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Review

Ecological–Health Risk Assessments of Copper in the Sediments: A Review and Synthesis

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Abstract: The ecological and children's Health Risk Assessments (HRA) of Copper (Cu) in aquatic bodies ranging from rivers, mangrove, estuaries, and offshore areas were studied using the Cited Cu Data in The Sediments (CCDITS) from 125 randomly selected papers published from 1980 to 2022. The ecological and children's HRA were assessed in all CCDITS. Generally, local point Cu sources (8%) and lithogenic sources were the main controlling factors of Cu concentrations. The present review revealed three interesting points. First, there were 11 papers (8%) documenting Cu levels of more than 500 mg/kg dw while China was the country with the highest number (26%) of papers published between 1980 and 2022, out of 37 countries. Second, with the Cu data cited from the literature not normally distributed, the maximum Cu level was higher than all the established guidelines. However, the median Cu concentration was lower than most of the established guidelines. The median values of the geoaccumulation index (I_{geo}) indicated a status of 'unpolluted' and 'moderate contamination' for the contamination factor (CF), and 'low potential ecological risk' for the ecological risk (ER) of Cu. However, the Cu ER could be based at present on the above mentioned 8% of the literature in the present study. Third, the calculated hazard index (HI) values were found to be below 1,

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indicating no potential chance of Cu non–carcinogenic effects in both adults and children, except for children's HI values from Lake Pamvotis of Greece, and Victoria Harbor in Hong Kong. Thus, regular monitoring (every 2 years), depending upon the available resources, is recommended to assess the ecological–health risk of Cu pollution in aquatic bodies to abate the risk of Cu exposure to children's health and avoid injurious impacts on the biota. It can be concluded that there is always a need for the mitigation and management of a Cu exposure risk assessment that can be used successfully for screening purposes to detect important human health exposure routes. Consequently, any sediments contaminated with Cu require rapid sediment remediation techniques.

Keywords: copper; sediments; geochemical indexes; health risk assessment

1. Introduction

There are five main reasons why copper (Cu) in sediments is focused on in this review. Firstly, based on global resources data in [1,2], Chile remains the dominant country with 658.2 Mt Cu and world copper resources were at least 1860 Mt Cu (including China). This is mainly caused by human mining for Cu due to the high demand for Cu used in manufacturing industries [1]. Currently, Cu and its compounds have been widely applied in various industries, including textiles, antifouling paints, electrical conductors, plumbing fixtures and pipes, cooking utensils, wood preservatives, pesticides fungicides, fertilizers, etc. [3].

Secondly, due to social-economic activities such as smelting, electroplating, leather production, electronics, agriculture (fertilisers and pesticides), and aquaculture sectors, Cu in sediments can be a secondary source of pollution in aquatic ecosystems. All of the aforementioned activities add to the stress on water and the environment by creating enormous amounts of municipal [4,5] and industrial wastewater [6,7] including potentially harmful compounds such as Cu.

Thirdly, Cu enters the marine environment mostly through rivers and estuaries, it is typically linked to particulate debris which settles and is absorbed into the sediment [8]. As a result, surface sediments constitute the major store and sink for metals and pollutants in aquatic ecosystems [8,9].

Fourthly, human expansion has prompted an increasing release to the environment due to rapid urbanization and industrialization which have led to significant increases in the levels, causing substantial pollution to the aquatic ecosystem. Therefore, it is important to address the role of anthropogenic activities on Cu pollution [8] in relation to many environmental media including sediments which end up as human health risks through different exposure pathways [10–13].

Fifthly, this topic has generated a lot of articles in the literature. According to the Scopus database, there were 674 publications published in scholarly journals between 1930 and May 2022 that featured the words 'copper' and 'sediments,' according to the Scopus database. If the names of the papers included words such as 'metals' and 'elements,' the number would be substantially higher. If non-Scopus indexed journals were taken into consideration, the number would likely be substantially greater. These figures demonstrate the need to keep an eye on Cu levels in aquatic sediments. It is realistic to expect that monitoring studies will become more common in the future.

Sediment refers to a layer of solid particles on the bed of a water body, which consists of any insoluble particulate matter [14]. These particulate matters could be transported from one area to others by various means, for instance, wind, and flowing rivers. Throughout the fate of a sediment particle, there might be a temporary settlement in between its origin and its final resting place. These sediments may also become settled in a delta at the river mouth or become beach deposits by the action of tides, currents, and waves. Coastal sediments are a major sink for metals of both anthropogenic and natural origin [15]. Under

certain conditions, these accumulated metals in sediment may be remobilized, changing the surrounding aquatic ecosystem [16].

