HEALTH RISK ASSESSMENT OF HEAVY METALS IN KUANTAN RIVER BASIN, PAHANG

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ABSTRACT

Introduction: Industrialisation, urbanisation, and nearby human activities can lead to heavy metal pollution in the Kuantan River basin, posing a health risk. **Objective:** This quantitative secondary data analysis study aimed to estimate the health risk assessment (HRA) of the arsenic (As), lead (Pb), cadmium (Cd) and mercury (Hg) in the Kuantan River basin. **Methods:** All the secondary data on heavy metals concentration, physicochemical parameters and rainfall were obtained from the Department of Environment (DOE) and the Meteorological Department (MET), respectively. In reducing the uncertainties in health risk estimation, the Monte Carlo simulation technique was used to estimate the Hazard Quotient, HQ and Lifetime Cancer Risk, LCR of studied heavy metals via ingestion and dermal absorption. Sensitivity analysis (SA) was used to measure the most influential parameters on the estimated risk. **Results:** The results demonstrated that in 2017, the ascending order of mean concentrations of heavy metals was Pb > As > Cd > Hg. However, from 2018 to 2021, Cd and Hg consistently fell below the detection limit, leaving Pb > As as the dominant heavy metals during this period. All mean HQ values of studied heavy metals did not exceed the acceptable limit of one for both ingestion and dermal uptake. The mean LCR values for As via ingestion and dermal exposure remained within acceptable limits (1×10^{-4}). However, in 2017, the mean LCR value for Cd through dermal uptake (1.40×10^{-4}) slightly exceeded the acceptable level. Meanwhile, the worst-case predicted values at the 95th percentile showed that the LCR values of As via ingestion route (all years), ranged from 1.48×10^{-4} to 2.98×10^{-4} and dermal uptake (except for 2019 and 2021), ranged from 3.80×10^{-3} to 1.99×10^{-4}, exceeded the acceptable limit. SA demonstrated that the concentration of As and Cd had the greatest influence on HRA via the ingestion pathway and dermal uptake in 2017. **Conclusion:** It indicates As and Cd potentially increasing the chance of developing cancer. This study will help individuals in high-risk heavy metal exposure areas be more cautious and aid authorities in mitigating contamination.

KEYWORDS: heavy metals, river water, hazard quotient, lifetime cancer risk, Monte Carlo simulation

INTRODUCTION

Over 700 substances have been identified as contaminants in the waters, with heavy metals posing the biggest threat to the environment and individuals due to their high toxicity and carcinogenicity (Ustaoglu and Aydin, 2020). It has been documented that in developing countries of Africa, Asia, and South America, river water often contains elevated heavy metal concentrations, exceeding global threshold limits (Zhou et al., 2020). Chromium (Cr), nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd), and lead (Pb) are examples of heavy metals that tend to exhibit greater persistence compared to organic contaminants (Lou et al., 2017), and some are able to accumulate and biomagnified. Therefore, long-term exposure to heavy metals, can exacerbate health issues and contribute to chronic conditions, with unregulated heavy metal deposition in rivers, as noted by Liu & Ma (2019), posing significant health risks such as carcinogenic effects, neurological damage, impaired growth, and even fatalities.

Heavy metal pollution, originating from natural and human activities, is particularly prominent in Asian countries, primarily due to mining and manufacturing (Zhou et al., 2020). Malaysian rivers, including those in the Kuantan district, grapple with heavy metal contamination. Prior studies have identified elevated levels of copper (Cu), arsenic (As), iron (Fe), manganese (Mn), aluminum (Al), and magnesium (Mg) in various Malaysian rivers (Liang et al., 2020; Razak et al., 2021; Maharjan, Wong, and Rubiyatno, 2021). Rapid urbanisation and industrialisation in Kuantan, coupled with a high-density population, have led to concerns about heavy metal pollution. Recent reports indicate contamination of Tunggak and Balok rivers (Gebeng Rivers), Kuantan, with Cd, Co, Cu, Pb, Mn, and Ni from industrial sources (Islam et al., 2022). Kuantan's population, the largest among nine districts in Pahang, engages in land-use practices that may exacerbate heavy metal pollution in the rivers. Despite this, the water quality status of the Kuantan River basin has consistently been classified as slightly polluted from 2016 to 2020, according to data published by the Department of Environment (DOE, 2017-2020). This persistent pollution could pose health risks to individuals engaging in activities like swimming, fish consumption, or groundwater usage.

Health risk assessment (HRA) as suggested by the United States Environmental Protection Agency (US EPA) serves as a valuable tool for evaluating health risks in humans. There are numerous published studies on heavy metal contamination in water and their health risk assessment (Jin et al., 2015; Ustaoglu and Aydin, 2020; Islam et al., 2015; Fahimah et al., 2023; Adesiyan et al., 2018; Mahad et al., 2019). This study aimed to determine the hazard quotient (HQ) and lifetime cancer risk (LCR) of As, Cd, Pb and Hg in the Kuantan River basin coupled with the Monte-Carlo simulation technique in reducing the uncertainty of HRA (Liu et al., 2019; Jin et al., 2022), together with a sensitivity analysis to determine which parameters are the most influential on the health risk estimation's outputs. The identification of parameters with the greatest impact on health risk can guide related stakeholders in efficiently channeling their efforts and resources toward addressing these critical parameters.

MATERIALS AND METHODS

Study Design and Source of Data

Quantitative secondary data analysis was used as the study design for this research study. The term "secondary analysis" describes the method of using existing data from previous studies or primary data collected by other researchers or agencies to address new questions (Tripathy, 2013). The secondary data which is the concentration of As, Cd, Hg, and Pb in the Kuantan River basin were obtained from the Department of Environment (DOE) for HRA. Taking into account the influence of physicochemical parameters and rainfall, the data on dissolved oxygen (DO), biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids (SS), pH, and ammoniacal nitrogen (NH_3-N) from DOE for the Kuantan River basin were also obtained. Additionally, we sourced rainfall data for the Kuantan district from the Meteorological Department as supplementary information, as it may impact the measured levels of As, Hg, Cd, and Pb.

