Geochemistry pollution status and ecotoxicological risk assessment of heavy metals in the Pahang River sediment after the high magnitude of flood event

K. Y. Lim, N. A. Zakaria and K. Y. Foo

ABSTRACT

The present work is aimed at assessing the aftermath effects of the 2014 flood tragedy on the distribution, pollution status and ecological risks of the heavy metals deposited in the surface river sediment. A series of environmental pollution indexes, specifically the enrichment factor (*EF*), geo-accumulation index (I_{geo}), contamination factor (*CF*), modified degree of contamination (mC_d), pollution load index (*PLI*), potential ecological risk index (*PERI*) and sediment quality guidelines (*SQGs*) have been adopted. Results revealed that the freshly deposited sediments collected soon after the flood event were dominated by Cu, Fe, Pb, Ni, Zn, Cr and Cd, with the average concentrations of 38.74, 16,892, 17.71, 4.65, 29.22, 42.36 and 0.29 mg/kg, respectively. According to the heavy metal pollution indexes, Pahang River sediments were moderately to severely contaminated with Pb, Ni, Cu, Zn and Cr, while Cd with the highest E_f^i risk of 91.09 was the predominant element that illustrated an aesthetic ecological risk to the water body after the tragic flood event. The findings highlighted a critical deterioration of the heavy metals content, driven by the catastrophic flood event, which has drastically altered their geochemical cycles, sedimentary pollution status and biochemical balance of the river's environment.

Key words | environmental pollution index, flood, heavy metal, river, sediment

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HIGHLIGHTS

- The 2014 flood has been recognized as the most severe tragedy in the history of Malaysia for the past 30 years.
- Heavy metal pollution and sediment geochemical profile of Pahang River were analyzed.
- A series of geochemical pollution indexes, including enrichment factor (*EF*), geo-accumulation index (I_{geo}), contamination factor (*CF*), modified degree of contamination (mC_d), and pollution load index (*PLI*) were thoroughly evaluated.
- The synergistic ecotoxicological risks in relation to sediment quality were studied using comprehensive potential ecological risk index (*PERI*) and consensus-based sediments quality guidelines (*SQGs*).
- Establishment of a reliable reference on the sedimentology, geochemistry, pollution status and ecological risk assessments under different environmental settings for future protection, restoration or rehabilitation of the river system after the flood tragedy.

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INTRODUCTION

Riverine sediment, which naturally consists of loose sand, clay and soil particles, is a complex mixture of organic and inorganic components, notably silicates, carbonates, sulphide and minerals, deposited at the bottom of the water body by means of precipitation, ion exchange, adsorption, hydrolysis and chelation processes. Sediment is well-known to be the sensitive indicator of the environmental and geochemical contaminations to indicate the secondary sources of heavy metal pollution status in the river water. The mineralogical compositions, organic matter contents and textural characteristics of the sediment sequence could highlight the best natural archives of the recent environmental changes, as it serves as the major carrier and ultimate repository site for deleterious chemical contaminants in the aquatic ecosystem (Martínez-Salvador & Conesa-García 2019).

Heavy metals, members of an ill-defined subset of elements, are the main environmental pollutants that are likely to be associated closely with the colloid elements and fine-grained particulates, at the bottom sediment, governed by a complex dynamic set of physical and chemical interactions and equilibria. They originate from the ore formation and weathering of rocks, with toxic, indestructible and non-biodegradable properties, and demonstrate bio-accumulative potential and low self-purification capacity, with high ecological significance to the aquatic environment (Quinn & Dussaillant 2018). The intensity of heavy metal contamination and distribution at a watercourse may be largely affected by the flow velocity, massive overland flow and rate of erosion during the peak phase of rainfall drops, and natural features of the sediment matrix and the adsorbed compounds. Accordingly, the changing physical–chemical properties of the water sediment have been inferred to be the factor determinant on the heavy metal accumulative behaviour, holding over 90% of the metal content in the aquatic environment, and the presence of Pb, Hg, Zn, Cd and Cu fractions in the polluted sediment were at the magnitude of three times higher than the natural fluxes (Zhang *et al.* 2014).

These heavy metals are known to be accumulated in the benthic invertebrate and terrestrial organisms, either through bio-accumulation or bio-magnification in the food chain, and might be remobilized into the liquid phase during acute conditions. The interaction between heavy metals with different contaminants may result in additive and synergistic implications, with a huge threat to the environment and human body when the permissible concentration limits have been exceeded. Excessive and long-term exposure to the heavy metals could lead to the disruption of mental and neurological functions, and metabolic pathways, resulting in the impairment of energy and neurotransmitter production, and dramatic alterations of the blood and cardiovascular, gastrointestinal, nervous, reproductive and urinary systems overall (Dukes *et al.* 2020).

As the partitioning and distribution patterns of these heavy metals are of critical importance to the potential toxicity, mobility and route of contamination, a unique understanding of the metal speciation in the sedimentary environment, rather than the concentration, is essential for the hazard identification, and the residual implications to the food chain equilibrium. Although heavy metal pollution in the aquatic environment continues to be the aesthetic concern and is the focus of attention at the global level, the potential ecological risk after the devastating flood events has not been widely explored. In this sense, this study has been carried out with the key objectives to determine the metals distribution of the sediments along Pahang River after the 2014 historical flood event. A series of environmental assessment indexes, specifically the enrichment factor (*EF*), geo-accumulation index (I_{geo}), contamination factor (*CF*), modified degree of contamination (mC_d), pollution load index (*PLI*), potential ecological risk index (*PERI*) and consensus-based sediments quality guidelines (*SQGs*) for the detected metals were evaluated as reliable sedimentological datasets on the mobilization, origins of metal species and the potential availability of metals to biota within different environmental settings. This useful information could serve as a benchmark and useful tool for the establishment of an up-to-date status of the sediment contamination along Pahang River and highlight the probable adverse impacts on the natural ecosystem, for river pollution control, environmental management and ecosystem conservation in the near future.

STUDY METHODOLOGY

Environmental settling

Pahang River, located between the longitudes of E 101° 16′ 31″–E 103° 29′ 34″, and between the latitudes of N 2° 48′ 45″–N 3° 40′ 24″, is the longest river in Peninsular Malaysia, with a maximum length and breadth catchment of 459 and 236 km, respectively. This area has a humid tropical climate and is characterized by a bimodal pattern of southwest and northeast monsoons, with the annual rainfall ranging from 1,488 to 3,071 mm/year. It is one of the most important water resources for domestic water supply, fishing, agricultural and industrial activities and recreational purposes for the local community. The monsoon climate-controlled hydrology of the Pahang River plays an important role in its sediment erosion, transportation and deposition processes. The flow regime of Pahang River is characterized by low flows during the pre-monsoon season and extremely high flows during the flooding and post-monsoon seasons, with a higher river discharge of 845.78–1,008.50 m^3/s. The scenario worsened during the 2014 flood tragedy and had devastated 18,000 km^2 of the lowland areas lying along Pahang River. The swift river flow has transported vast quantities of pollutant loads, including eroded sediments, debris and tremendous amount of toxic heavy metals from the adjacent landmass into the river by inflowing water during the flood event (Lim *et al.* 2019).