Both direct and indirect pathways are likely to play a role in the entry of sediment-bound metals into the human body [17]. Therefore, heavy metal pollution in coastal ecosystem is a serious concern. Despite these concerns, there are few studies that have focused on or have investigated the impact of sediment-bound metals on human health directly. The Cu bound to beach sand particles could enter the human body via inhalation of the sand or sedimentary particles, and direct ingestion via hand-to-mouth action especially by children [18]. Therefore, this implies that humans, especially children playing on beaches, may be exposed to these metal contaminants settled in the sand.

The Hazard Index (HI), Hazard Quotient (HQ), or Target Hazard Quotient (THQ) are used to measure the health risk posed by toxic metals. The HI value reported in urban park soils [18] and kindergartens soils [19] was noted to be comparatively higher in children than adults. Therefore, the risks posed by the ingestion route of sedimentary particles by both children and adults were the highest, followed by dermal contact/inhalation. Therefore, the health risk assessments (HRA) for children should be given a higher priority.

The objectives of this paper are to (a) review Cu concentrations in the sediments from 125 publications from 1980 to 2022 based on Scopus and Google Scholar databases and, (b) make commentary based on the reassessment of ecological–health risks of Cu from 125 articles on adults and children's HRA based on the reported Cu concentrations in the sediments.

2. Materials and Methods

2.1. Data Collection

Cu data reported in Scopus and Google Scholar databases ranging from 1980 to April 2022 were used. A total of 125 publications were randomly selected, with a special focus on different regions and countries. The keywords for the search were 'copper' and 'sediment' which were mostly found in the titles of the articles. However, only those papers that reported the Cu ranges (minimum and maximum concentrations) were selected in the present study for easy and direct comparative purposes, and the standardization of the calculation of the ecological–health risks of Cu.

The current review used Moher et al. [20]'s Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) systematic literature review approach to contribute to the existing body of information on 'copper' and 'sediment.' PRISMA is an evidence-based reporting standard that can be used for critical evaluation. Figure 1 shows the metrics of the formal method that were updated for this review work.

2.2. Data Treatment

2.2.1. Geoaccumulation Index

The geoaccumulation index (I_{geo}) has been shown to be a useful tool for assessing sedimentary heavy metal pollution [21]. The degree of Cu contamination in the area was determined using the geoaccumulation index (I_{geo}). The calculation of I_{geo} was based on Equation (1) [21].

$$I_{geo} = \log_2\left(\frac{\text{Sample}}{1.5 \times \text{Bg}}\right) \quad (1)$$

where sample is the total Cu concentrations in the sediments and Bg is the geochemical background value for Cu. The present study used the background concentrations in the earth's upper continental crust (UCC) which were Cu (14.3 mg/kg) based on Wedepohl [22].

The value (1.5) was the correction factor to mitigate the lithogenic effluents. There are six established classifications of pollution: 'practically unpolluted' (<0), 'unpolluted' (0–1), 'moderately polluted' (1–2), 'moderately polluted to strongly polluted' (2–3), 'strongly polluted' (3–4), 'strongly to very strongly polluted' (4–5), and 'very strongly polluted' (>5) [21].