Study Area

The 87 kilometres long Kuantan River basin flows through the Kuantan district, which is in Pahang State's north-eastern region. Kuantan River basin was chosen due to the urbanisation and rapid development of industrial progress as it can increase the discharge of pollutants into the ecosystems. This is because there are many development areas near the Kuantan River basin including commercial, residential, lodging and chemical industries. There are 192 manual river water quality monitoring (MRWQM) stations and two automatic river water quality monitoring (CRWQM) stations located throughout the Kuantan River basin. The data obtained was based on these water quality monitoring stations. The selected rivers in the Kuantan River basin included rivers of Belat, Charu, Galing Besar, Galing Kecil, Kenau, Kuantan, Pandan, Pinang, Reman, Riau and Talam. The water monitoring stations in Kuantan River and Belat River were selected due to being the largest and second largest rivers in Kuantan respectively, with Belat River being a part of Kuantan River. Both rivers serve as the source of water supply for domestic, industrial, and agricultural usage. Additionally, the Kuantan River is near to Gebeng Industrial Estate as the water supply for industry purposes. Meanwhile, the Galing River (part of the Kuantan River) was chosen as it is located in the most urbanised area in Kuantan.

Health Risk Assessment

HRA of As, Cd, Hg, and Pb was observed via the two main exposure pathways: ingestion of water and dermal absorption through the skin. Hence, the average daily dose (ADD) for ingestion of water ($ADD_{ingestion}$) and dermal absorption (ADD_{dermal}) was required to be calculated using equations (1) and (2) suggested by EPA (1989) and had been used in the previous studies such as (Adesiyan et al., 2018; Jin et al., 2022; Liang et al., 2018; Ustaoğlu & Aydın, 2020). Both the $ADD_{ingestion}$ and ADD_{dermal} expressed in unit µg/kg/d.

$$ADD_{ingestion} = \frac{(C_{water} \times IR \times ABS_g \times EF \times ED)}{(BW \times AT)} \qquad (1)$$

$$ADD_{dermal} = \frac{(C_{water} \times SA \times K_p \times ET \times EF \times ED \times CF)}{(BW \times AT)} \qquad (2)$$

Where;

C_{water}	= concentration of heavy metals in water	SA = surface area
IR	= ingestion rate	K_p = dermal permeability coefficient
ABS_g	= gastrointestinal absorption factor	EF = exposure frequency
ED	= exposure duration	ET = exposure time
BW	= body weight	AT = averaging time
CF	= conversion factor	

The calculation for non-carcinogenic and carcinogenic risk is required to evaluate HRA indicated by the Hazard Quotient (HQ) and Lifetime Cancer Risk (LCR), respectively. The ratio of the average daily dose for direct ingestion or dermal contact to the reference dose (RfD) is known as HQ calculated using the following equation below (3 & 4). Additionally, LCR indicates the carcinogen elements that can induce potential danger for humans. It is calculated with the following equation (5) where cancer slope factor (CSF) is required:

$$HQ_{ingestion} = \frac{ADD_{ingestion}}{RfD_{ingestion}} \qquad (3)$$

$$HQ_{dermal} = \frac{(ADD_{dermal})}{(RfD_{dermal})} \qquad (4)$$

$$LCR = ADD \times CSF \qquad (5)$$

If HQ > 1, it indicates that heavy metal exposure has a negative impact on human health. Meanwhile, if HQ < 1, then there will be no adverse effects arising on human health (Ustaoglu and Aydin, 2020). There is a carcinogenic risk if $LCR \geq 10^{-4}$, which indicates a high risk of developing cancer in humans (Ustaoglu and Aydin, 2020).

In estimating the Hazard Index (HI), the total of As, Pb, Cd, and Hg was computed to determine the impact of multiple heavy metals in the Kuantan River basin to the residents. HI was calculated using the USEPA guideline (Mohammadi et al., 2019; US EPA, 1991), as shown in equation (6) below:

$$HI = \sum_{HQ} = HQ_{As} + HQ_{Pb} + HQ_{Cd} + HQ_{Hg} \qquad (6)$$

Monte Carlo simulation

Both Monte Carlo simulation and sensitivity analysis have been conducted by using Crystal Ball software (version 11.1.3.0.0; Oracle Corp., USA). Apart from that, HRA as a deterministic assessment commonly has uncertainty or variability characteristics that cause inaccuracies in the assessment results due to single-point input parameters. Therefore, a probabilistic method such as Monte Carlo analysis was carried out because it could produce a result that was more accurate by reducing uncertainties (Liu et al., 2019; Qu et al., 2018). In this study, iterations of 10,000 were applied to determine the ingestion and dermal risk associated with exposure to As, Hg, Cd and Pb. Additionally, the parameter used in the Monte Carlo simulation such as heavy metal concentration was defined as the log-normal distribution. Meanwhile, the parameters (IR, ED, BW, AT, SA, EF, ET, CF, K_p, RfD, ABS_g, CSF) that have fixed values were defined as uniform distributions. Moreover, the Monte Carlo technique can identify the most influential parameter using sensitivity analysis (Liu et al., 2019). To identify the parameters influencing the uncertainty risk of HQ and LCR, the outcomes of the most influential parameters were presented in the rank correlation coefficient. This is because the correlation coefficient between each of the parameters and the value of risk will determine the sensitivity. Each element's contribution is bigger when it has a higher correlation coefficient since it contributes more to the outcomes (Qu et al., 2019).

RESULTS AND DISCUSSIONS

Concentration of heavy metals

Figure 1 below shows the mean for the concentration of studied heavy metals in the Kuantan River basin from 2017 to 2021. In 2017, the mean concentrations of the examined heavy metals followed the order Pb > As > Cd > Hg. However, from 2018 to 2021, Cd and Hg consistently were below the detection limit, with Pb > As emerging as the dominant heavy metals during this period. The figure shows the highest mean concentration of Pb recorded in 2019 (3.53×10^{-3} mg/L) and the highest mean concentration for As was reported in 2017 with 2.00×10^{-3} mg/L. In the year 2017, Cd and Hg were third (1.00×10^{-3} mg/L) and fourth-ranked (2.00×10^{-4} mg/L), respectively. However, the mean concentrations of Cd and Hg from 2018 to 2021 are unable to be analysed due to those two heavy metals below the detection limit (< 0.001 mg/L).