Sediment sampling activity

The sediments were sampled at 26 stations (Figure 1) along the Pahang River that has been impacted by the nearby sources of contamination. The sampling period was

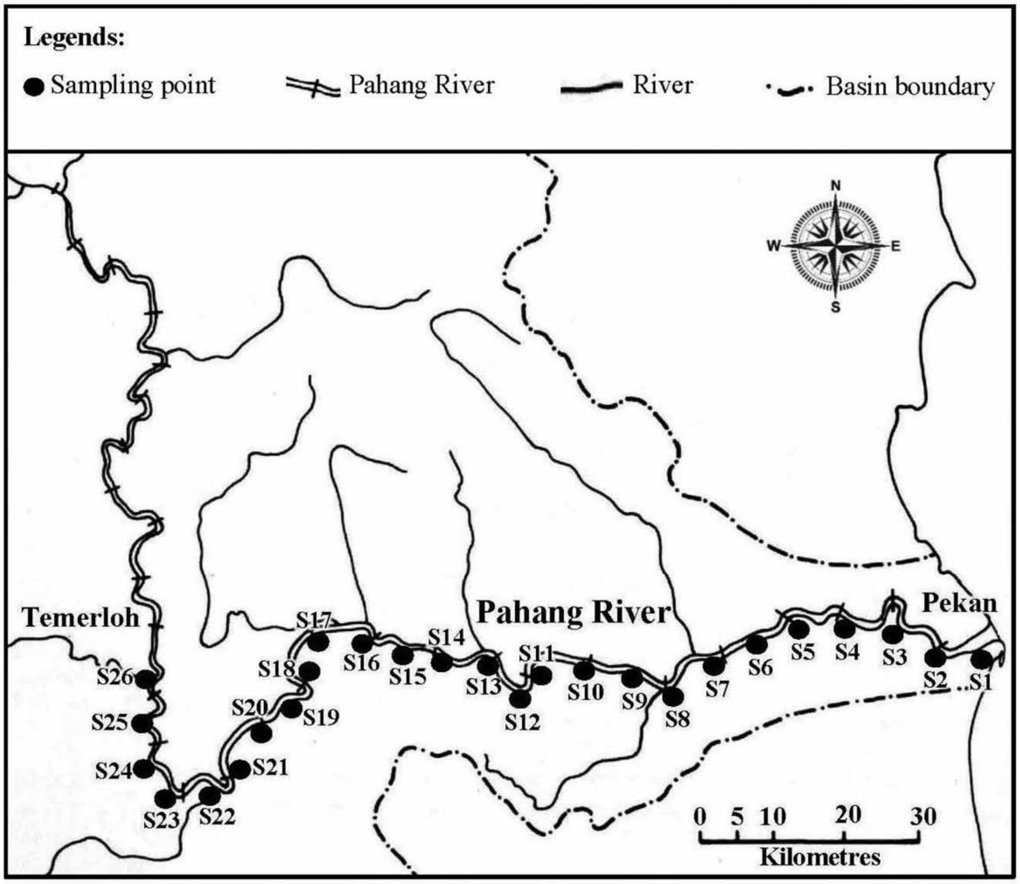

Figure 1 | The sampling stations along Pahang River.

coincident with the extreme flood event in the history of Malaysia, deploying pre-flooding and post-flooding data for July–October 2014, and January–March 2015, respectively. The superficial sediment samples were collected from each sampling site using a stainless steel Ponar-type box corer sampler. To avoid metal contamination from the grab's wall, the outer fraction of these sediment samples was removed, while the inner fraction was refrigerated at 4 °C for further analysis. The samples were oven-dried and grounded with clean mortar and pestle and sieved through a 63-μm stainless steel aperture for extraction purposes. The granulometric analysis was conducted according to the standard dry and wet sieving techniques for the specific quantification of clay (<4 μm), silt (4–63 μm) and sand (>63 μm) fractions. As the samples contain a high percentage of clay, the samples were subjected to pipette analysis as proposed by Krumbein & Pettijohn (1938), and each fraction of sucking pipette was dried and weighted to the nearest 0.0001 g.

Heavy metals analysis

The wet acid digestion, also known as the aqua regia+ hydrofluoric acid (HF) digestion method, was adopted. The laboratory apparatus and glassware were soaked in 10% of concentrated nitric acid, HNO_3 (v/v) overnight, rinsed with deionized water, and oven-dried before being used to minimize potential contamination as part of the quality assurance and quality controls (QA/QC). The heavy metals (Cu, Fe, Pb, Ni, Zn, Cr and Cd) were analysed according to the United States Environmental Protection Agency (USEPA) 3052 guideline, by digesting 50 mg of dried and homogenized sediment under the grain size of <63 μm using a microwave oven digestion system, with 1.5 mL of mixed concentrated acids (HF: HNO_3: HCl) in the ratio of 2: 3: 3. The qualitative and quantitative analyses were carried out using an Inductively Coupled Plasma-Mass Spectrometer (ICP-MS; model 7500 Perkin Elmer Ltd.) in triplicates, and the findings were presented in mg per kg (mg/kg) of dry mass of sediments. The differences ($p < 0.05$) in heavy metals content before and after the 2014 flood were analysed by one-way analysis of variance (ANOVA) using the Statistical Package for Social Sciences (SPSS) version 22.0.

Quality assurance and quality control

The quality assurance and quality control (QA/QC) for the strong acid digestion method, including reagent blanks, triplicate samples and standard reference materials (SRM1646a), were prepared and analysed using the same procedure and reagents to ascertain the precision of the analytical method using ICP-MS. The analytical results were reliable with the mean errors below 5%, and the analytical precision for the replicated samples was within ±10%.

Environmental pollution indexes

Enrichment factor

EF, a linear relationship between a reference metal with the metal of interest under natural sedimentation conditions, is an effective tool to measure the magnitude of heavy metal contamination in the sediment (Buat-Menard & Chesselet 1979). To minimize the mineralogical and grain size variation effects on the examined heavy metals, the concentrations were normalized to the textural characteristics of the Earth's crust by using iron (Fe) as the reference metal.

Geo-accumulation index

The I_{geo} approach, which was first proposed by Müller (1969), has been widely applied for the effective measurement of sediment quality and served as a reference to the heavy metal pollution, with regards to the pre-industrial levels of the bottom sediment and the background matrix correction factor of 1.5, which is specifically applied to counteract the variations of the background values, driven by the lithogenic effects.

Contamination factor and modified degree of contamination

The *CF* is an indicator of sediment contamination, commonly applied to evaluate the pollution of the environment by a given toxic substance over a period of time (Hákanson 1980). This environmental pollution assessment requires at least five surficial sediment samples to produce a mean pollutant concentration, to be compared with the baseline pristine reference level. Hákanson's study has proposed a sedimentological approach using a diagnostic tool, C_d, to indicate the accumulating status of heavy metals in the sediment for a particular core or sampling site, by defining the numeric sum of the n specific contamination factors (CF). With regard to an overall measurement of heavy metal contamination applicable to sediment, a modified and generalized model, the mC_d pollution impact equation, has been established to provide an integrated assessment of the overall enrichment and contamination impact for different groups of pollutants, by determining the sum of the CF for a given set of pollutants divided by the number of analysed pollutants.

Pollution load index

PLI is a standardized system that provides simple and comparative means to assess the overall toxicity status, and contribution of several heavy metals in the sediment samples, to allow the comparison of pollution levels between different locations and at different timeframes (Tomlinson *et al.* 1980). The average shales were adopted as the undisturbed background values for those metals in the same way as for the computation of *CF*.

Potential ecological risk index

PERI is a precise measurement, to ascertain the synergetic ecological risk of heavy metals according to the sedimentary theory (Hákanson 1980). This assessment takes into account the geological background conditions, environmental chemistry, biological toxicology and ecological variability. In the *PERI* concept, the standardized biological toxic-response factors (T_f^i) for the single pollutant in the surficial sediment are adopted from the Hákanson (1980) theory, where Cu, Pb, Ni, Zn, Cr and Cd are 5, 5, 5, 1, 2 and 30, respectively.