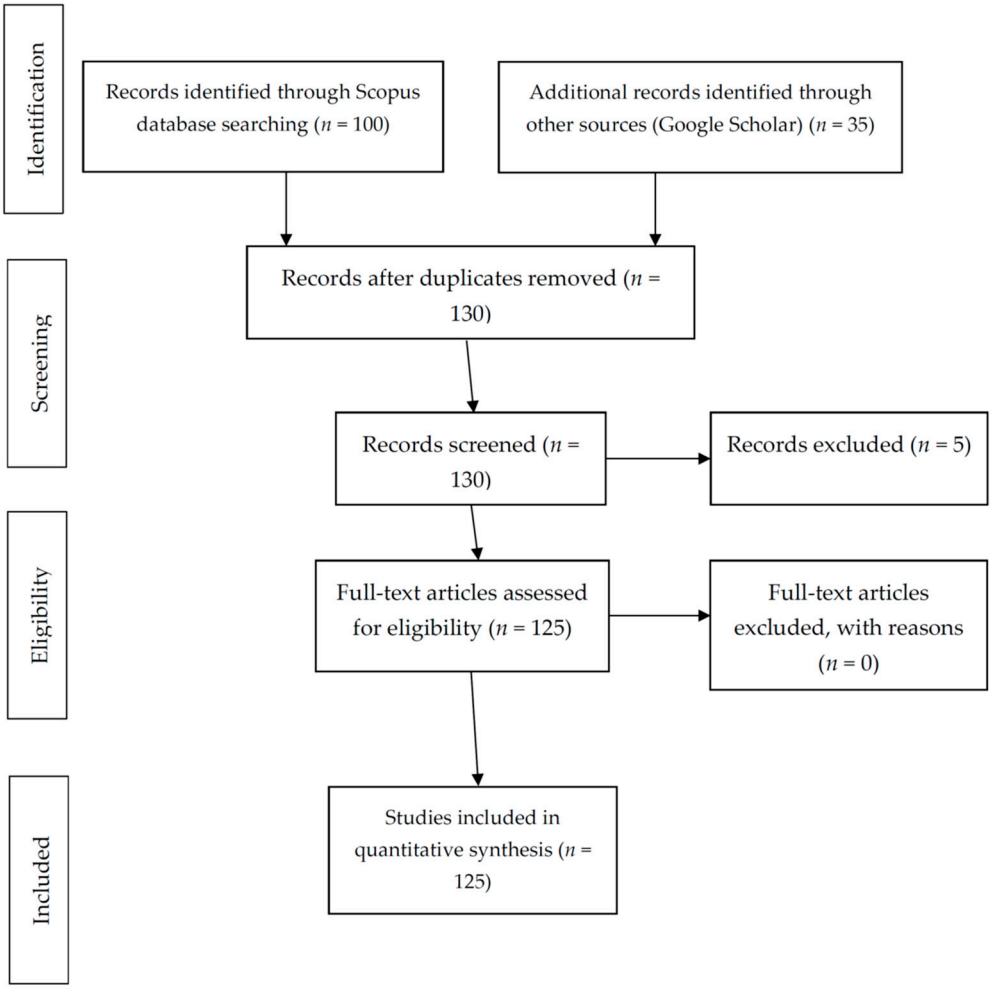


Figure 1. Flowchart of the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) (adapted from Moher et al. [20]), used in the present study.

2.2.2. Ecological Risk Index

Firstly, the calculation of the contamination factor (CF) was based on the pollution of a single Cu factor in Equation (2).

$$CF = \frac{C_S}{C_B} \quad (2)$$

where CF is the contamination factor; C_s is the mean Cu concentration in the sediment; and C_B is the Cu background level by based on Wedepohl [22], as mentioned previously. According to Hakanson [23], 4 classifications for CF values are 'low contamination' (CF< 1), 'moderate contamination' ($1 \leq CF < 3$), 'considerable contamination' ($3 \leq CF < 6$), and 'very high contamination' ($CF \geq 6$).

Later, the calculation of the ecological risk (ER), which is the potential ecological risk of a single element, was calculated based on Equation (3).

$$ER = T_R \times CF \quad (3)$$

The Cu toxic response factor (T_R) value used in the present study was 5.00 [22]. According to Hakanson [23], 5 classifications for the ER are 'low potential ecological risk' ($ER < 40$), 'moderate potential ecological risk' ($40 \leq ER < 80$), 'considerable potential ecological risk' ($80 \leq ER < 160$), 'high potential ecological risk' ($160 \leq ER < 320$), and 'very high ecological risk' ($ER \geq 320$).

2.2.3. Human Health Risk Assessment

Human health risk assessment (HHRA) of sediments is commonly used to assess both carcinogenic and non-carcinogenic risks to people through three exposure pathways: ingestion, inhalation, and skin contact. The HHRA technique was based on the US Environmental Protection Agency's guidelines and Exposure Factors Handbook [24–27]. The average daily doses (ADDs) (mg/kg day) of Cu through ingestion (ADD_{ing}), inhalation (ADD_{inh}), and dermal contact (ADD_{der}) for both children and adults were calculated by using Equations (4)–(6) as follows:

$$ADD_{ing} = C_{sediment}\left(\frac{IngR \times EF \times ED}{BW \times AT}\right) \times 10^{-6} \quad (4)$$

$$ADD_{inh} = C_{sediment}\left(\frac{IngR \times EF \times ED}{PEF \times BW \times AT}\right) \quad (5)$$

$$ADD_{der} = C_{sediment}\left(\frac{SA \times AF \times ABS \times EF \times ED}{BW \times AT}\right) \times 10^{-6} \quad (6)$$

where all the abbreviations are given and explained in Table 1. The daily levels of exposure to metals (mg/kg day) by ingestion, inhalation, and dermal contact are represented by ADD_{ing}, ADD_{inh}, and ADD_{der}, respectively. The hazard quotient (HQ) and hazard index (HI) were used to calculate the non-carcinogenic risk (NCR) of Cu in this investigation [24,25]. Table 1 shows the definition, exposure parameters, and reference values used in the literature to determine the intake values and health hazards of Cu in sediments.