As and Pb were recorded as the highest and second-highest average concentrations over the subsequent five years. It can be associated with uncontrolled bauxite mining activities in Kuantan until it was banned in early 2016 (Abdullah et al., 2016). Apart from that, Abdullah et al. (2016) also stated the main contaminants of bauxite are Al and Fe but due to geological characteristics of the land and land-use activities, the other toxic metals such as As, Hg, Cd, Pb, Mn and Ni may deteriorate the quality of water when the natural ecosystem is aggressively discovered and excavated. Pb concentration peaked in 2019 meanwhile As in 2017. Although Pb is very little present on the earth's crust, it can be transported and dissolved in water (ATSDR, 2020). Moreover, there is a high possibility that Pb concentration may be high due to the municipal waste that may contain metal such as the usage of batteries or from the types of transport that use fuels containing Pb. This is because the major sources of Pb commonly come from paint, pesticides, fuels, batteries, industrial waste or fertilizers (Tadesse, Tsegaye & Girma, 2018).

Additionally, Kusin et al. (2016) stated, that an elevated concentration of As in the water can be associated with the leaching process of bauxite stockpiles that allows the heavy metals to enter the water directly. Since the mining operation was stopped in 2016, there is a high chance the high concentration of As in 2017 was caused by the leaching process. In addition, Cd, Hg and Pb concentration levels also showed approximately similar trends in 2017. Moreover, the previous studies found the elements of Cd, Hg and Pb in the soil particles (Ismail et al., 2018) and the Pb levels exceed the recommended value which is > 35 mg/kg in sediment (Kusin et al., 2018) in the vicinity of the Kuantan mining areas.

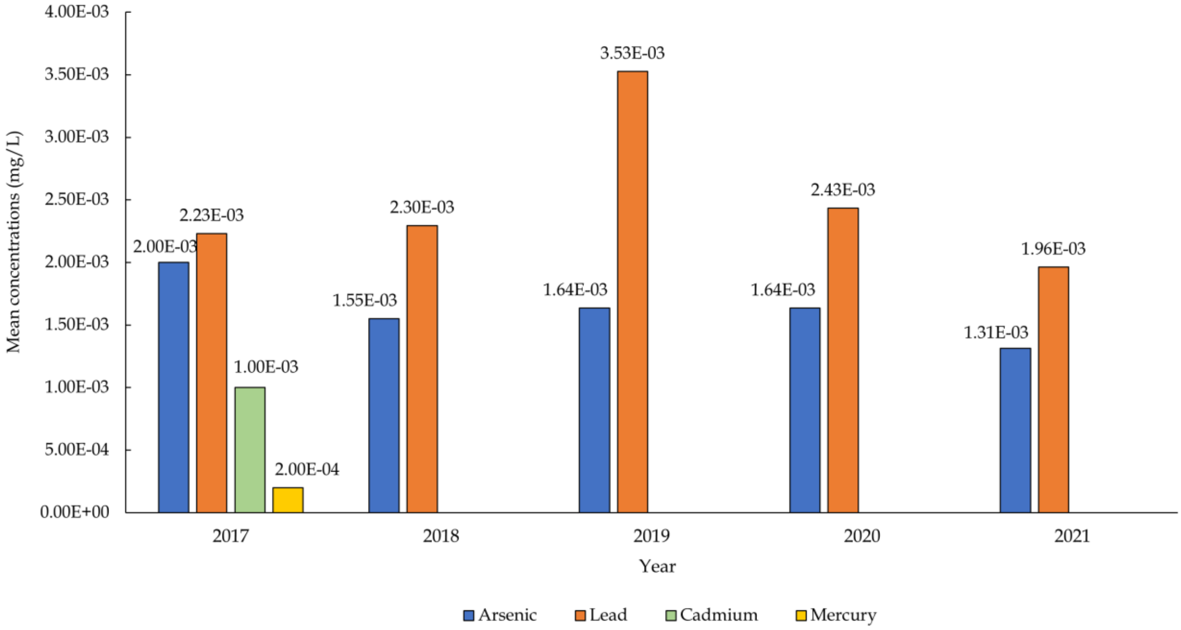
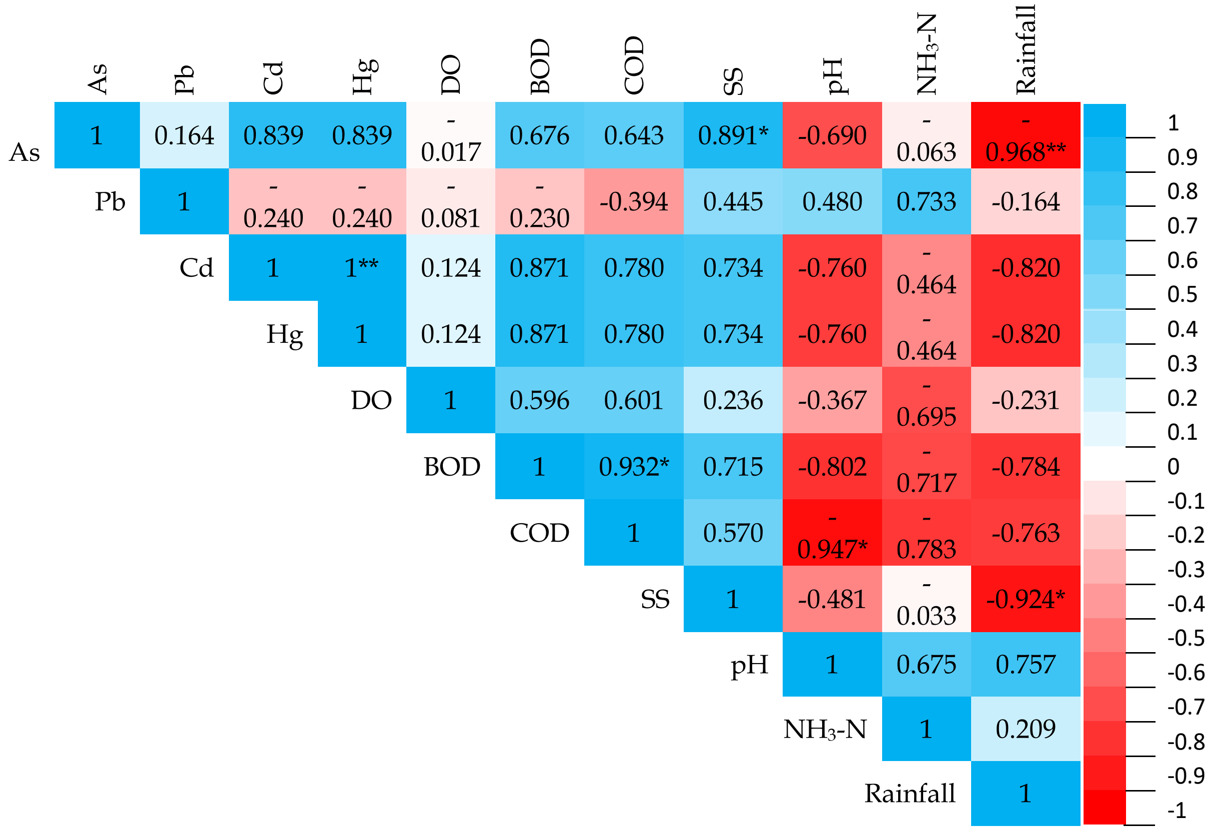


