RESEARCH ARTICLE

Mobility and risk assessment of heavy metals in benthic sediments using contamination factors, positive matrix factorisation (PMF) receptor model, and human health risk assessment

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Abstract

Metal pollution in benthic sediments was fractionated and modelled to quantify the risk of anthropogenic activities on river ecosystems. In this study, the individual contamination factor (ICF) and the global contamination factor (GCF) were used to measure the contamination levels in the sediments. On the other hand, the mobility factor (MF) was used to quantify the mobility of heavy metals in benthic river sediments. The factors used to assess pollution in benthic sediments employ bioavailable fractions of heavy metals, which have a greater chance of release into aquatic sediments and hence are more dangerous to the environment. Heavy metal mobility (MF) is highest in the post-monsoon season for Zn, Pb, Cu, and Co; Fe in winter; Mn in pre-monsoon; and Cd in monsoon. This means that heavy metals accumulate in benthic sediments during the post-monsoon season when river flows are less turbulent. ICF and GCF data show that pollution levels are higher post-monsoon than the rest season levels. Sediment samples were further subjected to the positive matrix factorization (PMF) model, which identified four factors that explained the variation in the study: factor 1 is concerned with anthropogenic Cu, Cd, and Co pollution, while factors 2, 3, and 4 are concerned with Fe, Mn, and Zn pollution. Finally, the total cancer risk (TCR) and hazard index (HI) are employed to quantify the risk to human health from accidental ingestion and dermal exposure. According to the risk outcomes from probabilistic and deterministic approaches, river exposure is dangerous to human health, with dermal absorption being the most significant concern of the exposure paths.

Keywords Contamination factors · Health risk assessment · Heavy metals · Mobility factor · Positive matrix factorization

Introduction

In recent years, the pollution of benthic sediments due to various anthropogenic activities has emerged as a serious issue. As a result of excessive effluent discharge (mostly from domestic, agricultural, and industrial wastewater), many heavy metals have accumulated in the aquatic ecosystem, posing a severe threat to global sediment flux (Armagan et al. 2008; Azhar et al. 2015; Yao et al. 2021). These heavy metals are not only persistent, but they are also harmful when their concentrations exceed permissible limits since they do not degrade or decompose over time. In addition, these compounds are less mobile in water than other chemicals. As a result, they accumulate in natural aquatic systems, leading them to adhere to the top of the sediment column of the water body. As a result, sediment columns of water bodies are potential sources of heavy metals, which can be released into the water column or aquatic flora and fauna by natural or anthropogenic processes, and so participate in the food chain (Yin et al. 2011; Yao et al. 2021).

In contrast to typical pollutants like nitrogen and phosphorus, toxic metals are dangerous at extremely low quantities, such as a few micrograms per millilitre (µg/mL). As a result, residents of polluted areas, children and adults, are exposed to hazardous compounds via various sources and pathways, resulting in a wide range of toxic exposures for them (Wang et al. 2017b, a; Chen et al. 2019; Xu et al. 2020). Chromium (Cr) and lead (Pb) are two heavy metals that have been identified as human carcinogens (Lyon 1994).

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On the other hand, the systemic non-cancerous metals (e.g. Cd, Co, Cu and Hg) can also cause negative health consequences even at minute concentrations (Djadé et al. 2021). Heavy metals can be found in sediments in several chemical fractions, each related to their mobility and bioavailability (Liang et al. 2017; Wang et al. 2019). Because total metals do not fully reflect geochemical processes, chemical speciation and metal bioavailability, sequential extraction is useful for investigating these topics (Sundaray et al. 2011; Zhang et al. 2017; Hing et al. 2020). It is worth noting that exchangeable metals are volatile and easily absorbed by humans and aquatic species (Liang et al. 2017). Heavy metals generated from anthropogenic activities are commonly found in an unstable state with greater mobility (Zhang et al. 2017). On the other hand, those lithogenic in origin are frequently associated with sediments that have taken on stable forms (Xiao et al. 2015). As a result, determining the heavy metal chemical fractions is crucial for source identification and risk assessment, particularly in areas with high levels of human activity.

Several modelling and prediction tools have been developed in the context of heavy metal concentrations in sediment and their influence on aquatic ecosystems (Chen and Chau 2019; Ardabili et al. 2020). However, a detailed qualitative and quantitative investigation must be carried out to identify potential pollution sources contributing to soil stratum contamination. Various methods have been used to identify pollution sources over time, including the absolute principal component scores-multivariate linear regression (APCS-MLR), chemical mass balance (CMB), maximum likelihood principal component analysis (MLPCA) and positive matrix factorization (PMF) (Jiang et al. 2017; Lv 2019). The PMF technique is well known and widely recognized as a powerful tool for source allocation, particularly for air pollutants, and the United States Environmental Protection Agency (USEPA) endorses it (Hsu et al. 2017; Manousakas et al. 2017). Other researchers have used the same statistical method to analyse water quality data (Gholizadeh et al. 2016; Zhang et al. 2020). In order to deal with environmental data and handle the uncertainty and distributions inherent in those data, positive matrix factorization (PMF), a multivariate analysis, has been developed (Paatero and Tapper 1994). When compared to other multivariate methods, PMF can provide a more accurate representation of the system by

(1) accounting for analytical uncertainties commonly associated with measurements of environmental samples and (2) requiring all values in the solution profiles and contributions to be positive (Reff et al. 2007).

A qualitative and systematic approach was used to identify the current literature on pollution in sediments due to heavy metals in India. The procedures below were performed to retrieve the needed literature. First, a comprehensive literature review for research and review papers is conducted in the Scopus database using different keywords. After eliminating duplicates and doing an initial screening, documents are selected. Table 1 lists the search phrases used to find the relevant publications and helps to understand the extent of work carried out and the need for the research work in India, a largely understudied region in this field. Heavy metals today have contaminated almost every aquatic ecosystem in the world. For evaluation of heavy metals in the sediments, two types of approaches are normally used. The first approach calculates the pollution level in the sediments using the total metal concentration, and the second approach uses the metal speciation fractions to evaluate the pollution level. Total metal-based approaches involved the calculation of pollution load index, geoaccumulation index, enrichment factor, potential ecological risk index etc. These indices evaluate the risk based on the background concentration of metals. In most cases, the background concentration unavailable and difficult to determine. In such cases, empirical relation or mathematical formula will be used to calculate the background, which can mislead the results. The metal speciation–based indices involved the calculation of pollution index, mobility factor, individual and global contamination factor, risk assessment code, modified ecological risk index etc. It has been found that the metal speciation approach is much more effective in describing the pollution status in sediments due to its understanding of the fractions that what metal contamination can be released from sediments and what is fixed to sediments. As such, different studies carried out the speciation of heavy metals in the sediment (Venugopal et al. 2009; Giridharan et al. 2010; Dhanakumar et al. 2013; Parthasarathy et al. 2021).

Exposure to heavy metal through ingestion or dermal contact pathways poses carcinogenic and non-carcinogenic effect on the human population. This is evaluated by calculating the hazard index and total cancer risk index. There

Table 1 Search terms used to identify the literature

Keywords	Occurrences	Relevant article
"Sediments" AND "heavy metals" AND "Hazard Index" AND "India"	11	7
"Sediments" AND "Metals" AND "positive matrix factorisation" AND "India"	1	1
"Sediments" AND "heavy metals" AND ("mobility Factor" OR "global Contamination factor") AND "India"	6	5

are two ways to calculate the two indices; one is deterministic method, and the second is probabilistic method. Models for deterministic risk assessment generally have only one value assigned to each model parameter, with the outcome being a single value for the risk associated with that parameter (Jiménez-Oyola et al. 2021). This technique is simple and easy to grasp, but input variables do not account for variability (Bazeli et al. 2020; Sheng et al. 2021). Moreover, this technique is built on a plausible exposure condition and thus conservative (Tong et al. 2018). The use of input parameter point values and assumptions might lead to an inaccurate risk assessment. The probabilistic method employs Monte-Carlo simulation to use a variety of distributions which fit the dataset to arrive at the risk results in the form of probability distributions. This method is useful for understanding the extent of risk and identifying the sensitive parameters of the risk assessment. Several researchers utilized these methods to conclude that children near the study areas have a high risk of carcinogenic and non-carcinogenic effects of heavy metals, more so than adults (Swarnalatha et al. 2015; Kumari et al. 2018; Bhat et al. 2022; Chaithanya et al. 2022; Kaur 2022; Arisekar et al. 2022).