Table 1. Definition, exposure factors, and reference values used to estimate the intake values and health risks of potentially toxic metals in sediment for the present study.

Factor	Definition	Unit	Values		References
			Children	Adults	
IngR	Ingestion rate of sediment	mg/day	200	100	[24]
ED	Exposure duration	Years	6.0	24	[24]
PEF	Particle emission factor	m^3/kg	1.36×10^9	1.36×10^9	[24]
AT	Average time	Days	365 × ED	365 × ED	[25]
BW	Bodyweight of the exposed individual	kg	15	55.9	[28]
EF	Exposure frequency	days/year	365	365	-
SA	Exposed skin surface area	cm^2	1600	4350	[28]
AF	Skin adherence factor	mg/cm day	0.20	0.70	[29]
Cu RfD_{ing}	Reference dose for ingestion	mg/kg day	4.00×10^{-2}	4.00×10^{-2}	[30]
Cu RfD_{inh}	Reference dose for inhalation	mg/kg day	4.02×10^{-2}	4.02×10^{-2}	[30]
Cu RfD_{der}	Reference dose for dermal contact	mg/kg day	1.20×10^{-2}	1.20×10^{-2}	[30]
InhR	Inhalation rate of sediment	m^3/day	7.63	12.8	[31]
ABF	Dermal absorption factor	Unitless	1.00×10^{-3}	1.00×10^{-3}	[32]

The HQ is the ratio of a metal's ADD to its reference dose (RfD) for exposure pathways that are similar [26]. The RfD (mg/kg day) is the maximum daily dosage of metal from a certain exposure pathway that is considered not to pose a significant risk of detrimental consequences to sensitive individuals over their lifetime, including both children and adults. Table 1 shows the RfD (mg/kg day) values of Cu used in this study for ingestion, inhalation, and dermal contact. If the ADD is less than the RfD value (HQ < 1), no adverse health effects are expected, but if the ADD exceeds the RfD value (HQ > 1), there are likely to be detrimental health effects [24,26].

HI, which is the total of the HQs in the three exposure paths, is used to calculate the NCR [33–35]. A HI value less than 1.0 indicates that there was no considerable risk of non-carcinogenic consequences. A HI of greater than 1.0 indicates the possibility of

non-carcinogenic effects. Non-carcinogenic effects are likely to have a favorable relationship with the increase in the HI value [31]. The HI was calculated according to Equation (7).

$$HI = \sum HQ_i = \sum \left(\frac{ADD_i}{RfD_i} \right) \quad (7)$$

Data Analysis

All graphical bar charts were plotted using the KaleidaGraph (Version 3.08, Sygnergy Software, Eden Prairie, MN, USA). The overall statistics were also obtained from KaleidaGraph.

3. Results and Discussion

3.1. Ecological Risk Assessments

Figure 2 shows the world map of Cu levels in the sediments cited in the 125 publications [36–160]. The detailed Cu concentrations (mg/kg, dw) in the sediments reported in the 125 papers are provided in Tables S1 and S2.