Figure 1 Mean concentrations of Arsenic (As), Lead (Pb), Cadmium (Cd) and Mercury (Hg) according to year from the Kuantan River

Further, the correlation between studied heavy metals, physicochemical properties and rainfall factors were observed. The influence of rainfall factor and suspended solid (SS) toward the elevation of As had statistically proved by the significant relationship between As and rainfall (p = 0.007) and between As and SS (p = 0.042) with both showing very high relationships (Figure 2). Rainwater has the ability to transport arsenic-bearing materials from an abandoned mine site into a river (Kusin et. al, 2018). Elevated levels of SS in a water body can lead to heavy metal precipitation within the SS, resulting in heavy metals settling as sediment, potentially causing adverse health effects, as discussed by Sujaul et al. (2013).



	As	Pb	Cd	Hg	DO	BOD	COD	SS	pH	NH_3-N	Rainfall
As	1	0.164	0.839	0.839	-0.017	0.676	0.643	0.891*	-0.690	-0.063	-0.968**
Pb		1	-0.240	-0.240	-0.081	-0.230	-0.394	0.445	0.480	0.733	-0.164
Cd			1	1**	0.124	0.871	0.780	0.734	-0.760	-0.464	-0.820
Hg				1	0.124	0.871	0.780	0.734	-0.760	-0.464	-0.820
DO					1	0.596	0.601	0.236	-0.367	-0.695	-0.231
BOD						1	0.932*	0.715	-0.802	-0.717	-0.784
COD							1	0.570	-0.947*	-0.783	-0.763
SS								1	-0.481	-0.033	-0.924*
pH									1	0.675	0.757
NH_3-N										1	0.209
Rainfall											1

* Correlation is significant at the 0.05 level (2-tailed) ** Correlation is significant at the 0.01 level (2-tailed)

Figure 2 Pearson correlation between studied heavy metals, physicochemical properties and rainfall factors (blue is positive and red is negative)

Meanwhile, the declining trend in heavy metals concentration of As and Pb in 2020 and 2021 can be associated with the implementation of the Movement Control Order (MCO) when the World Health Organization (WHO) declared the Coronavirus disease (COVID-19) outbreak as a global pandemic on 11th March 2020. Similarly, Malaysia has announced the COVID-19 lockdown starting on 18th March 2020 (Tang, 2022). According to Abdul Rahman & Abdul Halmi (2021), MCO has led to positive impacts on the environment due to restricted human movements, business or economic operations and anthropogenic activities. This is because anthropogenic activities contribute to the majority of water contamination.

Hazard Quotient

Figure 3 showed the results of HQ for both route of exposure, ingestion and dermal route of studied heavy metals. For ingestion route, the highest mean of HQ values of As and Pb were at 0.021 in 2017 and 0.023 in 2018, respectively. The mean HQ values were found to be at 0.004 for Hg and 0.002 for Cd in 2017. At 95th percentile, the highest HQ value of As was at 0.07 in 2017 and 0.08 in 2018, respectively. Hg and Cd possess a similar 95th percentile which is 0.01 in 2017.

Meanwhile, in 2017, the highest mean HQ for dermal uptake was found to be at 0.005 for As, while Pb reached its highest mean in 2019, also at 0.005. Estimated, Cd and Hg are in the third (0.020) and fourth-ranked (0.015) in terms of mean in 2017 respectively. The highest 95th percentile for As was reported at 0.02 in 2017 and Pb worst-case scenario' is at 0.02 in 2019. Both Cd and Hg showed similar 0.05 values which represents the 95th percentile in 2017. However, the mean and the 95th percentile for Cd and Hg were unable to be determined from 2018 - 2021 due to both heavy metals being below the detection limit (< 0.001 mg/L).