Source identification of heavy metals, i.e. whether the metals originated from geogenic or anthropogenic sources, is very important after establishing the risk associated with heavy metals. PMF is regarded among the best at source apportionment because it deals with variability and uncertainty in different datasets. This technique is mostly used in the case of atmospheric pollutants (Belis et al. 2015; Amil et al. 2016; Hsu et al. 2016) but has recently been to water quality, sediments or soil (Mustaffa et al. 2014; Chen et al. 2015; Gholizadeh et al. 2016). The advantage of using the PMF is its ability to segregate the pollution sources much more easily than other multivariate statistical analyses (Reff et al. 2007; Dash et al. 2020). So, in this study, PMF has been used to segregate the metal pollution sources.

From the above review, it was found that studies on the assessment of human health risks, heavy metal mobility and identification of pollution sources (using the PMF model) were rarely performed or carried out in India (Dhanakumar and Mohanraj, 2018; Giridharan et al., 2010; Kumari et al., 2018; Parthasarathy et al., 2021; Singh et al., 2014; Swarnalatha et al., 2015). As a result, extensive field research was conducted to learn more about how heavy metals behave in the ecosystem of a river. For this project, the specific goals are to (1) investigate heavy metal multi-phase distribution and partition behaviour; (2) determine chemical fraction metal mobility and ecological risk; (3) use the PMF model to assess river sediment pollution sources; and (4) determine non-carcinogenic and carcinogenic risk.

Materials and methods

Study area

The Kolong River is a tributary of the Brahmaputra, which originated as a spill channel or anabranch (Pcba 2013). The river has its source in the Kukurakata and Hatimura hills. The river's total length is roughly 218.62 km, and it passes through the metropolises of Nagaon, Morigaon and Kamrup and average depth of 1–2 m with a maximum height of 4–6 m during the highest rainfall period (Bora and Goswami 2017). The Kolong River combines with the Kopili River, a vast southern river of the Brahmaputra near Jagibhakatgaon, Morigaon district, before returning to its source, the Brahmaputra River near Guwahati. Nagaon District is home to the Kolong River, which flows for more than 100 km. The river divides Nagaon township into two east–west halves, Haibargaon and Nagaon, in the middle of the township. The Kolong River receives water from several tributaries, including the Diju, Misha, Diphalu, Haria-Nanoi and Titaimari or Rahasuti. After receiving water from the aforementioned rivulets, the Kolong River swells. The Kopili River joins the Kolong River course in Morigaon district at Dukhutimukh in Jagibhakatgaon town (Singh et al. 2020; Bora and Goswami 2015). Kolong River was formerly the only source of potable water, and settlements built up along its banks. Nagaon's portion of the Kolong River is currently degraded, posing a health and hygiene risk to the community. The Kolong River in Assam Nagaon's district illustrates how human activities have exacerbated environmental issues over the previous half-century. The Kolong River turned into a succession of intermittent dry stretches and sluggish pools due to this massive human intervention over the years that followed. Because of its limited capacity for self-purification, the river is currently under a considerable degree of anthropogenic stress and acts as a major recipient of pollutants. As a result, the Central Pollution Control Board lists the Kolong River as one of India's most contaminated rivers. The Kolong River's quality can only be improved over the long run with a comprehensive restoration scheme (Bora and Goswami 2014; CPCB 2015).

Sample and data collection

Nine benthic sediment samples were obtained in the study zone, sequentially from upstream to downstream in the Kolong River, as depicted in Fig. 1. Between 2018 and 2019, monthly benthic sediment samples were collected from the river using a shovel (Simpson and Batley 2016; Spellman 2016; Tuit and Wait 2020) at the different

Fig. 1 Study area
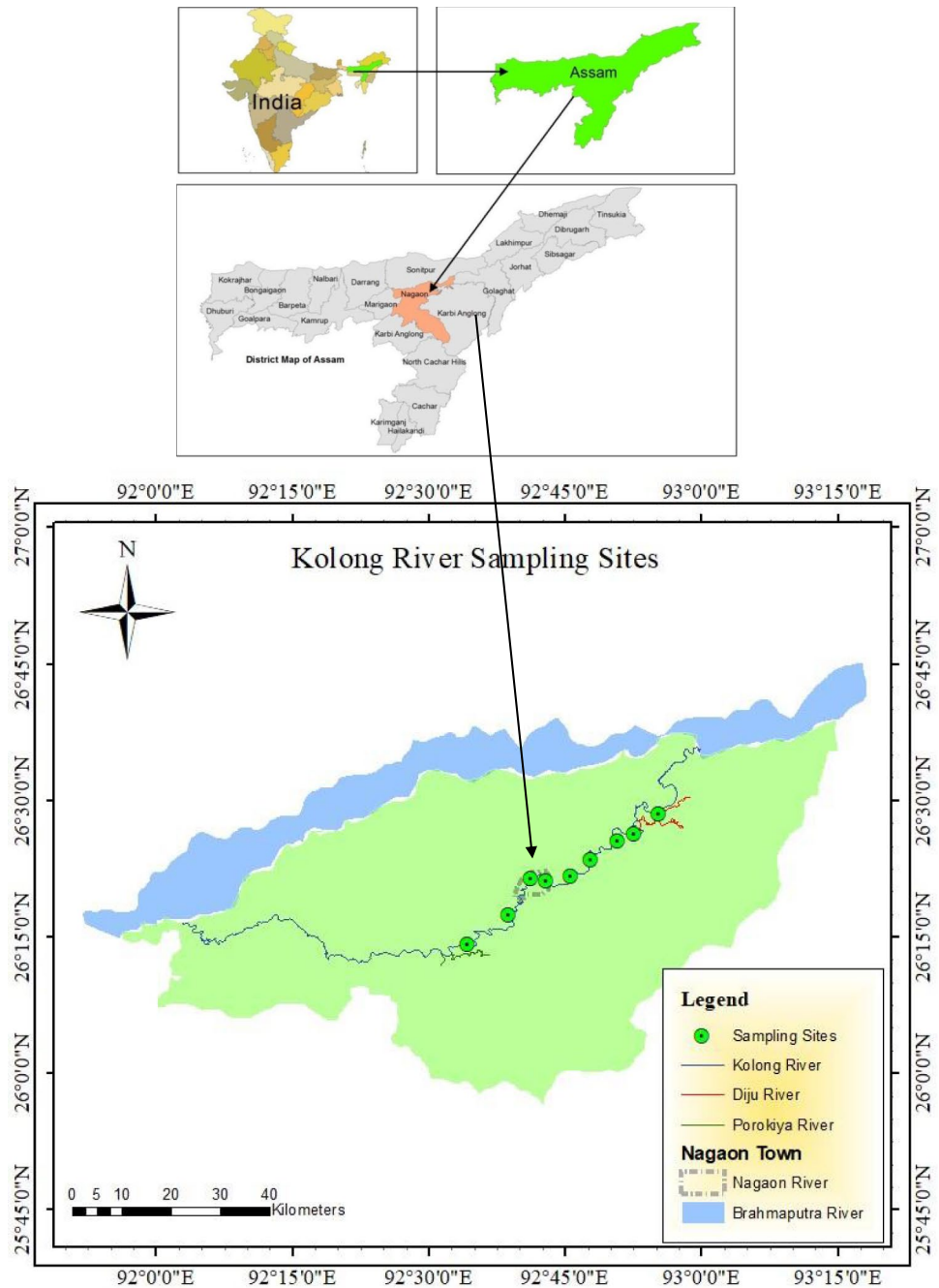
sampling points over four seasons: winter (W): December–February; pre-monsoon (PRM): March–May; monsoon (M): June–August; and post-monsoon (PTM): September–November (Goswami et al. 2021). The sediment samples were collected from the bed of the river where water depth was approximately 1 m by wadding the river from the upstream side in the four seasons (USEPA 2020). The depth of water in the winter, pre-monsoon and post-monsoon season is such that the samples were collected around the middle section whereas in the monsoon season, the samples were collected near the bank where the water

depth is safe to wade through (Bora and Goswami 2017). The obtained samples were stored in laboratory-grade zipped polythene bags. After collecting the samples, they were oven-dried at 105 ºC, and undesirable particles such as small pebbles, stone, wooden and grass particles were manually picked out. Samples were sun-dried and broken by hand before being oven-dried to eliminate water particles from the soil matrix and sieved through a 200-µ-mesh sieve for further testing. According to Tessier's sequential extraction procedures, analysis techniques were used to determine chemical partitioning (Tessier et al. 1979).