Sediment quality assessment guidelines

Numerical *SQG*s have been developed using a variety of approaches, typically involving statistical comparisons of chemical concentrations, and measurement of adverse biological effects during the exposure to sediment, with the primary purpose to protect the aquatic biota against the deleterious effects associated with sediment-bound heavy metals, to rank or to prioritize contaminated areas or chemicals of concern for further investigation (Long & MacDonald 1998). The mean quotient of these guidelines is a useful tool to summarize multiple datasets or figures into a single digit for a mixture of heavy metals associated with sediment. The sediment-associated *PEL*s of the specific heavy metals concentration are derived from the National Oceanic and Atmospheric Administration (NOAA) (MacDonald *et al.* 2000), with the Cu, Pb, Ni, Zn, Cr and Cd contents (mg/kg dry weight) of 197, 91, 36, 315, 90 and 3.5, respectively. A summary of the single and integrated sedimentary pollution indexes and their descriptions are listed in Table 1.

RESULTS

Sediment profile

Granulometric composition is a useful tool in the particle search and analysis techniques, commonly applied in the forensic evaluation of sediment, soil and a wide range of related particulate materials. This analysis is a very important indicator to the sediment properties, provenance, transport history and depositional conditions, where it provides data on the concentration of particles with different grain sizes in the observed sediment, including the percentage contents of the gravel, sand, pebbles, silt, mud and clay. Figure 2 demonstrates sand, silt and clay compositions in the sediment samples for both pre- and post-flooding conditions.

Field studies along with granulometric analysis of the collected sediment samples showed that the surface sediments of Pahang River were characterized by a high percentage of sand (6–100 wt%, mean = 58.9 wt%), and the spatial distribution was more uniform along the river during the ambient condition (Figure 2(a)). The sand content increased significantly towards the river mouth, and reached the higher range of 85–100 wt% at S1–S9 after flooding (Figure 2(b)) that were identified to be the most vulnerable zones of erosion, driven by the high flood wave energy, mixing up great amounts of sewage and heavy

Table 1 | Description of sediment quality indices for the assessment of heavy metal pollution in the aquatic sediment

Sedimentary pollution index	Mathematical equation	Description		Reference
Enrichment factor (*EF*)	$\mathrm{EF} = \dfrac{\left[\frac{C_x}{\mathrm{Fe}}\right]_{\mathrm{Sample}}}{\left[\frac{B_x}{\mathrm{Fe}}\right]_{\mathrm{Background}}}$	$[C_x/\mathrm{Fe}]_{\mathrm{Sample}}$	= The ratio of the concentration of the tested metals $[C_x]$ in relation to Fe content in the water sediment	Buat-Menard & Chesselet (1979)
		$[B_x/\mathrm{Fe}]_{\mathrm{Background}}$	= The ratio of the background values of the tested metals to Fe	
Geo-accumulation index (I_{geo})	$I_{\mathrm{geo}} = \log_2\left(\dfrac{C_x}{1.5 \times B_x}\right)$	C_x	= The concentration of metal '*x*' in the sediment sample	Müller (1969)
		B_x	= The background concentration of metal '*x*'	
		1.5	= The background matrix correction factor	
Contamination factor (*CF*)	$\mathrm{CF} = \dfrac{[C_x]_{\mathrm{Sample}}}{[B_x]_{\mathrm{Background}}}$	$[C_x]_{\mathrm{Sample}}$	= The mean concentration of a pollutant in the contaminated sediment	Hákanson (1980)
		$[B_x]_{\mathrm{Background}}$	= The pre-industrial 'baseline' sediments or average shale	
Overall degree of contamination (C_d)	$C_\mathrm{d} = \sum_{i=1}^{n} \mathrm{CF}^i$	n	= Number of analysed elements	Hákanson (1980)
		i	= The ith element (or pollutant)	
		CF	= Contamination factor	
Modified degree of contamination (mC_d)	$mC_\mathrm{d} = \dfrac{\sum_{i=1}^{n} \mathrm{CF}^i}{n}$			
Pollution load index (*PLI*)	$\mathrm{PLI} = (\mathrm{CF}_1 \times \mathrm{CF}_2 \times \mathrm{CF}_3 \times \mathrm{CF}_4 \times \mathrm{CF}_5 \times \mathrm{CF}_6 \times \mathrm{CF}_7)^{\frac{1}{n}}$	n	= Number of analysed elements	Tomlinson *et al.* (1980)
		CF	= Contamination factor	
Potential ecological risk index (*PERI*)	$PERI = \sum_{i=1}^{n} E_f^i = \sum_{i=1}^{n} C_f^i \times T_f^i$ $C_f^i = \frac{C_s^i}{C_n^i}$	E_f^i	= The potential ecological risk coefficient for a single pollutant i	Hákanson (1980)
		T_f^i	= The biological toxic-response factor for the single pollutant i in the surficial sediment	
		C_f^i	= The pollution coefficient of pollutant i	
		C_s^i	= The measured concentration of pollutant i in the surface sediment	
		C_n^i	= The pre-industrial background value for the pollutant i in the surface sediment	
Numerical sediment quality guidelines (*SQGs*)	$\mathrm{mean} - \mathrm{PEL} - \mathrm{quotient} = \sum\left(\dfrac{C_x}{\mathrm{PEL}_x}\right)/n$	C_x	= The concentration of metal '*x*' in the sediment samples	Long & MacDonald (1998)
		PEL_x	= The sediment quality guideline value of metal '*x*'	
		n	= Number of analysed elements	