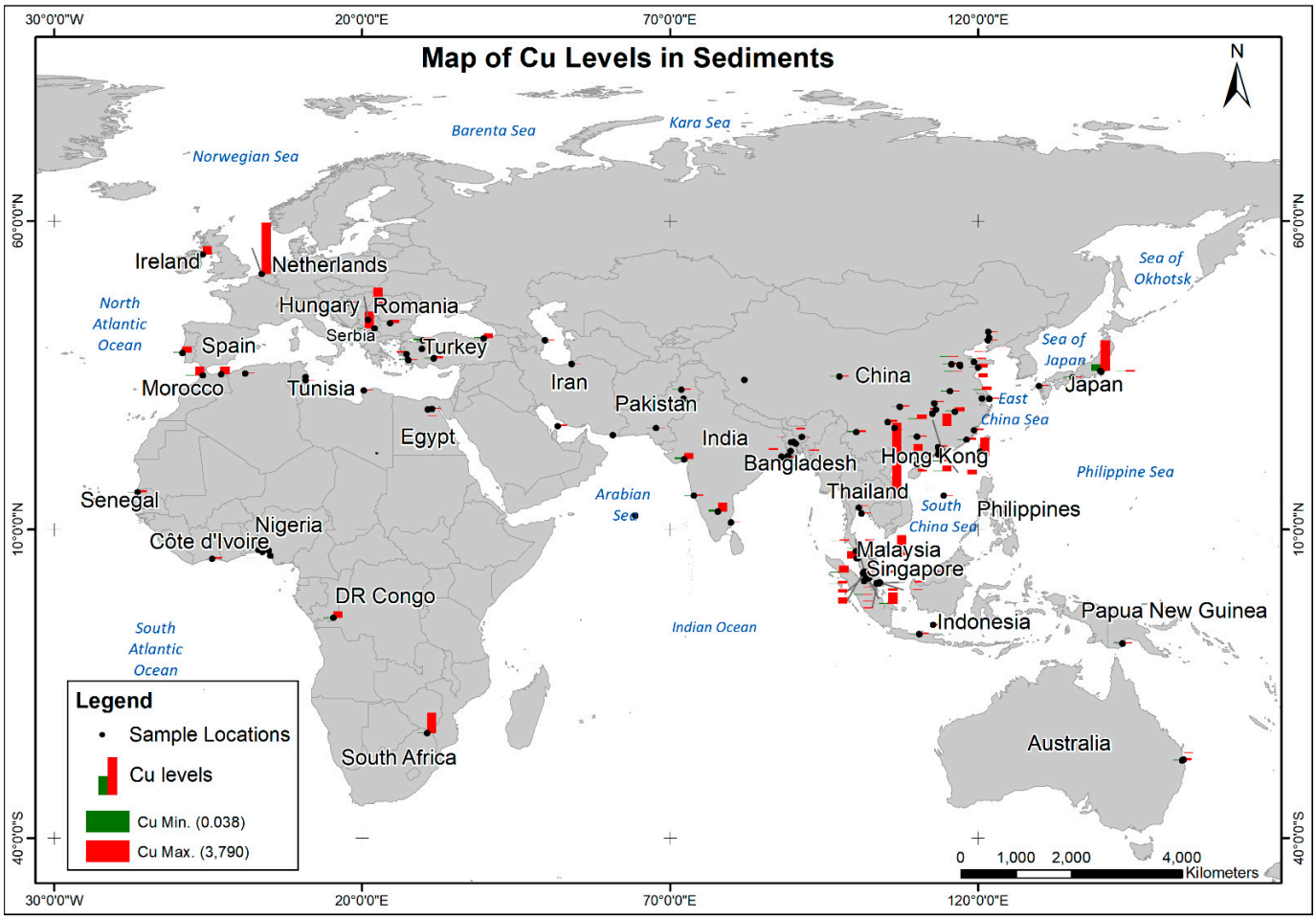


Figure 2. A world map covering the continents of Africa and Europe and the Asia–Pacific areas for the Cu levels in the sediments cited in the 125 publications. Source: https://www.efrainmaps.es/english-version/free-downloads/world/ (assessed on 1 June 2022).

A total of 37 countries (Table 2; Table S1) were coincidentally documented with their respective Cu concentrations in the sediments, mostly reporting the Cu concentrations in combination with other heavy or trace metals in the sediments from the coastal areas, mangrove, rivers, lakes, or estuaries. Clearly, many studies of Cu levels were reported in Asian countries, and there were scattered studies in Europe and African countries. Among the Asian countries, China topped the list with 33 papers, followed by Malaysia (19), India and Tunisia (7), Bangladesh (6), Japan (4), Hong Kong (3), Singapore (3), Indonesia (3), Iran (3), Pakistan (3), 8 other countries with two papers, and 18 other countries with one paper [36–160].

Table 2. Overall number of papers (NP) reported in different countries out of the 125 papers cited in the present study.

No.	Country	NP	No.	Country	NP
1	China	33	20	Netherlands	1
2	Malaysia	19	21	Ghana	1
3	India	7	22	Greece	1
4	Tunisia	7	23	Hungary	1
5	Bangladesh	6	24	Ivory Coast	1
6	Japan	4	25	Libya	1
7	Hong Kong	3	26	Morocco	1
8	Indonesia	3	27	Netherlands	1
9	Singapore	3	28	Nigeria	1
10	Iran	3	29	Oman	1
11	Pakistan	3	30	Papua New Guinea	1
12	Algeria	2	31	Philippines	1
13	Australia	2	32	Senegal	1
14	Taiwan	2	33	USA	1
15	Thailand	2	34	South Africa	1
16	Nigeria	2	35	Oman	1
17	Turkey	2	36	Spain	1
18	Egypt	2	37	Romania	1
19	Serbia	2			