Chemical separation, according to Tessier et al. (1979), is defined as a set of operations that divide a sample into 5 fractions: exchangeable (M1), carbonate bound (M2), Fe–Mn oxide bound (M3), sulphide and organic matter bound (M4) and residual (M5) fractions. Atomic absorption spectrophotometer (AAS) was used to analyse the metals (Co, Cu, Cd, Fe, Mn, Pb and Zn). Quality control protocols were followed throughout the experiment's operations, including calibrating the equipment with freshly made standards before running it. All the chemicals used in the studies were of laboratory quality, and all dilutions were made with deionized water (Baird et al. 2017).

Mobility of heavy metals

To evaluate the retention of heavy metals in benthic sediment samples, the mobility factor (MF), individual contamination factors (ICF) and global contamination factor (GCF) of metals were computed.

Mobility factor

The proportions of the metal fractions are used to determine the mobility of heavy metals in the environment. The chemical fractions of metals bound to sediments were split into two categories based on the degree to which they were associated with distinct phases. The mobility factor was then calculated using the information from these two groups. The M1 and M2 fractions comprise the mobile group, whereas the stationary group comprises of the M3, M4 and M5 fractions. MF adapted from Kabala and Singh (2001) was modified for the 5-step sequential extraction technique of chemical partitioning, which describes the potential movement of the metals (Sut-Lohmann et al. 2022). The mobility factor (MF) was used to calculate the heavy metal mobility factor shown in Eq. (1):

$$MF = \frac{M1 + M2}{M1 + M2 + M3 + M4 + M5} \times 100(\%) \quad (1)$$

Contamination factors

Evaluating the heavy metal contamination factor is critical because it indicates the level of risk that heavy metals pose to the environment. Using the results from chemical speciation, ICF for each metal was computed by dividing the total of the first three extractions (M1, M2 and M3) by the residual (M5) fraction for each metal. The GCF for each site was calculated by summing the ICF for each heavy metal detected (Ikem et al. 2003; Naji and Sohrabi 2015). The ICF and GCF were determined with the help of the following equation:

$$ICF_i(\text{metal}) = \frac{M1 + M2 + M3}{M5} \quad (2)$$

$$GCF = \sum_{i=1}^{n} ICF_i \quad (3)$$

According to Zhao et al. (2012), the ICF and GCF categories were as follows: GCF < 6 and ICF < 1 indicates low contamination, 6 < GCF < 12 AND 1 < ICF < 3 moderate contamination, 12 < GCF < 24 and 3 < ICF < 6 considerable contamination and GCF > 24 and ICF > 6 high contamination.

PMF

The positive matrix factorization (PMF) receptor model is one of the most important and widely used methods in source allocation (Yan et al. 2019). In the PMF model, n (samples) × m (species concentration) matrix (X) is decomposed into two-factor matrices: profiles (F) and contributions (G) and residual (E) (Eq. 4). Two files were used as input to the PMF model: one comprising concentrations of the assessed parameters, and the other including uncertainty values derived using Eqs. (5) or (6). The ideal number of factors is obtained by repeatedly running the model for several iterations and then selecting the best run or solution with the minimum value of Q (robust), which indicates the model's ability to fit data (Jiang et al. 2019). Equation (7) has been used to determine this parameter. The primary goal of PMF factor analysis is to minimize Q for G and F while ensuring that all or some of G and F's components have non-negative values (Paatero 1997):

$$X = FG + E \quad (4)$$

If the species concentration (i.e. concentration of heavy metal (C_s)) was more than the standard deviation (SD), the uncertainty (Unc.) was calculated by Eq. (5); otherwise, the C_s was substituted by SD/2, and the uncertainty was presumed to be supplied by Eq. (6) (Wang et al. 2016):

$$Unc. = 0.1 \times C_s + \frac{SD}{3} \quad (5)$$

$$Unc. = 5 \times \frac{SD}{6} \quad (6)$$

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{s_{ij}} \right)^2 \quad (7)$$

where e_{ij} is the sum of squared differences between the original matrix data (X) and output of PMF (GF), and s_{ij} is the calculated uncertainties.

The investigation was conducted using the EPA PMF 5.0. The optimum number of factors was estimated by looking at the value of *Q*, which indicates how well the model fits the data. A global minimum was obtained for each model run by varying the seed value from 1 to 20 times (Bzdusek et al. 2006).

Human health risk assessment

While establishing whether or not heavy metals are harmful to human health, the assessment of health risks is a valuable tool. Both carcinogenic and non-carcinogenic effects of polluted soil or sediments have been observed due to pollution.

Exposure parameters

Dermal contact and incidental intake of sediments (during recreational activities) were used to estimate human health risks from exposure to heavy metals in children and adults. The chronic daily intake (CDI) was calculated using the USEPA's procedures (Eqs. (8) and (9)) in mg/kg) (US EPA 1989, 2001):

$$CDI_{ingestion} = \frac{C_S \times EF \times ET \times IR_S \times ED}{AT \times BW} \times CF \quad (8)$$

$$CDI_{dermalcontact} = \frac{C_S \times EF \times ED \times ET \times AF \times SA \times ABS}{AT \times BW} \times CF \quad (9)$$

The details and values of the terms in Eqs. (8) and (9) are given in Table 2.

Risk characterization

A risk assessment of heavy metal exposure, both non-carcinogenic and carcinogenic, was conducted to determine the health consequences following Eqs. (10) and (11). Hazard quotients (HQs) were used to compute the non-carcinogenic risk for all heavy metals through the exposure pathways. The hazard index (HI) was computed by adding all HQs. If HQ and HI are more than 1, the suggested acceptable limits have been exceeded (Phillips and Moya 2014). Potential carcinogenic health impact (CR) from inadvertent intake of sediments was estimated using Eq. (12). The cancer risk was evaluated using reported slope factors (SF_oral). The total cancer risk (TCR) was determined by

Table 2 Deterministic and probabilistic assessment parameters and values

Parameters		Deterministic approach	Probabilistic approach		Reference
		Point estimate (RME)	Distribution	Values	
EF	Exposure frequency: adults and child (day/year)	120	Triangular	120 (26 -260)	
ED	Exposure duration (year)				
	Adults	30	Lognormal	11.36 ± 13.72	Israeli and Nelson (1992)
	Child	6	Uniform	01–06	Anon (2019)
ET	Exposure time: adults and child (h/event)	2.6	Triangular	2.6 (0.5–6)	Anon (2019)
SA	Skin surface area (swimming) (cm^2)				
	Adults	23,000	Normal	$18,400 \pm 2300$	Phillips and Moya (2013)
	Child	7280	Normal	6800 ± 600	Anon (2019); Carr (1994)
BW	Body weight (kg)				
	Adults	70	Normal	72 ± 15.9	Carr (1994)
	Child	15	Normal	15.6 ± 3.7	Phillips and Moya (2013)
IR$_s$	Ingestion rate of sediments (mg/event)				
	Adult	12.5	–	12.5	Goldblum et al. (2006)
	Child	50	–	50	Goldblum et al. (2006)
AT	Averaging time (day)				
	Non-carcinogen	365xED	–	365xED	USEPA (1989)
	Carcinogen	365×70	–	365×70	USEPA (1989)
AF	Adherence factor (mg/cm^2)				
	Adult	0.07	–	0.07	USEPA (1989)
	Child	0.2	–	0.2	USEPA (1989)
ABS	Dermal adsorption factor (unitless)	0.001	–	0.001	USEPA (2011); Wang et al. (2017b, a)
K_p	Permeability constant (cm/hour)	0.001	–	0.001	RAIS (1992a, b)
CF	Conversion factor	0.000001	–	0.000001	

adding the CR values for each route of exposure and comparing them to permissible standard values (Yang et al. 2018; US EPA 2001). The reference dosage (R_fD) and slope factors (SF) were provided on the RAIS website (RAIS 1992a, b):