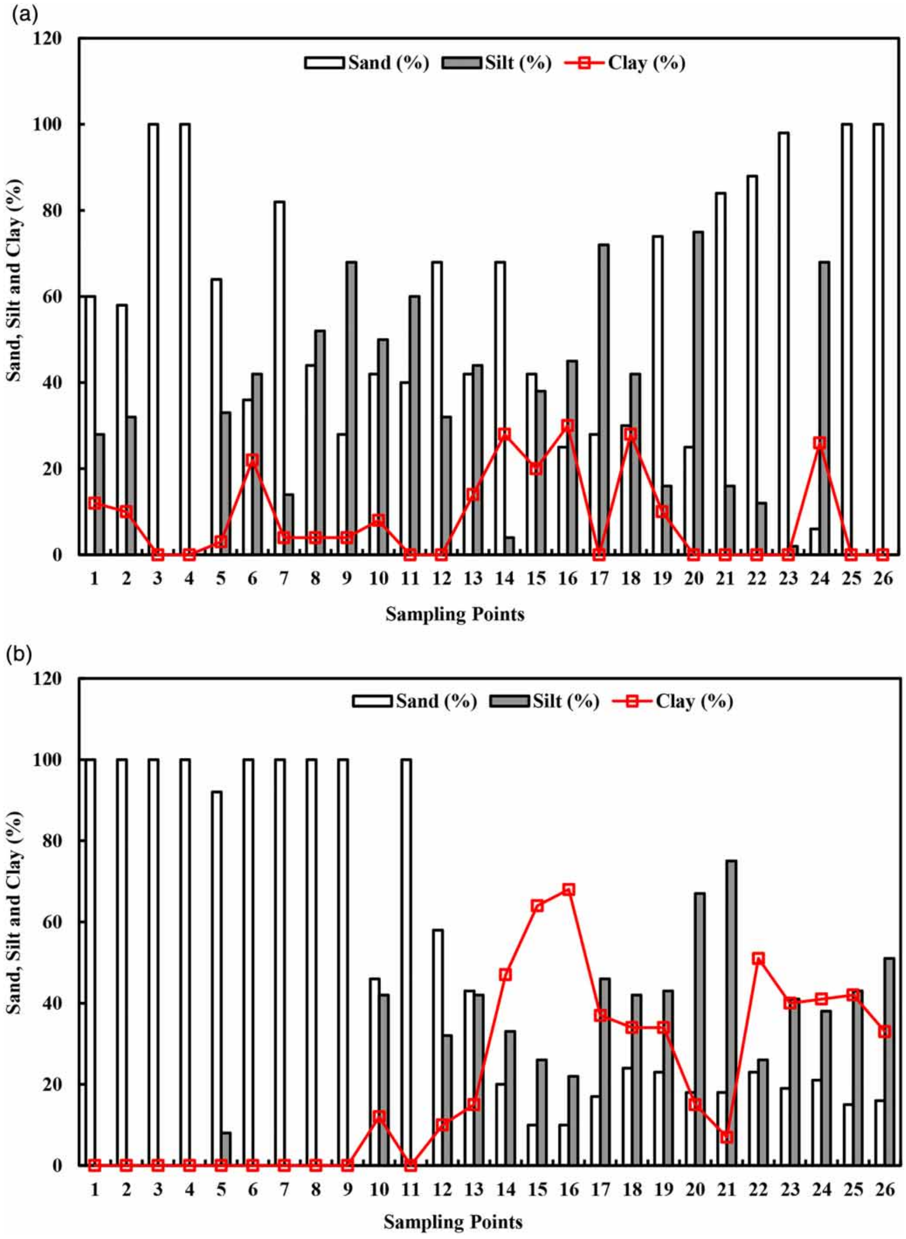

Figure 2 | Sediment profile of Pahang River before (a) and after (b) the 2014 tragic flood event.

land drainage, which allowed the settling or enrichment of fine particles to the bottom river sediment.

In the latter sector, the sediment samples at the middle part of the river exhibited the highest silt content (75 wt%) as compared to the mean silt content of 32.5 wt% for all sites. The silty nature could be due to the transportation and deposition of finer particles. However, the clay content showed an inverse spatial distribution pattern, with a lower range of 0–30 wt% (mean = 8.6 wt%) at the pre-flooding condition, and higher clay compositions of 35–68 wt%, with an average clay content of 21.2 wt% after the severe flood event. The general scarcity of clay-sized sediment in shallow water, due to winnowing through the action of water waves, tides and currents waves, were contrasted with the huge accumulation of argillaceous deposits in the rest of the river system. From the textural compositional analysis, sand was identified to be the major dominating component at both pre- and post-flooding seasons at the river ecosystem.

Hydrodynamic conditions, particularly flood incidence, are of primary importance in the hydrologic balance of the aquatic system, and the transport of freshwater, sediment, suspended matters and pollutants from the upstream catchments to the coastal zone in a short time period. In assessing the 2014 flood, the information on the river response to storms, in terms of physico-chemical behaviour of the sediment column, sediment yield and mobilization of particulate matters and pollutants, were investigated. According to the Department of Irrigation and Drainage Malaysia (DID 2014), the mean cumulative rainfall depth from 16 to 22 December 2014 recorded at the two gauging stations, particularly Pekan and Temerloh rainfall stations, was 738.5 mm. The rainfall pattern had resulted in higher river discharge and swift water flow along Pahang River, leading to the overland flow phenomena, water erosion and soil particle detachment at the upstream area and hilly terrain. These relevant variations may in turn affect the annual sediment budget, resulting in a noticeable increase in the loading of particulate-associated pollutants that could be harmful to the receiving coastal zone ecosystem (Lim *et al.* 2020).

Seasonal variation of metal load in sediment

The seasonal variation of heavy metals in the sediment samples before and after the flood tragedy is illustrated in Figure 3. Generally, the heavy metals deposited in the sediments were identified at the descending order of: Fe > Zn > Pb > Cr > Cu > Ni > Cd, with the concentration ranges of 9,922–19,559 mg/kg, 47.8–67.9 mg/kg, 17.2–35.6 mg/kg, 15.6–30.0 mg/kg, 11.6–30.0 mg/kg, 10.2–28.4 mg/kg, and 0.05–0.25 mg/kg, respectively. Specifically, the concentrations of Fe, Cu, Cr and Cd have increased by 17.68, 83.25, 77.24 and 61.11%, respectively, while the Zn, Pb and Ni contents were reduced by 49.99, 55.77 and 24.95%, respectively, under the flooded condition, and all the changes in the metal loads are significantly different at $p < 0.05$.

Among all, Fe was found to be the most abundant natural heavy metal in the water sediment, as tropical soil is rich in iron contents (Lim *et al.* 2013; Shaari *et al.* 2015). Similarly, copper ions in water could be easily bounded to the organic matter, and later deposited at the riverbed or along the riverbank (Manceau & Matynia 2010). Meanwhile, the prevailing water salinity has

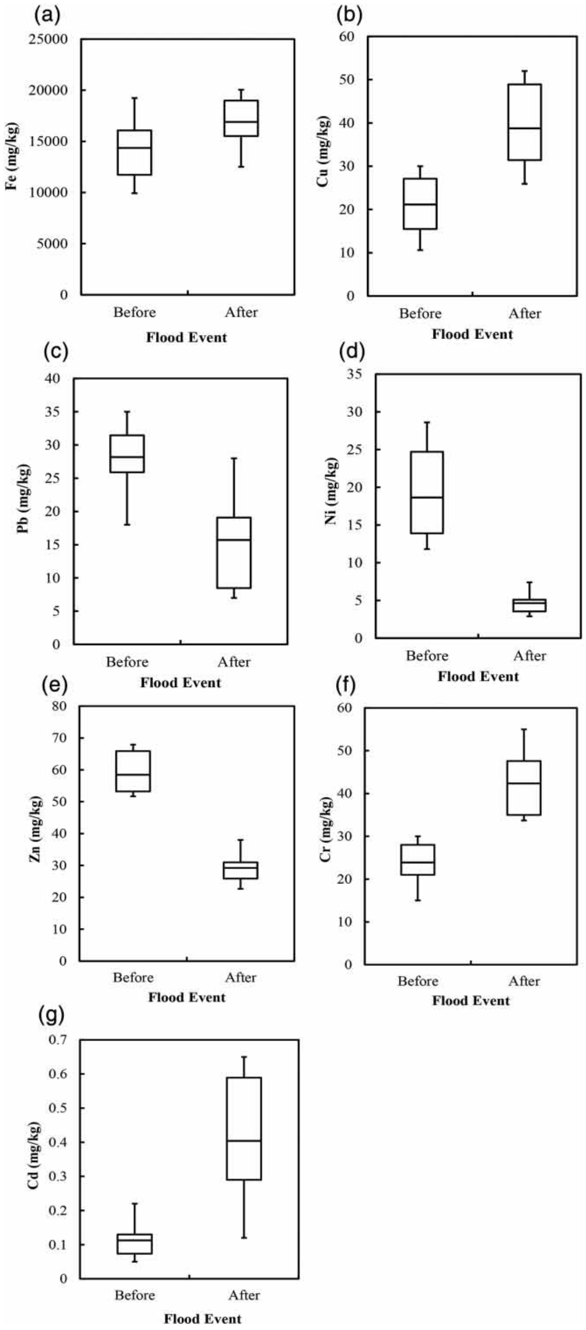

Figure 3 | Seasonal variation of heavy metals in the sediment samples at Pahang River before and after the catastrophic flood event.

demonstrated apparent influence on the distribution and accumulation of heavy metals in the sediment-water system by changing their insoluble form into the soluble form, to be trapped by the fine sediment (Wang *et al.* 2018).

The temporal patterns of Cr and Cd shared the aforementioned similarity after the monsoon flood incidence. Relatively, the presence of Cd is related to the industrial effluents emitted from the power plants and combustion related facilities, that may be flowing out to the adjoining water system. In contrast, the concentrations of Zn, Pb and Ni showed downward trends. Similar results have been outlined by Zhang *et al.* (2010), in which this metal group is strongly bounded to the exchangeable phase of the minerals, and probably more easily influenced by the dilution effect of heavy rainfall and strong floodwater current. A list of studies also recorded the parallel findings, in which the concentration of heavy metals in the water sediment was lower than the ambient condition (Forghani *et al.* 2009; Zhang *et al.* 2010; Karimian Torghabeh *et al.* 2020). The phenomenon could be attributed to the high disturbance by the massive overland flow during the flood event and mineral dissolution effect, which suggested that these metals originated mainly from the land-based runoff and river discharge at the study area.