Based on the 125 reviewed papers, there were 11 papers (8%) documenting the total Cu concentrations of more than 500 mg/kg dw (Tables S1 and S2; Figure 3). These papers included Lake Pamvotis of Greece (24985 mg/kg; [92])> Victoria Harbor of Hong Kong (3790 mg/kg; [46]) > Scheldt Estuarine of the Netherlands (2600 mg/kg; [47]), Old Nakagawa River of Tokyo, Japan (1565 mg/kg; [76]) > Mvudi River of South Africa (1027 mg/kg; [66]) > polluted drainage sediments from Peninsular Malaysia (1019 mg/kg; [72]) > Kaohsiung Harbor in Taiwan (946 mg/kg; [68]) > Serbia (870 mg/kg [98], 859.9 mg/kg; [143]) > Kaohsiung Harbor in Taiwan (760 mg/kg; [108]) > Shima River of China (630 mg/kg; [110]). From these top eleven citations with elevated Cu levels in the sediments, the levels in the Kaohsiung Harbor of Taiwan were reported to be twice as high, between 760–946 mg/kg, by Chen et al. [68] and Chen et al. [108]. However, based on HQ, only Lake Pamvotis of Greece (24,985 mg/kg; [92]) was found to have a HQ value over 1, indicating a potential chance of Cu NCR at this site.

Table 3 shows the comparisons of the concentrations of Cu sediment quality guidelines in the available literature, with the overall statistics of the 125 reviewed papers containing Cu data in the present study. There are three patterns that can be observed.