$$HQ_{ingestion} = \frac{CDI_{ingestion}}{R_fD_{oral}} \qquad (10)$$

$$HQ_{dermalcontact} = \frac{CDI_{dermalcontact}}{R_fD_{dermal}} \qquad (11)$$

$$CR_{ingestion} = CDI_{ingestion} \times SF_{oral} \qquad (12)$$

Deterministic method

Models for risk assessment generally have only one value assigned to each model parameter, with the outcome being a single value for the risk associated with that parameter (Jiménez-Oyola et al. 2021). This technique is simple and easy to grasp, but input variables do not account for variability (Bazeli et al. 2020; Sheng et al. 2021). Moreover, this technique is built on a plausible exposure condition and thus conservative (Tong et al. 2018). The use of input parameter point values and assumptions might lead to an inaccurate risk assessment. The deterministic approach's exposure parameters and generic exposure factors are shown in Table 2.
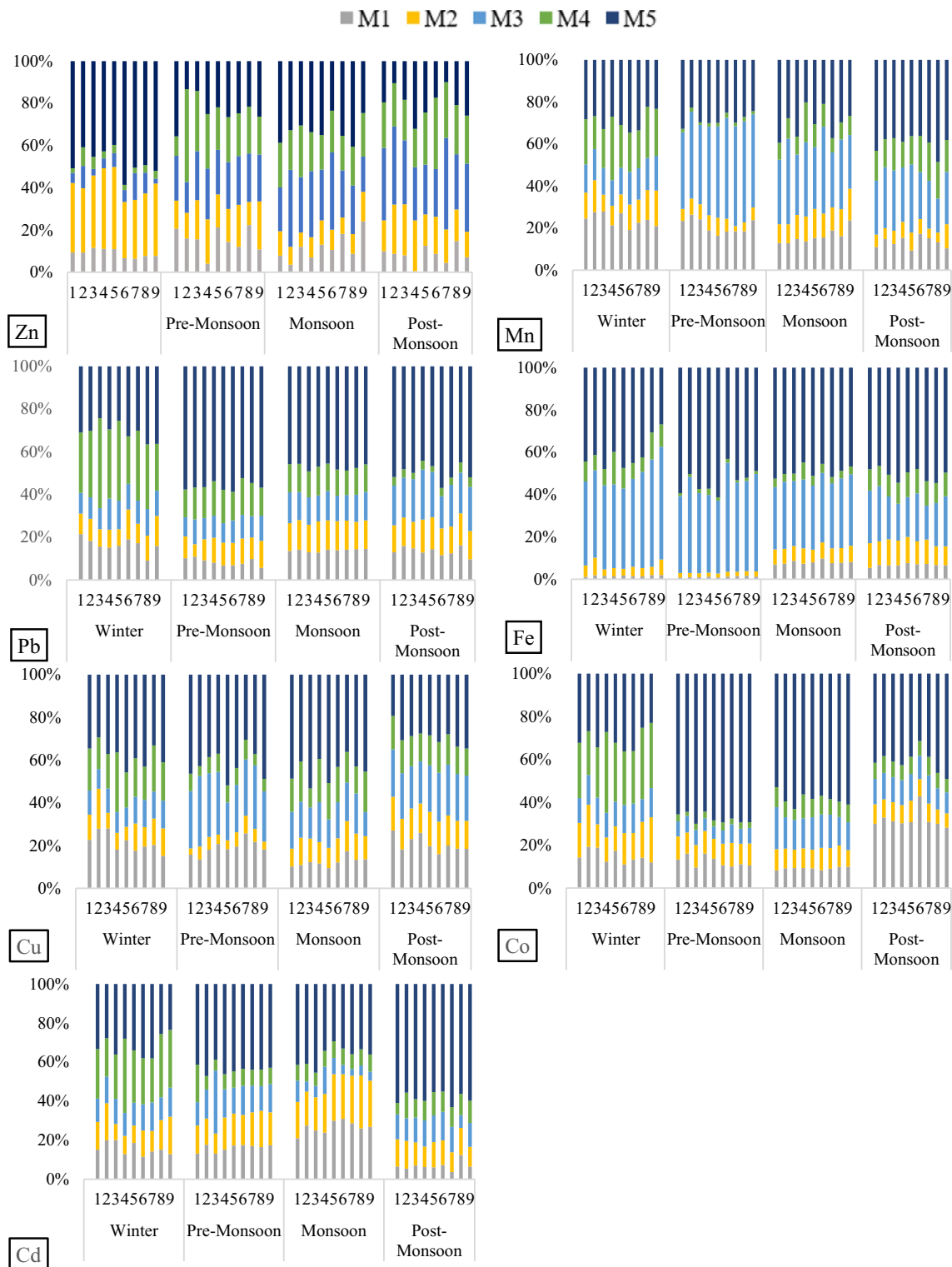
Probabilistic method

Monte Carlo simulation (MCS) is a frequently used approach in probabilistic risk assessment, in which variables are described by their distribution (Saha and Rahman 2020; Jiménez-Oyola et al. 2021). A wide variety of probable outcomes is generated using statistical sampling procedures in MCS in the form of probability distributions, which considers the data's inherent unpredictability and uncertainty (Low et al. 2015; Tong et al. 2018). MCS and other probability-based approaches are frequently used in human health risk assessment because they examine the sensitivity of the model input

Table 3 Concentration of heavy metals in Kolong river sediments

		Winter	Pre-Monsoon	Monsoon	Post-Monsoon
Zn (mg/kg)	Max	456.54	38.02	141.90	115.39
	Min	300.08	22.94	95.88	62.11
	Average	354.82	29.78	111.34	87.67
	Std. Dev	57.37	4.40	16.02	15.18
Mn (mg/kg)	Max	115.91	495.73	360.09	199.67
	Min	75.31	266.71	233.13	140.69
	Average	98.05	370.29	312.27	169.57
	Std. Dev	13.75	81.76	42.54	18.99
Pb (mg/kg)	Max	16.81	40.68	60.60	4.04
	Min	12.99	30.13	52.72	3.18
	Average	14.79	34.84	55.88	3.57
	Std. Dev	1.19	3.03	2.22	0.29
Fe (mg/kg)	Max	3440.40	9222.24	4229.54	2097.40
	Min	1898.21	6483.93	3442.47	1735.99
	Average	2912.47	7754.13	3927.93	1933.86
	Std. Dev	504.00	901.02	220.13	131.39
Cu (mg/kg)	Max	20.94	25.79	12.27	24.81
	Min	11.97	11.88	6.90	22.11
	Average	16.68	17.71	9.45	23.32
	Std. Dev	2.83	4.12	1.68	0.90
Co (mg/kg)	Max	14.69	33.42	28.42	11.05
	Min	9.16	27.73	23.21	8.68
	Average	11.82	29.67	24.85	9.61
	Std. Dev	1.71	1.96	1.76	0.80
Cd (mg/kg)	Max	12.67	6.87	5.18	3.36
	Min	7.50	4.93	4.03	2.64
	Average	10.04	5.35	4.52	3.03
	Std. Dev	1.64	0.65	0.39	0.25

variables (US EPA 2001; Saha et al. 2017; Bazeli et al. 2020; Saha and Rahman 2020). The non-carcinogenic and carcinogenic hazards were assessed in this work, using MCS as probabilistic modelling in Oracle Crystal Ball (Gitter et al. 2020; Xu et al. 2020). The number of iterations (for each run) was set at 10,000 to construct the probabilistic risk distributions (Yang et al. 2019). Heavy metal concentrations prior to MCS were fitted with the triangular distribution. For the

Fig. 2 Chemical speciation of surficial sediments in Kolong river

probabilistic evaluation, the standard distributions and exposure parameter values are shown in Table 2.