Environmental pollution status

Enrichment factor

Measurement of the total concentration of heavy metals, as the only criterion for the assessment of heavy metal contamination in the sediment environment, is not satisfactory for tracing the natural or anthropogenic origins according to their grain sizes and mineralogy. The *EF* technique is a powerful tool to differentiate between the natural and anthropogenic sources of metal enrichment in sediment, and to discern the status of environmental contamination. Within this framework, the normalization of element contents is usually adopted, in which the metals concentration is normalized to the textural or compositional characteristics of the tested water sediment (Hornung *et al.* 1989).

In the present work, Fe was applied for the determination of *EF* in the sediment samples, for the following reasons: (i) the presence of Fe in the sediment is mainly due to the natural weathering processes, which could be applied to normalize the metals concentration, grain size and compositional influence of the metals; (ii) Fe is a better predictor than Al as a background heavy metal, due to the similarity between Fe with other metals in both toxic and anoxic conditions; and (iii) the natural sediment concentration of Fe tends to be uniform, mainly because of its good stability with fine solid surfaces as oxide coating in the sediment.

According to the analytical factor, the contamination levels are recognized into five major categories: $EF < 1$ demonstrates 'no enrichment', $1 < EF < 3$ denotes as 'minor enrichment', $3 < EF < 5$ refers to 'moderate enrichment', $5 < EF < 10$ indicates 'moderately severe enrichment', $10 < EF < 25$ represents 'severe enrichment', $25 < EF < 50$ signifies 'very severe enrichment', and $EF > 50$ implies 'extremely severe enrichment'. The $EF = 1.5$ is usually selected as the threshold value, with a value $EF < 1.5$ suggesting that these trace metals are originated entirely from crustal materials or natural weathering processes, whereas an $EF > 1.5$ indicates an appreciable portion of the trace metals is delivered as non-crustal. The alteration of *EF* for different deposited heavy metals in the water sediments at Pahang River before and after the 2014 severe flood event are described in Figure 4.

Under ambient conditions, the calculated *EF* for Pb, with an average value of 13.03, appeared to be the metal of concern at the study area. Meanwhile, Ni, with the

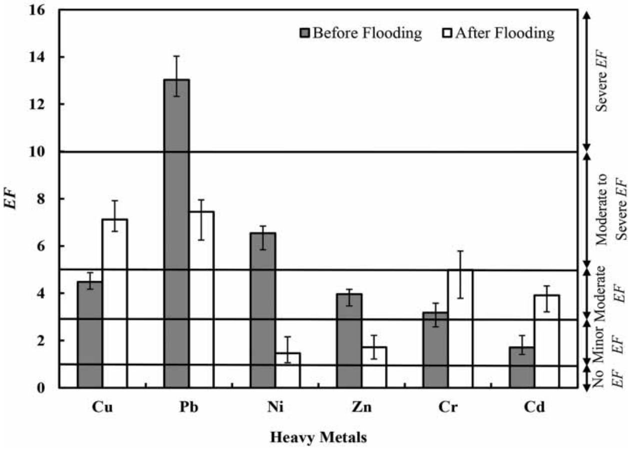

Figure 4 | The alteration of *EF* for different deposited heavy metals in the water sediments at Pahang River before and after the 2014 severe flood event.

EF value of 6.54, indicates a moderate to severe *EF*, that was one to three times higher than the Earth mean values, while Cr (3.17), Zn (3.96) and Cu (4.48) were identified as moderately enriched. In contrast, Cd, with the mean *EF* value of 1.70, was denoted as a minor *EF* along Pahang River. In agreement with the research findings as depicted in Figure 4, the *EF* values for Pb, Ni and Zn soon after the flood event are traumatically lower than the ambient conditions. The results could be related to the presence of autochthonous components and biogenic carbonates, which may significantly affect the binding capability of these metals within the water sediment. Nevertheless, the *EF* for Cu, Cr and Cd was significantly higher, with average values of 7.12, 4.99 and 3.91, respectively, after the flood tragedy.

The variability of the individual *EF* value may be a result of the nature of the heavy metals themselves, the pollution loads and their speciation forms of occurrence in the sediment–water complex. Technically, the greatest *EF* value for Pb, which could be associated with the surface runoff and municipal waste, recorded a higher distribution in the aqueous medium under huge water flow. Meanwhile, Cd that has been related to the colloidal particles deposited at the water sediment forms a stronger electrostatic attraction or Van der Vaals forces with the clay or silt-based substances. Technically, the bioavailability and toxicity of these heavy metals are deeply related to the concentrations or their chemical forms, with metals that have the highest and greater labile fraction in the sediment structure, and have a better mobility and bioavailability in the aquatic ecosystem. This hypothesis has been supported by Meng *et al.* (2008), who suggested that the finer grain size detected in the sediment samples appears to be the major factor influencing and affecting the enrichment status of heavy metals in the surficial sediment of the Wanggu River, Indonesia. Nevertheless, the *EF* method is criticized for its arbitrary 'cutoff value', with the assumption of the constancy of heavy metal/ reference element ratios in the natural process, and the available/reactive characteristics of heavy metals in nature (Dung *et al.* 2013).

Geo-accumulation index

The I_{geo}, originally introduced by Müller (1969), is applied to quantitatively evaluate the heavy metal contamination status in the sediment samples in comparison with the pre-industrial concentrations. The factor 1.5 is adopted for the correction of regional background difference, and according to the theory of I_{geo}, the contamination levels could be classified into seven major grades, given by: practically uncontaminated ($I_{geo} \leq 0$), uncontaminated to moderately contaminated ($0 < I_{geo} \leq 1$); moderately contaminated ($1 < I_{geo} \leq 2$); moderately to heavily contaminated ($2 < I_{geo} \leq 3$); heavily contaminated ($3 < I_{geo} \leq 4$); heavily to extremely contaminated ($4 < I_{geo} \leq 5$); or extremely contaminated ($I_{geo} > 5$) (Müller 1969).

The I_{geo} values of the individual metal in the surficial sediments of Pahang River before and after the 2014 flood event are illustrated in Figure 5. The major weakness of this index is that it does not take into account the geochemical variability of different pollutants, and the defined factor is not readily comparable with other enrichment indices, due to the nature of the I_{geo} calculation, which involves a log function, and the background multiplication of 1.5. In agreement with the previous findings recorded by Abrahim & Parker (2008), the presented I_{geo} indexes signified that the riverbed sediments were heavily contaminated by Pb (Class IV), moderately to heavily contaminated by Ni (Class III), and moderately polluted by Cu, Zn and Cr (Class II) at most of the sampling sites. The negative I_{geo} values (Class I) of Fe, Ni and Zn indicated negligible contamination levels along the course of the river stretch, due to the bioturbation in the upper mixed layers or dilution by course sediment. The depositions of Cu, Cr and Cd were significantly increased,

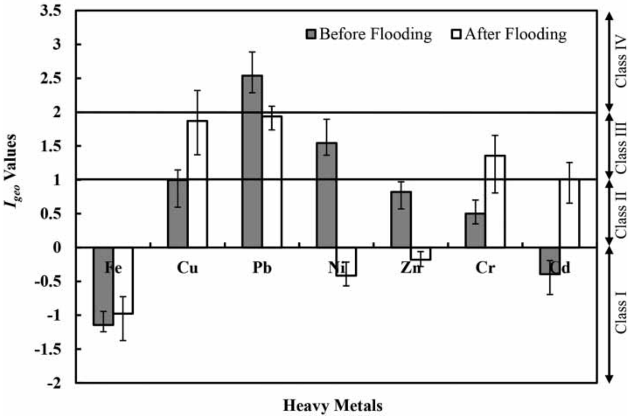

Figure 5 | The I_{geo} values of the individual metal in the surficial sediments of Pahang River before and after the 2014 flood event.

which could be flushed from the upper region to the downstream by the massive overland flow, as evident from the rising I_{geo} values of 1.87, 1.36 and 1.01, respectively.