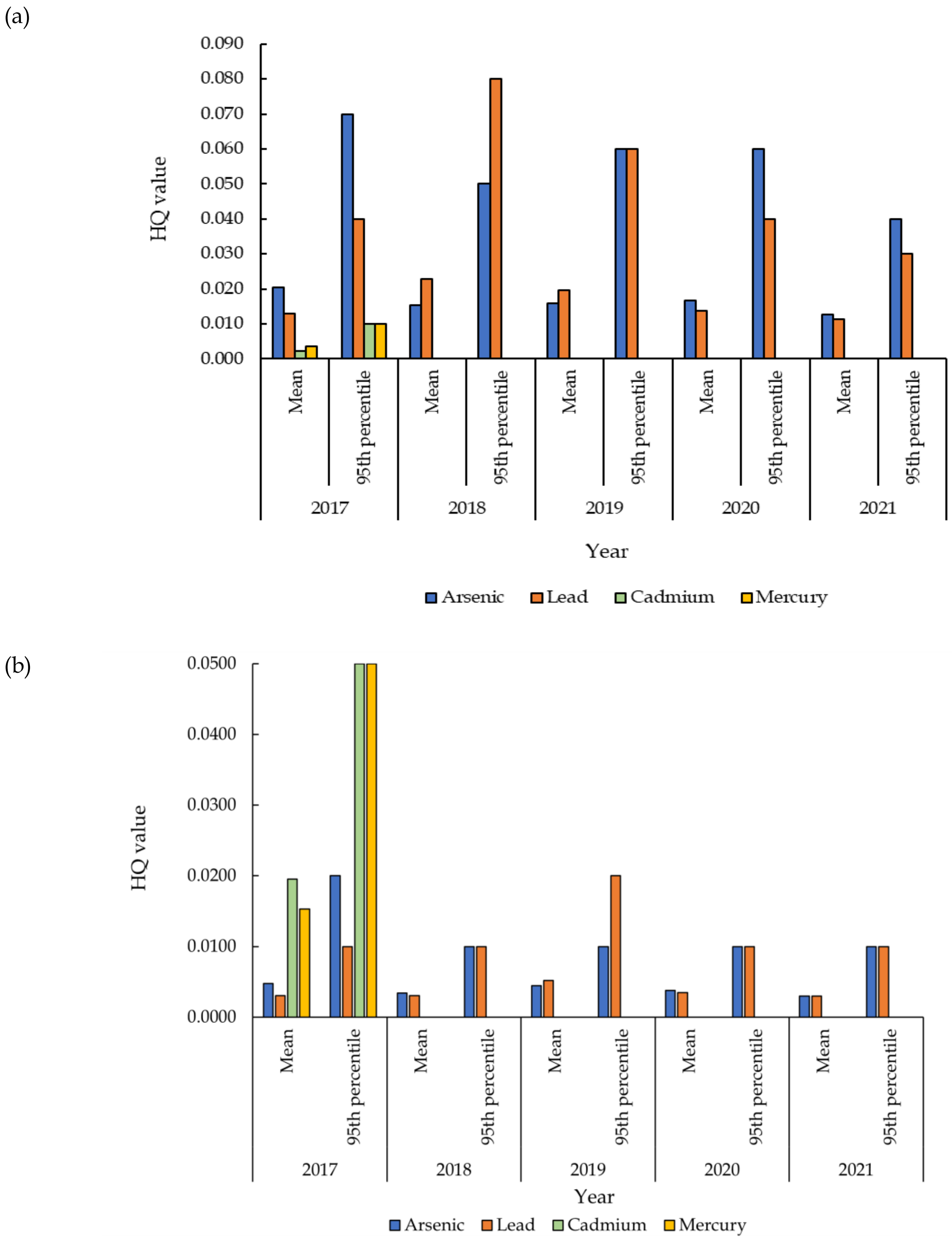


Figure 3 Result of Monte Carlo simulation of HQ via (a) ingestion and (b) dermal route according to year

Hazard Index

The cumulative impact of multiple heavy metals was assessed using the Hazard Index (HI). The mean HI values for both oral and dermal intake ranged from 0.006 to 0.043 while at the 95th percentile, they varied between 0.020 and 0.130. The cumulative HQ values for heavy metals calculated were consistently below one, indicating adverse health effects are not likely associated with exposure to multiple heavy metals.

Both HQ values and HI for ingestion and dermal routes are below the maximum acceptable value established by the US EPA guidelines which are less than one, representing an unlikely risk of adverse health effects of As, Pb, Cd, and Hg to the residents along the Kuantan River basin, whether through oral or dermal exposure. Similar findings were observed from the previous studies conducted in the Kuantan River by Mahad et al. (2019), through the consumption of marine fish in Peninsular Malaysia by Wan Azmi, Ahmad & Wan Mahiyuddin (2019), and in the Bertam River, Cameron Highlands by Razali et al. (2018).