Sensitivity analysis

The impact of the input factors on cancer and non-cancer risk estimations is assessed using a sensitivity analysis based on the contribution to variance or rank coefficient correlation (Chen et al. 2019). This approach is commonly used in decision-making and risk management to discover the most critical factors influencing risk outcomes (Harris et al. 2017). The sensitivity analysis in this study was performed using the Spearman rank order correlation coefficient, which assesses how strongly and in which direction quantitative variables are connected to outcomes rather than the values

Fig. 3 Mobility factor (MF), and individual contamination factor (ICF) of (**a**) Zn, (**b**) Mn, (**c**) Pb, (**d**) Fe, (**e**) Cu, (**f**) Co and (**g**) Cd and their global contamination factors (GCF)

Fig. 3 (continued)

(e) Cu

(f) Co

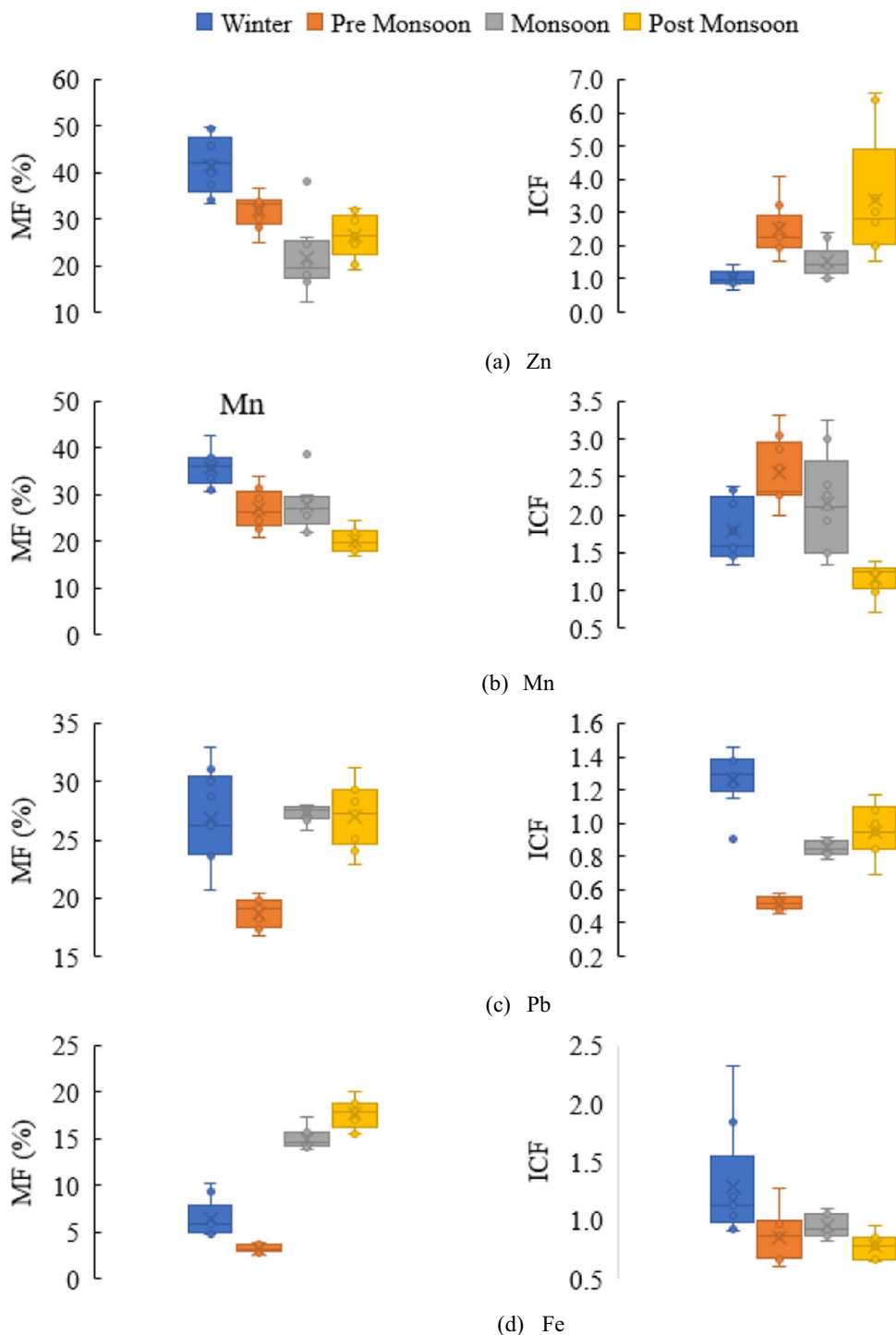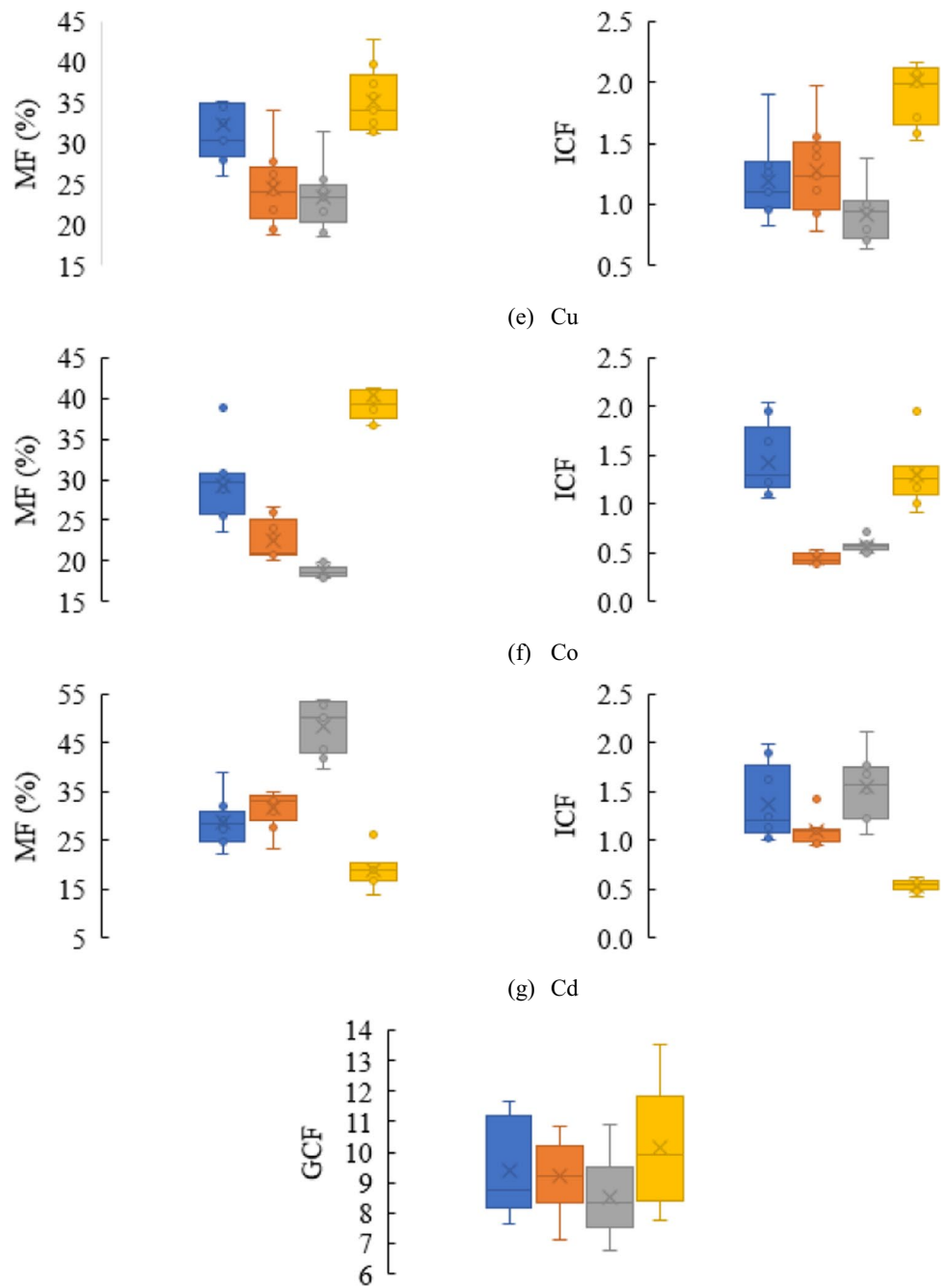(g) Cd

themselves (US EPA 2001). The sensitivity analysis was performed using a 95% confidence level and 10,000 iterations.