Parallel to the research findings, Feng *et al.* (2011) and Oliveira *et al.* (2011) have suggested that the changing hydrodynamic conditions were sufficiently strong to facilitate the dilution or diffusion of heavy metals along the water system. From the pollution index assessment, it was clearly revealed that the sediment quality along the sampling stations has undergone dramatic amplification in terms of heavy metals content. These results indicated a significant relationship between the distribution of heavy metals with the grain size of the sediment samples. An evaluation of the sedimentological characteristics of Pahang River has been carried out by Kamaruzzaman *et al.* (2010). The mean particle size of the water sediment ranged from 1,200 μm (−0.24 Ø) to 72 μm (3.79 Ø), with an average diameter of 46.5 μm (4.51 ± 3.13 Ø). Comparatively, the fine-grained bottom sediment which has an average particle size <63 μm as obtained in this work showed a greater accumulation of heavy metals than the coarser particles after the flood event.

Contamination factor and modified degree of contamination

For a better estimation of the heavy metals status in the surface sediment, the *CF*, C_d and mC_d have been chosen to be the powerful tools for the accurate assessment of the contamination level of different heavy metals to the surrounding environment over a period of time. Originally developed by Hákanson (1980), *CF* was applied for the monitoring of environmental pollution via a given toxic substance, in which C_d and mC_d could quantify the overall degree of contamination, with the specific aim of minimizing the grain size effects between the measured content and the background value, or to reveal the actual geochemical imbalance. The classification and description of the *CF* and mC_d for each metal with respect to the background value are categorized in Table 2 as: $CF < 1$ indicates a low degree of sediment contamination; $1 \leq CF < 3$ refers to a moderate degree of sediment contamination; $3 \leq CF < 6$ illustrates a considerable degree of sediment contamination; and $CF \geq 6$ represents a very high degree of sediment contamination, whereas the contamination level of mC_d was classified, ranging from: $mC_d < 1.5$ = nil to a very low degree of contamination; $1.5 \leq mC_d < 2$ = a low degree of contamination; $2 \leq mC_d < 4$ = a moderate degree of contamination; $4 \leq mC_d < 8$ = a high degree of contamination; $8 \leq mC_d < 16$ = a very high degree of contamination; $16 \leq mC_d < 32$ = an extremely high degree of contamination; and $mC_d \leq 32$ = an ultra-high degree of contamination.

Table 2 | The gradations of *CF* and mC_d for each metal with respect to the background value

Pollution index	Index	Contamination intensity of sediment quality
Contamination factor (*CF*)	$CF < 1$	Low degree of sediment contamination
	$1 \leq CF < 3$	Moderate degree of sediment contamination
	$3 \leq CF < 6$	Considerable degree of sediment contamination
	$CF \geq 6$	Very high degree of sediment contamination
Modified degree of contamination (mC_d)	$mC_d < 1.5$	Nil to very low degree of contamination
	$1.5 \leq mC_d < 2$	Low degree of contamination
	$2 \leq mC_d < 4$	Moderate degree of contamination
	$4 \leq mC_d < 8$	High degree of contamination
	$8 \leq mC_d < 16$	Very high degree of contamination
	$16 \leq mC_d < 32$	Extremely high degree of contamination
	$mC_d \leq 32$	Ultra-high degree of contamination

The variation of *CF* for different heavy metals detected in the deposited sediments along Pahang River is illustrated in Figure 6. From the presented results, the calculated *CF* for Pb ($CF = 5.74$), which has been defined as the metal of concern, has shown a great reduction after the flood event. Meanwhile, the *CF* values for Zn, Cr, Cd and Ni, which lay between 1.10 and 3.84, did not show great fluctuation, falling under the category of considerable or moderate degree of contamination. However, the *CF* values for Cu and Ni were moderately altered by the changing hydrological conditions, while

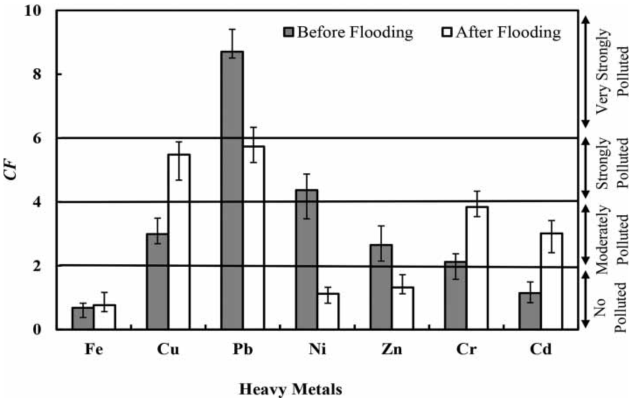

Figure 6 | The variation of *CF* for different heavy metals detected in the deposited sediments along Pahang River.

Fe was identified to show the lowest to nil contamination level. The overall degree of contamination (C_d) values were detected at 19.84 and 21.28, respectively, while the computed mC_d values were computed as 2.83 and 3.04, before and after the flood event, respectively. Comparison between the *CF* and m*C* values demonstrated the changing contamination degree of different metals driven by the flood events, which has contributed to the alteration in metal distribution, and overall metal contamination along the Pahang River.

Pollution load index

Different from the other indexes, *PLI* derived by Tomlinson *et al.* (1980) permits a comparison of the overall heavy metal pollution for a specific area and region. The *PLI* could be generally classified into five major categories: no pollution ($PLI < 1$); moderate pollution ($1 < PLI < 2$); heavy pollution ($2 < PLI < 3$); and extremely heavy pollution ($3 < PLI$). In this work, the *PLI* values were identified to be 2.2 and 2.5, respectively, before and after the extreme flood event. The slight fluctuation implied a different degree of deposition and distribution of heavy metals at the water sediment, which has normalized the overall contamination behaviour. The acquired *PLI* values indicated a moderate degree of pollution to the river system. This integrated index is coded by the advantage of being easily understood by the researchers but is criticized by the weakness of the ecotoxic potential of individual metal that has not been taken into account.

Potential ecological risk index

The potential ecological hazard index was originally proposed by Hákanson (1980), to integrate the concentration of heavy metals with ecological effect, types of pollutant, environmental impact, toxicology and sensitivity of a water body for the effective evaluation of the potential risk of heavy metals in the surface sediment to the aquatic ecosystem and human health. The methodology assumed that the sensitivity of the aquatic system depends primarily on its productivity. This potential ecological risk assessment system is based on the element abundance and several preconditions as follows: (1) concentration condition, which *PERI* would increase with the aggravated metal pollution degree in sediment; (2) species number condition, the metals in sediment possess additivity, namely potential ecological risk is larger with multiple metals; (3) toxic-response condition, the heavy biological-toxicity metals show larger evidence of *PERI* and magnitude for abundance correction; and (4) sensitivity condition according to biological production index, namely the sensitivity is different under different water quality systems. The terminology and grading standards of potential ecological risk assessment in relation to sediment quality as suggested by Hákanson (1980) are given in Table 3. The value of $E_f^i < 40$ indicates a low potential ecological risk; $40 \le E_f^i < 80$ is a moderate ecological risk; $80 \le E_f^i < 160$ is a considerable ecological risk; $160 \le E_f^i < 320$ is a high ecological risk; and $E_f^i \ge 320$ is a very high ecological risk, while $PERI < 150$ indicates a low potential ecological risk; $150 \le PERI < 300$ is a moderate ecological risk; $300 \le PERI < 600$ is a considerable

Table 3 | The grading standard of the potential ecological risk assessment (E_f^i and *PERI*) in relation to the sediment quality

Range of potential ecological risk index, E_f^i	Grade of ecological risk of single metal	Range of potential toxicity index (*PERI*)	Grade of potential ecological risk of environment
$E_f^i < 40$	Low risk	$PERI < 150$	Low grade
$40 \le E_f^i < 80$	Moderate risk	$150 \le PERI < 300$	Moderate
$80 \le E_f^i < 160$	Considerable risk	$300 \le PERI < 600$	Severe
$160 \le E_f^i < 320$	High risk	$600 \le PERI$	Serious
$320 \le E_f^i$	Very high risk		

ecological risk; and $PERI \geq 600$ is a very high ecological risk.