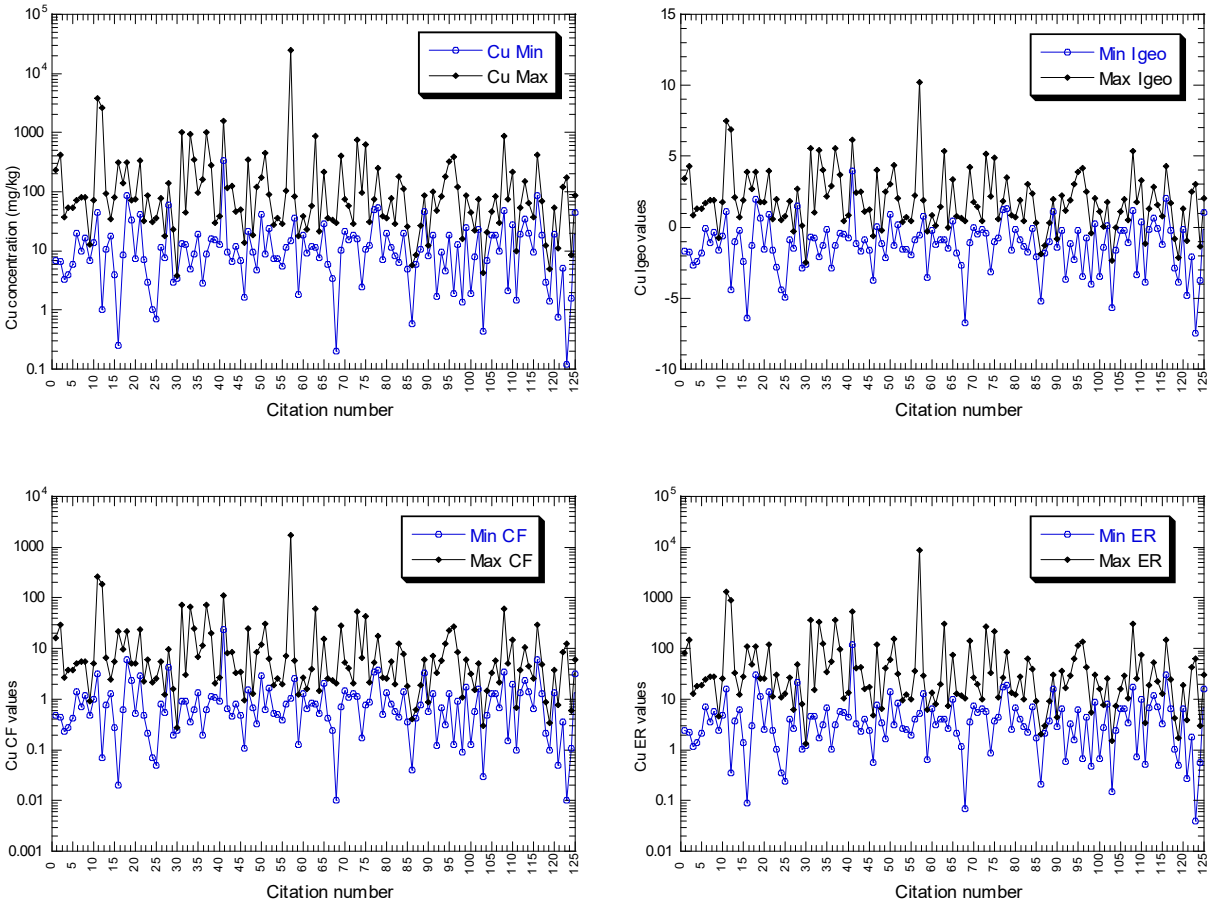


Figure 3. The calculated values of the geoaccumulation index (I_{geo}), contamination factor (CF), and ecological risk (ER), based on the Cu concentration ranges (mg/kg dry weight) (minimum and maximum) in the sediments cited from 125 papers in the literature published between 1980 and 2022. The citation numbers follow the detailed reference numbers presented in Tables S1 and S2.

Firstly, the average values of skewness and kurtosis were 14.8 and 224, respectively. This indicates that the Cu data cited from the literature were not normally distributed in which the Cu data were not within the normality ranges for skewness (−2 to +2) [161–163] and kurtosis (−7 to +7) [161,162]. Therefore, we used medians, rather than means, for a more meaningful interpretation with the understanding of outliers which were represented by the extremely elevated Cu data in 8% (11 papers as mentioned previously) of the data cited. Secondly, based on the 125 papers, the Cu concentrations (mg/kg dry weight) ranged from 0.12 to 24,985. The maximum Cu level was higher than all the established guidelines in Table 3. However, the median Cu concentration (22.95) was lower than all the established guidelines [22,23,164–168], except for the threshold effect level (TEL) [168] and upper continental crust (UCC) [22].

Thirdly, the I_{geo} values ranged from −7.48 to 10.19. This indicates that they ranged from 'practically unpolluted' to 'very strongly polluted' with a median (0.09) status of 'unpolluted' based on Muller [21]'s classifications. The CF values ranged from 0.01 to 1747. This indicates that they ranged from 'low contamination' to 'very high contamination' with a median (1.61) status of 'moderate contamination' according to Hakanson [23]. The ER values ranged from 0.04 to 8736. This means that they ranged from a 'low potential ecological risk' to a 'very high ecological risk' with a median (8.02) status of 'low potential ecological risk' according to Hakanson [23].

Table 3. Comparison of the overall statistics of the Cu calculated values of the geoaccumulation index (I_{geo}), contamination factor (CF), and ecological risk (ER), hazard quotient ingestion (HQ_{ing}), hazard quotient inhalation (HQ_{inh}), hazard quotient dermal contact (HQ_{der}), hazard index (HI) for adults (A) and children (C) based on the Cu concentrations ranges (mg/kg dry weight) (minimum and maximum) in the sediments cited from 125 papers in the literature published between 1980 and 2022 with the different established guidelines values.