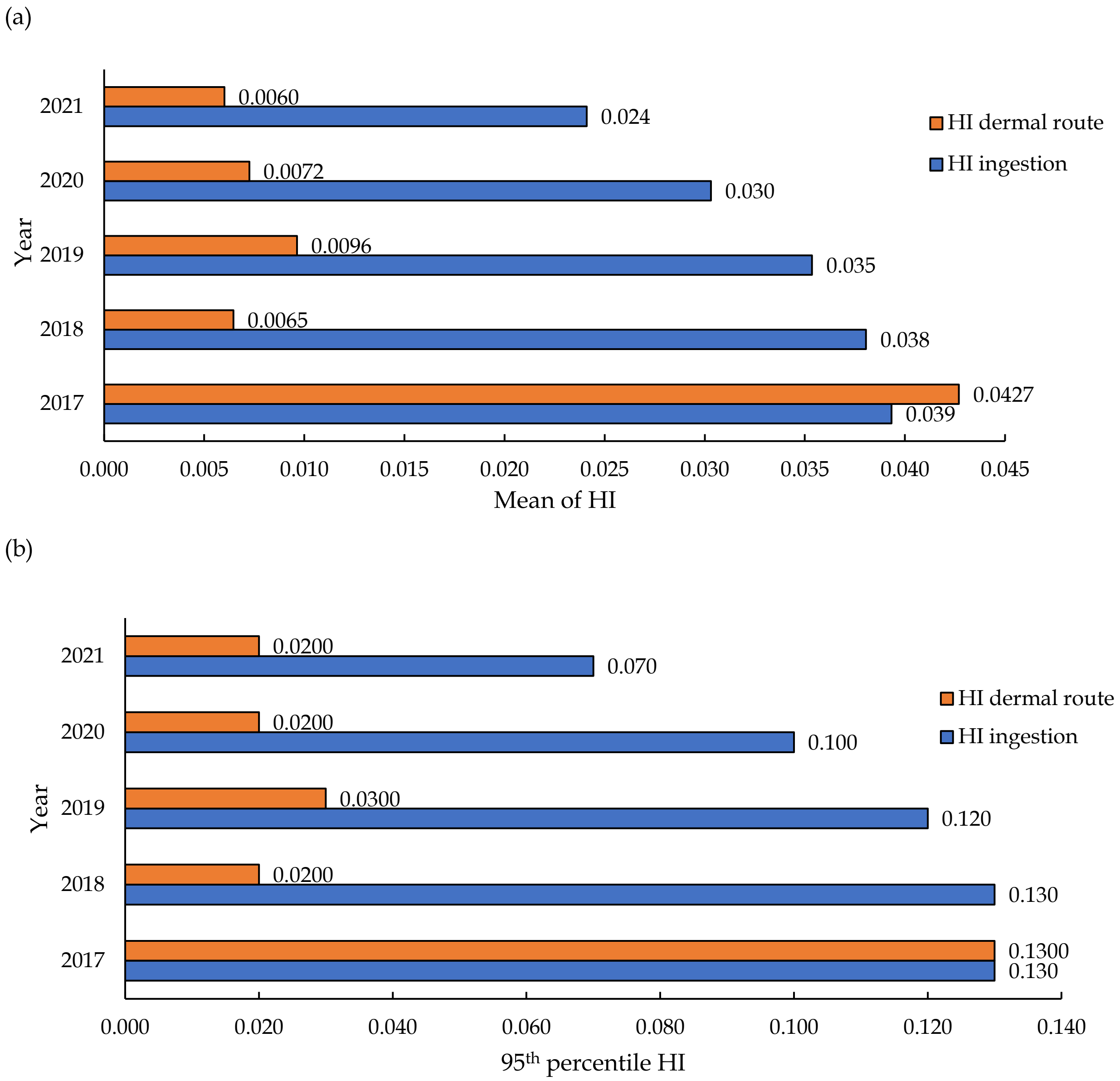


Figure 4 Results on (a) mean HI and (b) 95th percentile HI according to year

Lifetime Cancer Risk

LCR was determined only for As and Cd since both heavy metals are Group A or Class 1 human carcinogens (US EPA, 1998; IARC, 2012). Figure 5 demonstrates the mean LCR values via ingestion route across the five consecutive years were below the acceptable level of 1.00 x 10^{-4}, meanwhile, at the 95th percentile, the LCR values were beyond the acceptable level, which is represented by values of 2.98×10^{-4}, 2.75×10^{-4}, 1.97×10^{-4}, 1.99×10^{-4} and 1.48×10^{-4}, respectively. Cd recorded the mean and 95th percentile of LCR level at 2.97×10^{-6} and 5.64×10^{-6}, respectively which is below the value of 1 x 10^{-4}.

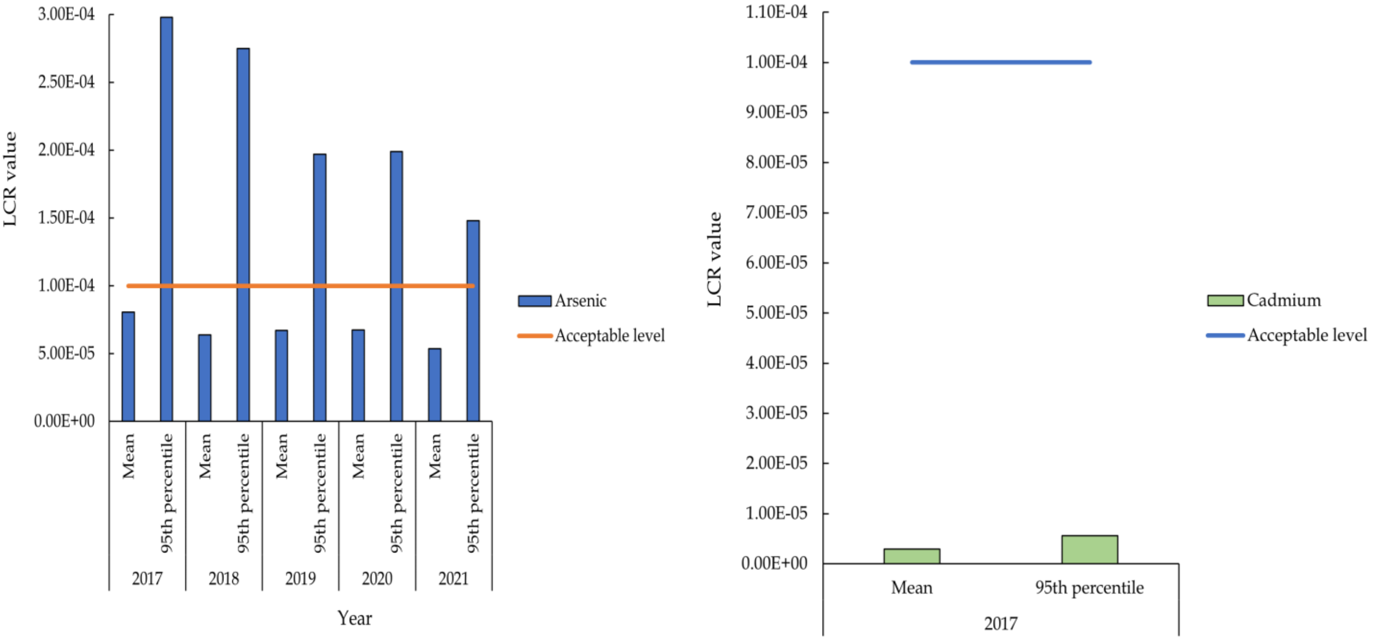


Figure 5 Result on Monte Carlo simulation of LCR via ingestion route according to year

The results of the mean of LCR due to dermal exposure to As and Cd are shown in Figure 6 below. The mean of LCR levels did not exceed the acceptable levels, however, among the five years, mean of LCR for As was highest at 3.72×10^{-5} in 2017 and lowest at 2.40×10^{-5} in 2019. Cd, on the other hand, recorded the reading of mean at 1.40×10^{-4} in 2017, which slightly exceeds the upper limit of 1×10^{-4}.

As reading for the 95th percentiles in all years shows that the readings exceeded the highest acceptable risk level of 1×10^{-4}, in which the values were 3.80×10^{-3}, 1.68×10^{-4}, $1.76 \text{ x } 10^{-4}$ 1.99×10^{-4}, and $1.11 \text{ x } 10^{-4}$, respectively. Similarly, Cd also reported the same result when Cd's 95th percentile (5.70×10^{-4}) surpassed the upper limit which is 1×10^{-4} in 2017. However, in the following years starting from 2018 to 2021, the estimation of the LCR value and 95th percentile can be achieved for As only due to the Cd concentration in the water being below the detection limit (< 0.001 mg/L).