Results and discussions

Chemical speciation of heavy metals

Maximum, minimum, mean and standard deviation of concentrations of heavy metals in mg/kg of sediments in Kolong

Table 4 Summary of PMF for different runs for heavy metals in surficial sediments in Kolong river

Factor	Q_{exp}	Q (robust)	Q (true)	Q (true)/Q_{exp}
2	31	8.8501	8.8500	0.2855
3	15	1.7390	1.7390	0.1159
4	−1	0.8216	0.8216	−0.8216
5	−17	0.3065	0.3062	−0.0180
6	−33	0.0063	0.0058	−0.0002

Table 5 Correlation coefficients (R^2) of heavy metals between observed and predicted values by PMF model

Species	Intercept	Slope	SE	R^2
Zn	0.85	1.00	4.18	1.00
Mn	2.18	0.98	3.03	0.96
Pb	0.30	0.98	0.11	0.99
Fe	55.47	0.98	97.61	0.97
Cu	0.26	0.98	0.22	0.99
Co	0.13	0.99	0.13	0.99
Cd	0.10	0.99	0.10	1.00

River benthic sediments during the various seasons of sampling are presented in Table 3.

For Zn, there is a drastic decrease in concentration with the onset of monsoon indicating metals get desorbed from the sediments into the water body. This can also be correlated with the chemical speciation results (Fig. 2), wherein there is decrease in the percentage share of M1, M2, and M3 fractions, which is part of the metal fractions which can easily get desorbed (Pardo et al. 1990). For Mn, the opposite phenomenon was recorded for the sediments, wherein metal concentration increased drastically from winter to pre-monsoon. The arrival of monsoon brings in pollutants from various sources of pollution in the water body, which get deposited onto the sediments. Similar increase is also observed in case Pb (winter to monsoon), Fe (winter to pre-monsoon) and Co (winter to pre-monsoon). This might be due to anthropogenic metal contamination, as in the case of Mn, Fe, Pb and Co, the increase in metal contamination directly correlates with the M3 fraction (Morillo et al. 2002). The M3 fraction of the metals might have adsorbed on the sediments during the turbulent flow of the river (Lim and Kiu 1995; Baruah et al. 1996; Akindele and Olutona 2014). The concentration of Cu and Cd is fairly constant as it is with the percentage of metal fractions in the sediments. Finally, from the chemical speciation study, it observed that anthropogenic addition of pollutants is present in the case of Mn, Pb, Fe, and Co.

Mobility of heavy metals

MF, ICF and GCF

The box plot in Fig. 3 depicts the changes in MF, ICF and GCF temporally and spatially. The MF factor represents the mobility of bioavailable fractions in the sediments. The greater the amount of mobile fractions present in the sediments, the more is the risk for the ecosystem, as the fractions can be released from the sediments into the stream. Zn, Mn and Pb have the highest MF in winter, whereas Cu and Co have the highest MF in the post-monsoon season,

i.e. in the dry season. In the wet season, i.e. the pre-monsoon and monsoon, the MF has decreased, which might be due to the desorption of mobile metal fractions from the sediments due to the increase in turbulence in the river (Akindele et al. 2020). For Cd, the exact opposite happened, wherein MF is higher in the wet season than in the dry season. This indicates that turbulence has an opposite effect on Cd, which leads to adsorption of Cd onto the sediment particles. For Fe, there is a constant increase of bioavailable metals from winter to post-monsoon season, which MF represents, indicating anthropogenic addition of metals in the river and, in turn, setting down onto the sediment particle.

The ICF value indicates the anthropogenic contamination with respect to each metal, whereas the GCF value gives an idea about overall contamination at a site. The ICF value for Zn and Cu is highest in post-monsoon, whereas Co, Fe and Pb have the highest values in the winter. For most metals, dry seasons pose a higher risk than wet seasons. This indicates that metal gets adsorbed onto the sediment particles during the lean flow period, i.e. during the winter and post-monsoon seasons, and gets desorbed during the high turbulent period, i.e. during the pre-monsoon and monsoon seasons. The ICF values of Zn constantly rose from low contamination in the winter to moderate contamination indicating the Zn is continuously added to the river anthropogenically or externally. Mn, Pb, Fe, Cu, Co and Cd have 'moderate to low' contamination, whereas Zn has the 'high' contamination value. The cumulative effect of these metals at a site is represented by GCF, which has increased after the monsoon season (in post-monsoon) to considerable contamination, compared to the pre-monsoon and monsoon seasons, where the majority of GCF values fall into the moderate contamination category. This indicates that with the onset of monsoon or with increase in turbulence in the river, desorption of heavy metals from sediment might have occurred as there is decrease in labile or bioavailable fraction of metals. In other words, with monsoon receding, i.e. in post-monsoon season, the bioavailable fractions of metals in sediments rise, indicating the metal getting adsorbed onto the sediment particles giving an increased risk (Akindele and Olutona 2014; Mna et al. 2021). From this study, it is observed that the tendency for metals to accumulate in sediments happens in dry seasons or the lean flow period of the river. Thus, the anthropogenic addition of metals in the Kolong River might be happening during the dry season, which can be the reason for high values during this period (Akindele et al. 2020). When metals are added to the sediments, they are an unstable bond which makes them bioavailable, and in such cases, the MF, ICF and GCF will show high values as in the study area.

PMF

Q values were computed after iterations with a number of factors ranging from 2 to 7. Four factors were considered ideal because $Q_{\text{True}}/Q_{\text{exp}}$ for iteration, in this case, is the least (Table 4). The model's fitness was determined by comparing observed and predicted heavy metal values, yielding the correlation coefficients (R^2) (Fig. S1). R^2 values greater than
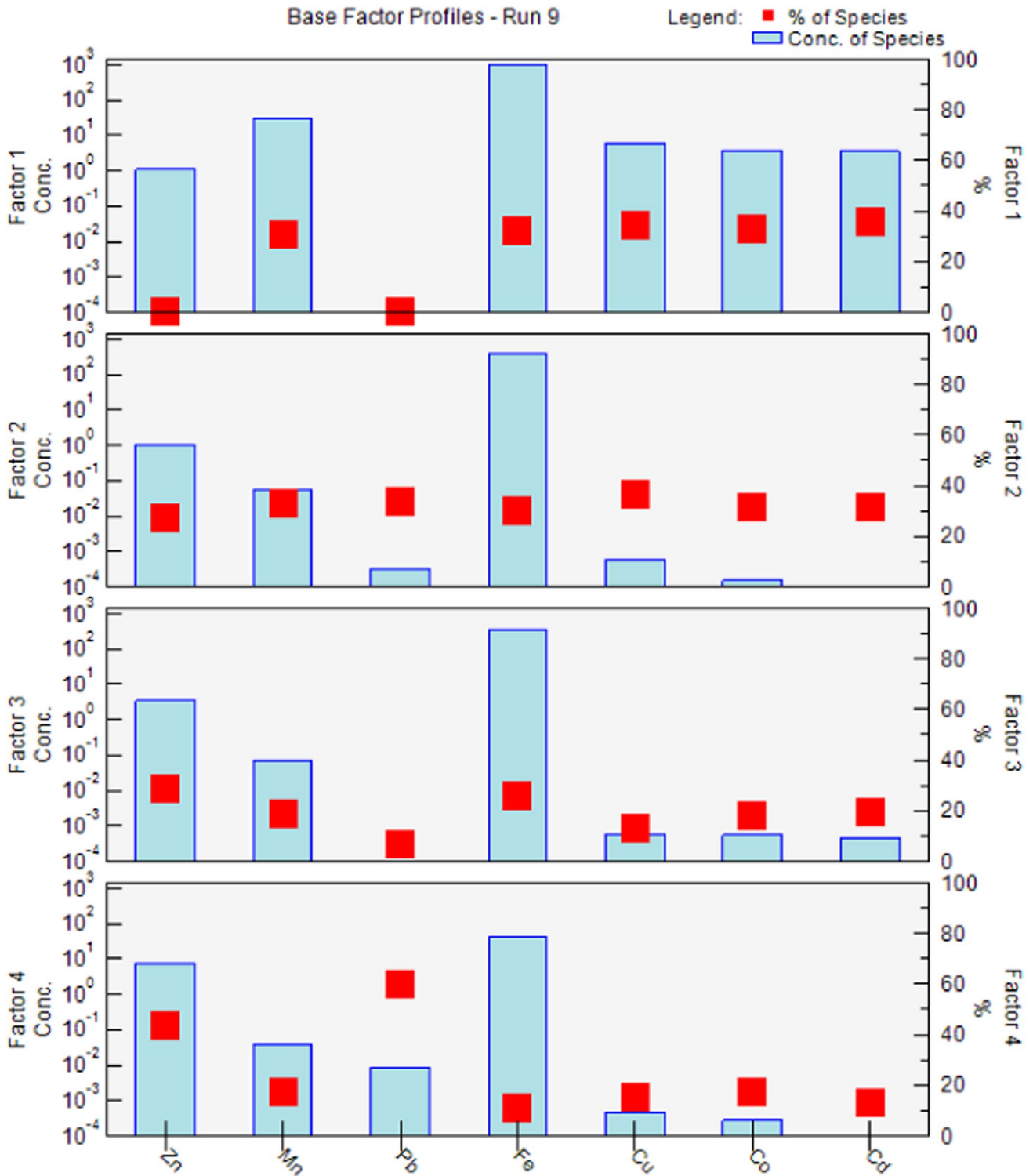

Fig. 4 Heavy metals (%) contribution to the 4 factors

0.95 were found in all heavy metals, indicating a strong relationship between predicted and observed values and therefore a high level of model dependability (Table 5).