The calculated potential ecological risk indices (E_f^i) for the seven heavy metals are presented in Figure 7. From Figure 7, the mean E_f^i values for each heavy metal before and after the flood event are according to the descending order of: $\text{Pb} > \text{Cd} > \text{Ni} > \text{Cu} > \text{Cr} > \text{Zn}$, and $\text{Cd} > \text{Cu} > \text{Pb} > \text{Cr} > \text{Ni} > \text{Zn}$, respectively. At the ambient conditions, the mean E_f^i values of Zn, Cu, Ni, Cr and Cd ranged between 2.62 and 25.42, to indicate a low degree of ecological risk. However, the average E_f^i value of Pb in the sediment samples, that reached the peak value of 46.95, was likely to show moderate risk compared to other metals, as it is manifested with a relatively broad dispersion, which may be attributed to the anthropogenic intrusions. On the whole, the mean *PERI* value before the flood event, which was found at 116.03 ($PERI < 150$), indicated a low ecological risk of the aquatic biota along Pahang River.

In contrast, the *PERI* acquired after the flood event was 158.96. The ecological risk factors for Pb, Zn, Cu, Ni and Cr that fall under the low ecological risk ($E_f^i < 40$) were identified to be 26.18, 1.31, 27.41, 5.45 and 7.52, respectively. Nevertheless, Cd, which has recorded the highest average E_f^i value of 91.09, demonstrated a high degree of ecological risk to the local natural environment. The high content of Cd in the surface sediment samples observed in this study could be attributed to the changing geological factors and sewage discharges. The obtained result was differed from the single factor pollution index, mainly due to the justification that some elements which showed a higher contamination level might illustrate great affinity to the small particles and could be easily transferred into water sediment by the suspended solids, to lower its toxicity to the aquatic ecosystem, and potential ecological risk in the coastal environment (Skarbøvik *et al.* 2014). The ecological distress driven by the contaminated water sources in the flood events has induced a dramatic financial loss exceeding US$10,000, with more than 5,000 patin and tilapia, as well as 300 kerai fish being killed at the Temerloh area along the river (The Star 2018).

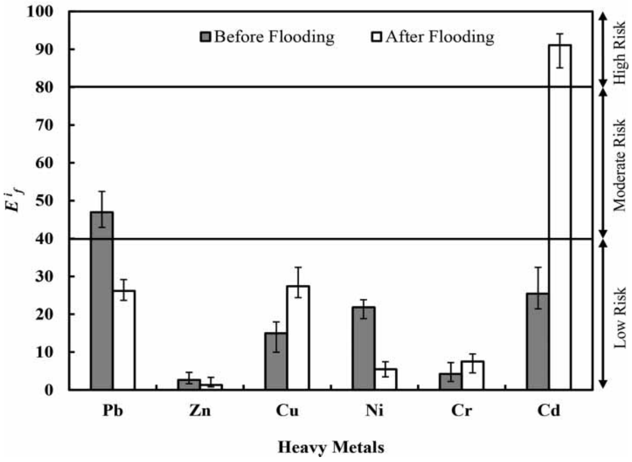

Figure 7 | E_f^i for the seven heavy metals detected in the sediments along Pahang River before and soon after the flood event.

Sediment quality assessment guidelines

Hazardous waste site evaluation involves the collection of substantial quantities of sediment chemistry data, and these data would serve as the backbone to support the screening level of ecological risk assessments (USEPA 2005). To effectively predict the heavy metal pollution, a comparative study was conducted according to the *SQG*s as proposed by the USEPA standards; and the analytical results were compared with *SQG*s, notably probable effect limit (*PEL*) to ascertain the true extent of sediment contamination, and to predict the potential biological effects to the natural environment (MacDonald *et al.* 2000). Generally, it is a powerful tool to be applied in numerous applications, including monitoring plans design, historical data interpretation, remedial investigations and developing sediment quality remediation objectives.

Accordingly, the mean-*PEL*-quotient of <0.1 has a 9% probability of toxicity; a mean-*PEL*-quotient of 0.11–0.5 has a 21% probability of toxicity; a mean-*PEL*-quotient of 0.51–1.5 has a 49% probability of toxicity; and a mean-*PEL*-quotient of >1.5 has a 76% probability of toxicity (Long & MacDonald 1998). In the present study, the mean-*PEL*-quotient for all the tested sediment samples after the flood event varied from 0.08 to 0.47, signifying 21% probability of toxicity on the biota, and the mean-*PEL*-quotient for Ni, Pb, Cu and Cr, with the quotient values 0.13, 0.17, 0.19 and 0.47, respectively, have exceeded the individual-based *PEL* values recommended by the USEPA. The obtained results verified the ecological risk driven by the distribution of heavy metals, that might have further implications for the natural environment.

DISCUSSION

Environmental pollution and ecological risk assessments are the two quantitative approaches for the reliable evaluation of accumulative hazards on the environment and ecological receptors, driven by single or a combination of different external stressors and affecting agents (USEPA 1998). The basic framework of the ecological risk assessment involves three major components, specifically problem formulation, analysis and risk characterization. The uncertainties involved in each individual step, and their cumulative results, could be integrated into the overall risk assessment. The selection of the assessment paradigm and/or their combinations will be conducted not only according to the changing conditions, plans, programs, policies, scopes and objectives of the projects, but may be limited by the available data and accessible information. These comprehensive approaches, particularly the framework of ecological risk and comparative risk assessments, were promulgated, and could be applied in environmental management, specifically for the identification of unique features in ranking contaminated sites; and while detailed information is available, the new sustainability indicators, derived mainly from the paradigm of sustainability assessment, would generate a new criterion to prioritize the contaminated sites.

Among all, heavy metals are of considerable environmental concern, primarily due to their tetra-toxicity, persistent and bio-accumulative behaviours. These heavy metals would undergo numerous changes in the natural media during their transport, assisted by the dissolution, precipitation and sorption phenomena. In this sense, the identification and quantification of the heavy metal sources, mobility, toxicity, bioavailability, as well as the fate of the individual metal, remains a critical challenge to be highlighted. The additivity, antagonism or synergism of multiple heavy metal contaminants may demonstrate the overall toxicity rather than an individual implication. In the present work, the geochemical studies with respect to the *EF*, I_{geo} and *CF* have been analysed to ascertain the magnitude of single metal pollution, and to estimate the impact of anthropogenic activities at the polluted sites; while the mC_d and *PLI* were applied for the evaluation of the overall risk of multiple heavy metals in the river sediment. These geochemical approaches, however, involve a different level of subjectivity, and the possible weighting role, combined antagonism as well as the ecological risk on the river sediment have been neglected (Ahamad *et al.* 2020).