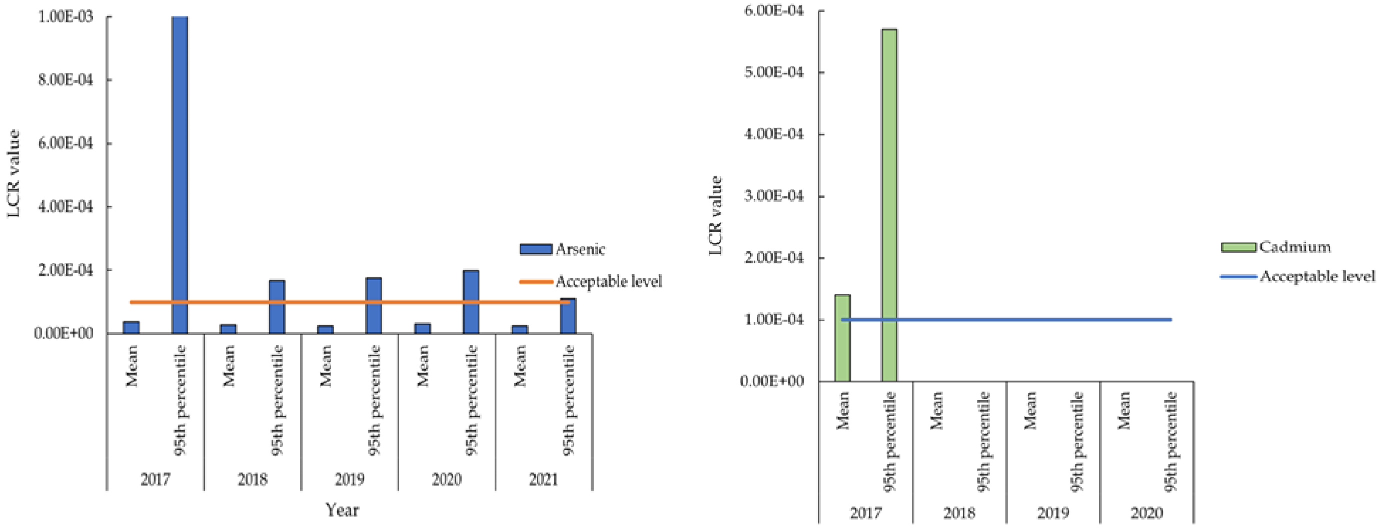


Figure 6 Result on Monte Carlo simulation of LCR via dermal route according to year

Carcinogenic risks, as defined by the EPA, refer to the increased likelihood of an individual developing cancer over their lifetime due to exposure to potential carcinogens (US EPA, 2023). The LCR values exceeding 1×10^{-4} are deemed unacceptable, as a carcinogenic risk at this level presents significant health hazards requiring intervention and remediation due to its substantial impact on health (US EPA, 2012). This recent study highlights that As and Cd pose substantial cancer risks, primarily through dermal uptake from polluted rivers and potential oral exposure. In essence, individuals exposed to As and Cd face an increased likelihood of developing cancer over their lifetime, either through dermal contact (such as bathing) or by ingesting untreated water, which may originate from groundwater sources.

Accumulation of ingested arsenic can have a variety of negative effects on humans, including an increased risk of cancer, high blood pressure, dermal effects, diabetes, and peripheral neuropathy (Ustaoglu & Aydin, 2020). Cadmium is soluble in water, whereas insoluble forms are immobile but can deposit into the water and form sediment (ATSDR, 2012). Therefore, high dermal absorption of Cd might represent a potential threat such as *'itai-itai'* disease, hypertension, gastrointestinal upset, cardiovascular disease, severe rheumatoid disease and fragility of bone (Mahad et al., 2019; ATSDR, 2012). Contaminated water with high As and Cd is required to undergo the treatment of either conventional or extensive treatment technologies to assure the safety of the water.

Sensitivity Analysis via Ingestion Route

Figure 7 shows in 2017, the factor with the greatest significance for sensitivity analysis was the concentration of heavy metals in the water, with As having a value of 1 and Cd having a value of 0.98. Likewise, the concentration of heavy metals was the greatest impact parameter for sensitivity analysis from 2018 to 2021. This is because all the readings for the concentration of heavy metals are 1. The other parameters such as IR, BW, AT, CSF, ABS_g, ED, EF and *RfD* were less sensitive and varied from -0.01 to 0.02 over the duration of the five years. However, due to the fact that the majority of the concentration values were below the detection limit, Cd does not display the results on sensitivity from 2018 to 2021.

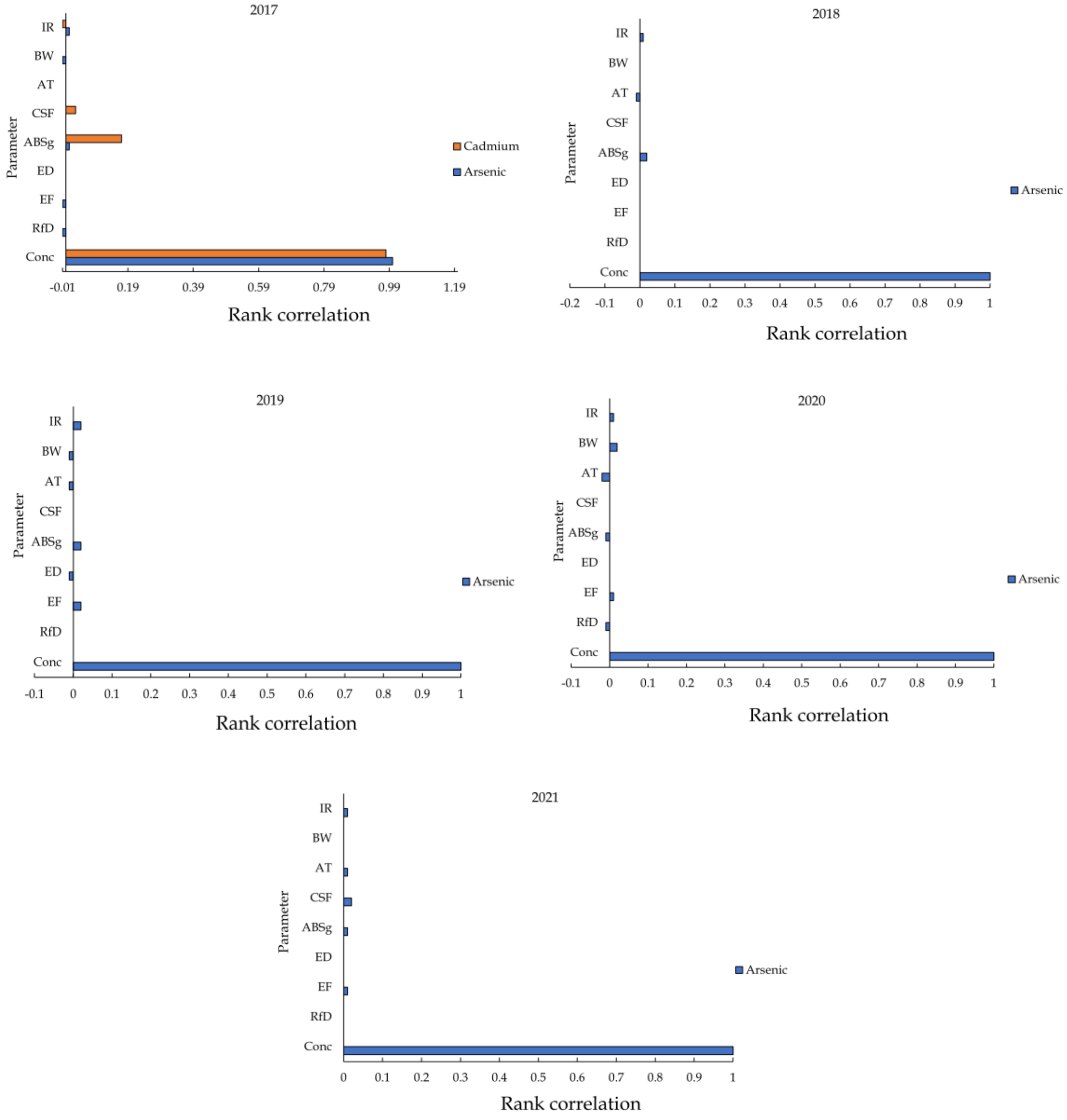


Figure 7 Sensitivity analysis result of As and Cd via ingestion route according to year