Pollution source apportionment

Heavy metal toxicity to human health and the environment involves the usage of source allocation models, viz. PMF model, in order to decrease the likelihood of future contamination and, as a result, to more efficiently manage resources. The current investigation considers four factors that influence the source of pollution in the Kolong River. Figure 4 shows that the contribution to factor 1 is shown only by Mn, Fe, Cu, Co and Cd. The presence of Cu, Co and Cd can mainly be attributed to anthropogenic sources of contamination from the nearby region. So, factor 1 can be said to be related to anthropogenic contamination. In the rest of the factors, it is observed that the contribution is only from Fe, Mn and Zn. As reported in various works of literature, the source of these metals in northeast India is natural or lithogenic (Borah et al. 2009; Haloi and Sarma 2012). Thus, from source apportionment analysis using PMF, it can be concluded that among the four factors, factor 1 can be related to anthropogenic contamination and factors 2, 3 and 4 can be said to be associated with natural or lithogenic contamination.

Health risk assessment

Human health is jeopardized if hazardous chemicals are found in sediment samples. The dangers of heavy metal exposure on the health of both adults and children were investigated in this study. Deterministic and probabilistic approaches produced risk results exceeding the USEPA's safe exposure limit (US EPA 1989, 2001).

Deterministic method

Figure 5 depicts the deterministic method of assessment of carcinogenic and non-carcinogenic risks associated with heavy metal exposure in sediments. During the sampling period, which included several seasons, the results of the deterministic analysis revealed unsatisfactory results, namely, HI > 1 and TCR > 10^{-5}. Non-carcinogenic risk for adults was highest in the pre-monsoon, with a value of 27.17 compared to a reference value of 1. They tend to decrease with increased precipitation, which directly correlates with MF, which is also seen to decrease with the onset of monsoon. The non-carcinogenic effect of these metals poses a serious health hazard for persons in the lower age group (i.e. children) compared to persons in the higher age group (i.e. adults). Fe poses serious health concerns among the metals

Fig. 5 HQ, HI and TCR outcomes of deterministic method for heavy metals through the different exposure routes
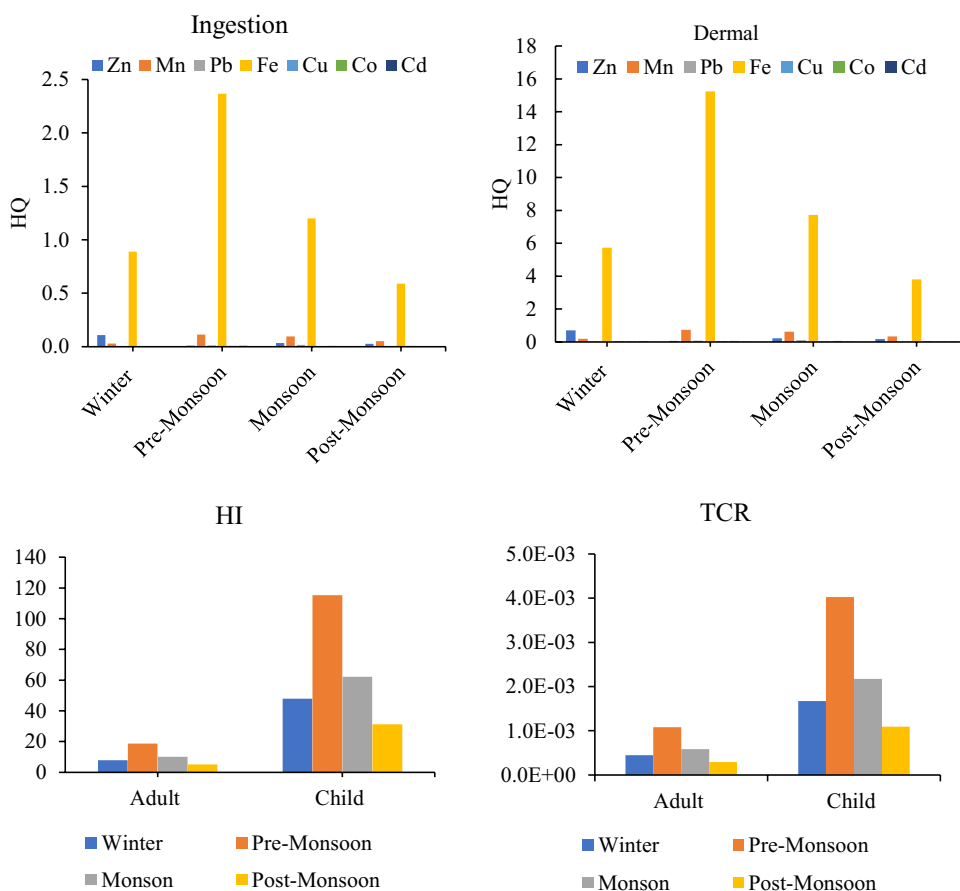
analysed in the study area as its values for all the seasons for adults and children are greater than 1 (Fig. 5). Among the exposure routes, dermal absorption poses more concern for the inhabitants of the nearby area, thereby mainly restricting any recreational usage of the river. Considering the carcinogenic impact of heavy metals on the population of humans, the direct usage of the river should be banned as the TCR values are more than 1×10^{-5} for all seasons.

Probabilistic method

The values of any parameter vary temporally and spatially during the sampling period; calculating risk using point deterministic values would give us an idea about the hazard posed by that parameter but would present a full picture of the risk posed for the analysis. Also, quantifying each and every point at different points of time would be tedious and may result in calculation errors. In such cases, the probabilistic risk calculation method would give a more descriptive result of the parameter variation and help in quantifying the spatial and temporal risk posed by the parameter. In this method, the data of heavy metals collected for different seasons is first fitted to a triangular distribution and then run through a Monte-Carlo simulation (10,000 iterations) to quantify the cancerous and non-cancerous risk posed through the different routes of

exposure, i.e. ingestion and dermal. The 5th and 95th percentile values of HI and TCR are given in Fig. 6, and their probability-frequency distribution curves for the complete dataset of the different seasons are provided in Fig. S2-S5. The results show that neither in the wettest or driest months do the HI and TCR values fall below the permissible value of 1 and 10^{-5}, respectively. The situation is harsher for the child population, where the values are even higher. The HI values in order 10^2 and TCR values are in order 10^{-3}, which is way higher than the permissible limits. The health risk assessment study thus calls for the treatment of the water before use and stringent policies to be framed for pollution abatement.