The complementary approaches of *PERI* and *SQG*s, which integrates sediment standard criteria, grain size effect, toxic-response factor, *EF*, I_{geo} and *CF* indexes are therefore necessary for an accurate assessment of heavy metals accumulation from various anthropogenic sources, and evaluation of the resultant environmental and ecological risks in the sedimentary environment, with possible adverse effects to the aquatic organisms, for the interpretation of sediment quality (Kulbat & Sokołowska 2019).

In the present study, the analysis with respect to *EF*, I_{geo} and *CF* illustrated that the river sediments were severely accumulated by Pb and Cu, and moderately contaminated by Cr, Cd, Zn and Ni, with the accumulative pollution risks, mC_d and *PLI* of greater than 1. The acquired results of both single- and multi-element sediment quality indices are important to better delineate the co-contamination and potential interactions of different anthropogenic heavy metal components along Pahang River. Meanwhile, the ecological quality balance of the river sediment, either defined as a qualitative threshold or focus on ecological risk assessment of a single metal, recorded a *PERI* of 158.96, with the highest E_f^i value of 91.09 for Cd, and mean-*PEL*-quotient of 0.08–0.47 after the heavy flood event, were made to provide excellent and comprehensive geological information, to allow a more realistic estimation of the real synergistic ecological risk in the sediment. These aforementioned findings provided a concluding remark that the integrated ecotoxicological index prevails to be more expressive and representative for the health risk estimation of heavy metal contamination in the aquatic river sediment.

A comparative summary of the single and integrated geochemical and ecotoxicological risk indexes for different heavy metals after the flood incidence at different regions is depicted in Table 4. Generally, this present work provides up-to-date and comprehensive information on the aftermath effects of heavy metal pollution status and environmental degradation of the river's ecosystem. From the results

Table 4 | A comparative summary of the single and integrated geochemical and ecotoxicological risk indexes for different heavy metals after the flood incidence at different regions for the past 10 years

Heavy metal **Single index**	**Pb**	**Cu**	**Cr**	**Cd**	**Zn**	**Ni**	**Fe**	**Flood event**	**Reference**
EF	7.45	7.12	4.99	3.91	1.72	1.46	0.99	Pahang River, Malaysia	This study
	4.00	4.90	1.70	4.80	4.60	1.60	–	Seine River, France	Le Gall *et al.* (2018)
	1.10	6.80	7.60	–	10.1	3.5	0.29	Deba River, Spain	Martínez-Santos *et al.* (2015)
	3.20	2.70	6.60	–	2.20	2.40	–	Hanjiang River, China	Guo *et al.* (2014)
I_{geo}	1.93	1.87	1.36	1.01	−0.18	−0.42	−0.98	Pahang River, Malaysia	This study
	2.74	3.96	–	–	3.90	3.09	–	Lososinka River, Russia	Slukovskii (2015)
	−0.7	0.10	1.20	–	−1.10	−0.60	–	Hanjiang River, China	Guo *et al.* (2014)
CF	5.74	5.48	3.84	3.01	1.32	1.12	0.76	Pahang River, Malaysia	This study
	0.93	1.46	1.02	2.72	1.43	–	–	Yanghe River, China	Kuang *et al.* (2016)
E_f^i	26.18	27.41	7.52	91.09	1.31	5.45	–	Pahang River, Malaysia	This study
	9.03	352.94	1.94	–	8.88	6.59	–	Pearl River, China	Liu *et al.* (2020)
	3.07	4.78	3.04	14.72	1.52	10.38	–	West Morava River, Serbia	Antić-Mladenović *et al.* (2019)
SQGs	$>PEL$	$>PEL$	$>PEL$	$<PEL$	$<PEL$	$>PEL$	$<PEL$	Pahang River, Malaysia	This study
	$<TEL$	$>TEL$	–	$<TEL$	$<TEL$	$<TEL$	–	Ganges River, India	Bonnail *et al.* (2019)
Integrated index									
PLI	2.5							Pahang River, Malaysia	This study
	1.08							Yanghe River, China	Kuang *et al.* (2016)
PERI	158.96							Pahang River, Malaysia	This study
	672.664							Pearl River, China	Liu *et al.* (2020)
	49.57							West Morava River, Serbia	Antić-Mladenović *et al.* (2019)
	30,711.8							Longjiang River, China	Zhao *et al.* (2018)

listed in Table 4, the *EF*, I_{geo}, *CF*, *PLI*, E_f^i and *PERI* values reported in the literature findings from different flood events as compared to 0.29–10.10, −1.10–3.96, 0.76–5.74, 1.08–2.50, 1.31–352.94 and 49.57–30711.8, are documented in this study. According to the sediment contamination criteria, the rising of *EF*, I_{geo}, *CF* and *PLI* values after the flood events in the environmental assessment represents a progressive deterioration of the sediment quality. Similar to the sedimentology approach, *PERI* and the potential ecotoxicity index, greater than the cut-off values, could be concatenated to the heavy metal pollutants with ecological, environmental and biological toxicology effects. Specifically, Fe has been adopted as the most suitable geochemical normalizing element to compensate for the granulometric and mineralogical variability of metal concentrations in sediments, as its concentration and distribution was uniform and not related to other examined heavy metals along Pahang River, indicating a good stability of this metal in the river system as a whole. Guo *et al.* (2014) and Martínez-Santos *et al.* (2015) have applied a similar approach in the normalization of geochemical calculations along Hanjiang River, China and Deca River, Spain after the tragic flood events, whereas thorium (Th) was introduced as a conservative tracer or normalizer for the quantification of heavy metal enrichment in the flood sediments at Seine River, France (Le Gall *et al.* 2018).

The variations of *EF*, I_{geo}, *CF* and E_f^i of the examined heavy metals summarized in Table 4 have demonstrated different degrees of pollution and toxicity levels for Cu, Fe, Pb, Ni, Zn, Cr and Cd. Overall, the sediments in the studied regions could be rated as 'extreme ecological risk degree' for Pearl and Longjiang Rivers, China, and 'low ecological hazard' for West Morava River, Serbia, which are mainly attributed to the local exogenous influence at different countries and geographical alterations, particularly the biogeochemical processes, heavy metals distribution pattern, nature of the sediment matrix and the properties of the parental compounds. The complexities of human activities, hydrological alterations and climatic change, coupled with the complex interaction phenomena, notably from the minerals release during the dry season, and their influx in the monsoons from the watershed or floodwater discharge, have eventually introduced a variety of implications with different degrees of pollution status and assessment results.

CONCLUSION

In this work, the complex relationships and contamination status between the sediment texture and heavy metal concentrations of the surficial sediment samples along Pahang River before and after the heavy flood event were analysed with respect to the distribution characteristics, geochemistry pollution features and ecotoxicological risks, by adopting the chemometric analysis, sediment quality criteria, regional sedimentary reference data and environmental hazard assessment indexes. Seven major heavy metals, specifically Fe, Cu, Pb, Ni, Zn, Cr and Cd, were detected in the deposited sediments, with the highest concentrations of 20,451, 54.20, 28.60, 7.40, 38.00, 55.30 and 0.65 mg/kg, respectively, which was 4–5-fold higher than the corresponding local natural background levels. The environmental assessment indexes revealed a polymetallic contamination of the river sediments, with: (1) the magnitude of single-element indexes, notably EF, I_{geo} and CF illustrated that the river sediments were severely accumulated by Pb and Cu, but moderately contaminated by Cr, Cd, Zn and Ni; (2) the combination of mC_d and PLI values was greater than 1; (3) while the $PERI$ value acquired after the flood event was observed at 158.96, with Cd hitting the highest single ecological risk factor, E_f^i of 91.09, indicating the possible long-term adverse biological implications for the ecosystem. The newly organized baseline data could prevail to be a valuable reference for the reliable assessment of heavy metal pollution status under different environmental settings, which are necessary for the aquatic system protection, and future restoration or rehabilitation of the river basin after the flood tragedy.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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