Sensitivity Analysis via Dermal Route

The sensitivity analysis results in Figure 8 indicate that the most significant parameter for arsenic (As) in 2017 was the concentration of heavy metals in water, with a value of 0.83. The parameter that significantly impacted Cd in 2017 was CF with a value of 0.56. The CF and the K_p both recorded the same value (0.56) in 2017 for As, CF is more significant than the K_p for Cd. CF parameter was analysed as a first ranked in 2018 to 2021 for As with values at 0.62, 0.70, 0.55, and 0.63, respectively. Meanwhile, the concentration value for Cd is below the detection limit (< 0.001 mg/L) during this period, hence the result of the sensitivity analysis was not estimated.

Sensitivity analysis was conducted to determine the most influential variable on the estimated risk. It is expected that the concentration of heavy metals in water is the most significant parameter reflected in the result of sensitivity analysis through the oral route. It reveals that toxic pollutants even at low concentrations can harm humans and deteriorate water quality. However, dermal uptake shows inverse results in which the CF had the highest impact except for As in 2017 which reported high heavy metal concentration as the most influential parameter.

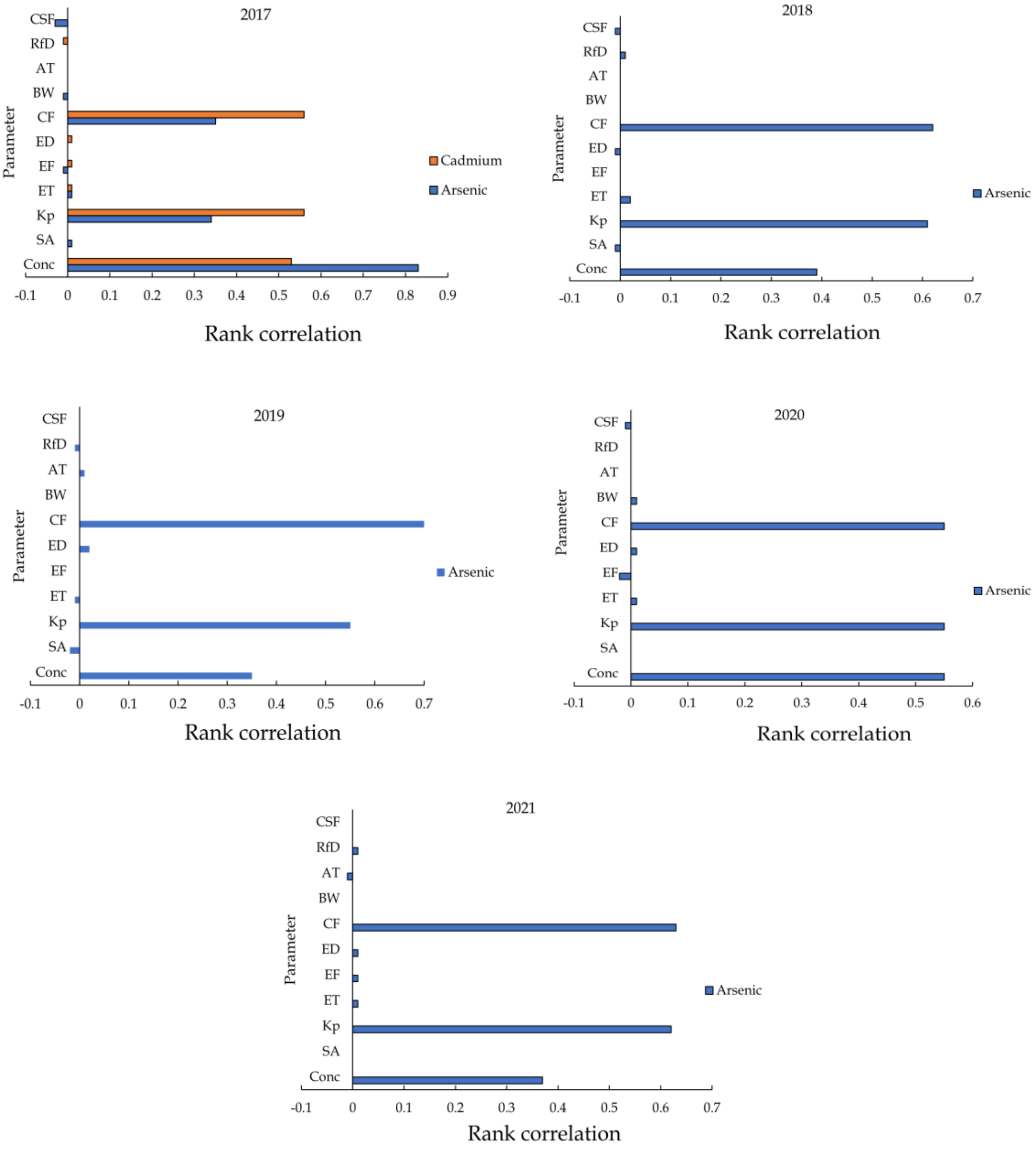


Figure 8 Sensitivity analysis result of As and Cd via dermal route according to year

CONCLUSION

LCR estimation indicated that Cd and As are the significant pollutants in the Kuantan River basin since they can increase the possibility of developing cancer in humans. These findings are crucial for both the environment and public health, highlighting the need for collaboration between the Department of Environment (DOE) and Ministry of Health (MOH) to mitigate heavy metal contamination. Strategic measures, increased monitoring, and public awareness campaigns are necessary to ensure water quality, especially for Kuantan's population, safeguarding their well-being and the aquatic ecosystem's sustainability.

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