Sensitivity analysis

Figure 7 shows the sensitivity analysis results to assess the prime parameters concerning the non-carcinogenic and carcinogenic risks. Exposure frequency (EF), exposure time (ET), body weight (BW) of the population exposed to the risk, contact surface area (SA) in case of dermal exposure and Fe concentration (Cs-Fe) are important risk factors for non-cancer risk. The contributing or differentiating factor causing the high risk in the winter months is the concentration of Zn (Cs-Zn). In the case of carcinogenic risk, the significant factors contributing to risk are body weight (BW) of the population

Fig. 6 HI and TCR from exposure to heavy metals through probabilistic method (5th and 95th percentile)
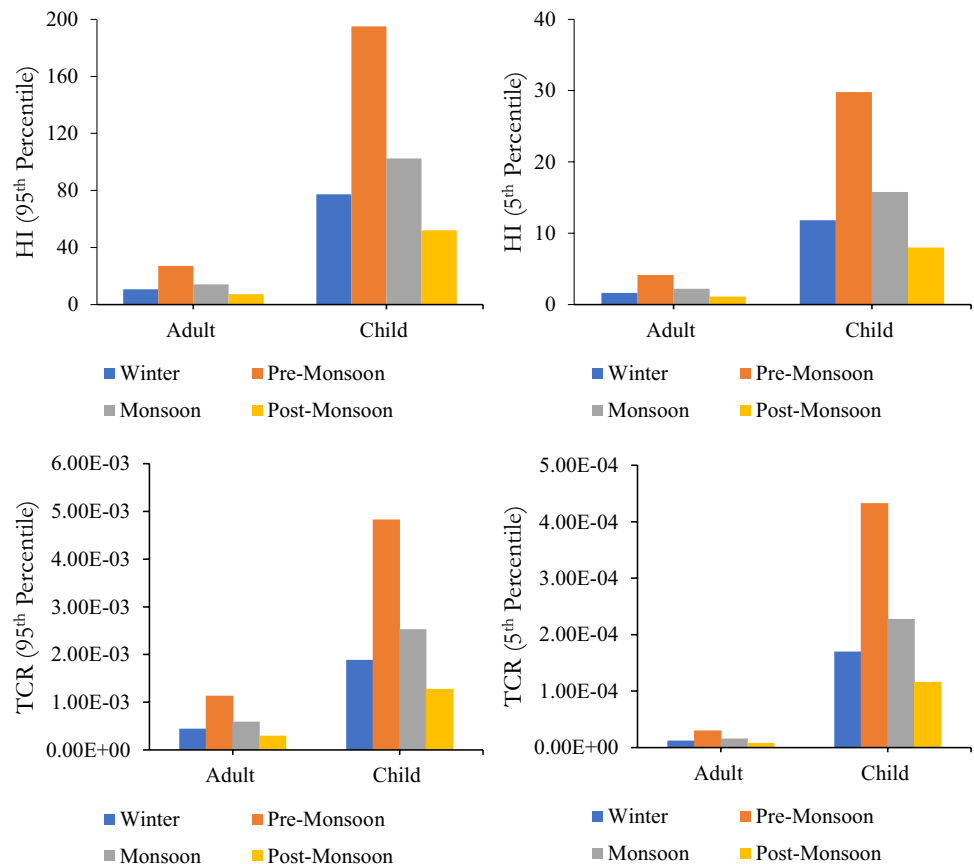
Fig. 7 Rank correlation charts of inputs of probability analysis (sensitivity analysis). **a** Winter. **b** Pre-Monsoon. **c** Monsoon. **d** Post-monsoon
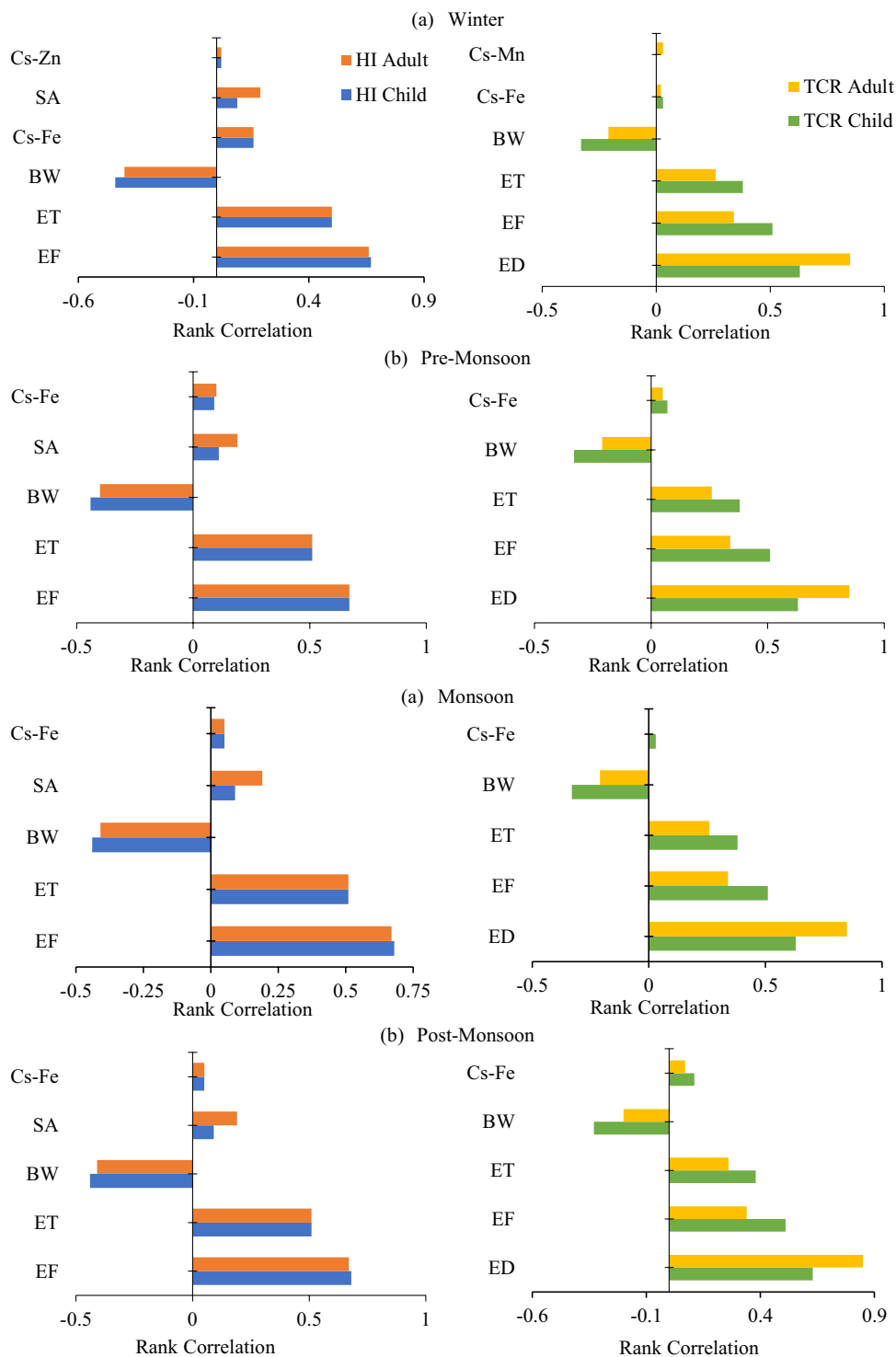
exposed to the risk, concentration of Fe (Cs-Fe), exposure duration (ED) and exposure frequency (EF) and exposure time (ET). The contributing or differentiating factor causing the high risk in the winter months is the concentration of Mn (Cs-Mn). Thus, from sensitivity analysis, it can be concluded that the focus of remediation study in the region should primarily focus on the concentration of Fe, Mn and Zn. If required, focus can also be shifted towards the rest of the heavy metals.

Conclusion

The mobility factor (MF) and the individual contamination factor (ICF) for most metals analysed were higher in the winter or post-monsoon months, i.e. during the dry season. However, the variation of Cd was the exact opposite to the other metals, i.e. higher values were found in pre-monsoon and monsoon season (wet period) and identified

the increase in the Fe and Zn contamination due to anthropogenic addition of pollutants. Global contamination factor (GCF) showed that the post-monsoon season had the most considerable contamination, followed by the winter, pre-monsoon and monsoon season. This contamination in benthic sediments leads to a very distinct result during high rainfall seasons wherein flow in the river is turbulent; the contamination falls from higher form contamination to lower form contamination due to the desorption of metals from sediments. The simulation of the PMF model segregated the metal contamination, the contamination of Cu, Co and Cd mainly due to anthropogenic sources while Fe, Mn and Zn contamination due to lithogenic or natural sources. Finally, health risk assessment revealed that exposure to Kolong River is hazardous to human health, with dermal absorption presenting the greatest concern among the exposure pathways. Sensitivity analysis attributed the variation in the risk to Mn for carcinogenic and Zn for non-carcinogenic risk. Thus, this study calls for adequate regulatory strategies to be formulated and continuous environmental monitoring programs to be carried out to reduce the pollution in the river.

Author contribution *Ankit Pratim Goswami:* conceptualization, methodology, formal analysis, resources, investigation, data curation, writing—original draft. *Ajay S. Kalamdhad:* conceptualization, resources, writing—review and editing.

Declarations

Ethics approval Not applicable.

Consent to participate For further clarification and information, I shall be free to contact any of the people involved in the research.

Consent for publication Not applicable.

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