	Cu	**I_{geo}**	**CF**	**ER**	**A HQ_{ing}**	**A HQ_{inh}**	**A HQ_{der}**	**A HI**	**C HQ_{ing}**	**C HQ_{inh}**	**C HQ_{der}**	**C HI**	**Reference**
Minimum	0.12	−7.48	0.01	0.04	5.50×10^{-6}	5.03×10^{-10}	5.58×10^{-7}	6.06×10^{-6}	4.10×10^{-5}	1.12×10^{-9}	2.19×10^{-7}	4.12×10^{-5}	This study
Maximum	24,985	10.19	1747	8736	1.15	1.05×10^{-4}	1.16×10^{-1}	1.26	8.54	2.32×10^{-4}	4.55×10^{-2}	8.58	
Mean	209	0.25	14.65	73.23	9.62×10^{-3}	8.79×10^{-7}	9.73×10^{-4}	1.06×10^{-2}	7.16×10^{-2}	1.95×10^{-6}	3.82×10^{-4}	7.19×10^{-2}	
Median	22.95	0.09	1.61	8.02	1.05×10^{-3}	9.62×10^{-8}	1.07×10^{-4}	1.16×10^{-3}	7.85×10^{-3}	2.14×10^{-7}	4.18×10^{-5}	$7.88E \times 10^{-3}$	
SD	1609	2.52	113	563	7.41×10^{-2}	6.76×10^{-6}	7.47×10^{-3}	8.12×10^{-2}	5.50×10^{-1}	1.49×10^{-5}	2.93×10^{-3}	5.53×10^{-1}	
SE	102	0.16	7.12	35.6	4.68×10^{-3}	4.28×10^{-7}	4.73×10^{-4}	5.13×10^{-3}	3.48×10^{-2}	9.45×10^{-7}	1.85×10^{-4}	3.50×10^{-2}	
Skewness	14.8	0.21	14.8	14.8	1.48×10	1.47×10	1.47×10	1.47×10	1.47×10	1.47×10	1.47×10	1.47×10	
Kurtosis	224	1.06	224	224	2.24×10^{2}	2.24×10^{2}	2.24×10^{2}	2.24×10^{2}	2.24×10^{2}	2.24×10^{2}	2.24×10^{2}	2.24×10^{2}	
Guidelines	Cu	I_{geo}	CF	ER	A HQ_{ing}	A HQ_{inh}	A HQ_{der}	A HI	C HQ_{ing}	C HQ_{inh}	C HQ_{der}	C HI	Reference
ERL	34.0	0.66	2.38	11.89	1.56×10^{-3}	1.42×10^{-7}	1.58×10^{-4}	1.72×10^{-3}	1.16×10^{-2}	3.16×10^{-7}	6.20×10^{-5}	1.17×10^{-2}	[166]
ERM	270	3.65	18.88	94.41	1.24×10^{-2}	1.13×10^{-6}	1.26×10^{-3}	1.36×10^{-2}	9.23×10^{-2}	2.51×10^{-6}	4.92×10^{-4}	9.28×10^{-2}	[166]
ISQV-low	65.0	1.60	4.55	22.73	2.98×10^{-3}	2.72×10^{-7}	3.03×10^{-4}	3.28×10^{-3}	2.22×10^{-2}	6.05×10^{-7}	1.18×10^{-4}	2.23×10^{-2}	[167]
ISQV-high	270	3.65	18.88	94.41	1.24×10^{-2}	1.13×10^{-6}	1.26×10^{-3}	1.36×10^{-2}	9.23×10^{-2}	2.51×10^{-6}	4.92×10^{-4}	9.28×10^{-2}	[167]
TEL	18.7	−0.20	1.31	6.54	8.57×10^{-4}	7.83×10^{-8}	8.70×10^{-5}	9.45×10^{-4}	6.39×10^{-3}	1.74×10^{-7}	3.41×10^{-5}	6.43×10^{-3}	[168]
PEL	108.2	2.33	7.57	37.83	4.96×10^{-3}	4.53×10^{-7}	5.04×10^{-4}	5.47×10^{-3}	3.70×10^{-2}	1.01×10^{-6}	1.97×10^{-4}	3.72×10^{-2}	[168]
PRL	50.0	1.22	3.50	17.48	2.29×10^{-3}	2.09×10^{-7}	2.33×10^{-4}	2.53×10^{-3}	1.71×10^{-2}	4.65×10^{-7}	9.11×10^{-5}	1.72×10^{-2}	[23]
UCC	25.0	0.22	1.75	8.74	1.15×10^{-3}	1.05×10^{-7}	1.16×10^{-4}	1.26×10^{-3}	8.54×10^{-3}	2.33×10^{-7}	4.56×10^{-5}	8.59×10^{-3}	[164]
UCC	14.3	−0.58	1.00	5.00	6.56×10^{-4}	5.99×10^{-8}	6.66×10^{-5}	7.22×10^{-4}	4.89×10^{-3}	1.33×10^{-7}	2.61×10^{-5}	4.91×10^{-3}	[22]
UCC	28.0	0.38	1.96	9.79	1.28×10^{-3}	1.17×10^{-7}	1.30×10^{-4}	1.41×10^{-3}	9.57×10^{-3}	2.61×10^{-7}	5.10×10^{-5}	9.62×10^{-3}	[165]

Note: UCC = Upper continental crust; ERL = Effects range low; ERM = Effects range median; ISQV-low = Interim sediment quality value-low; ISQV-high = Interim sediment quality value-high; TEL = Threshold effect level; PEL = Probable effect level; PRL = Pre-industrial reference level. SD = standard deviation; SE = standard error.

3.2. Health Risk Assessments

Figure 4 shows the values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI for adults and children based on the Cu data ranges (minimum, Table S1 and maximum, Table S2) in the sediments cited from 125 papers in the literature published between 1980 and 2022. The overall statistics of the above values are provided in Table 3.

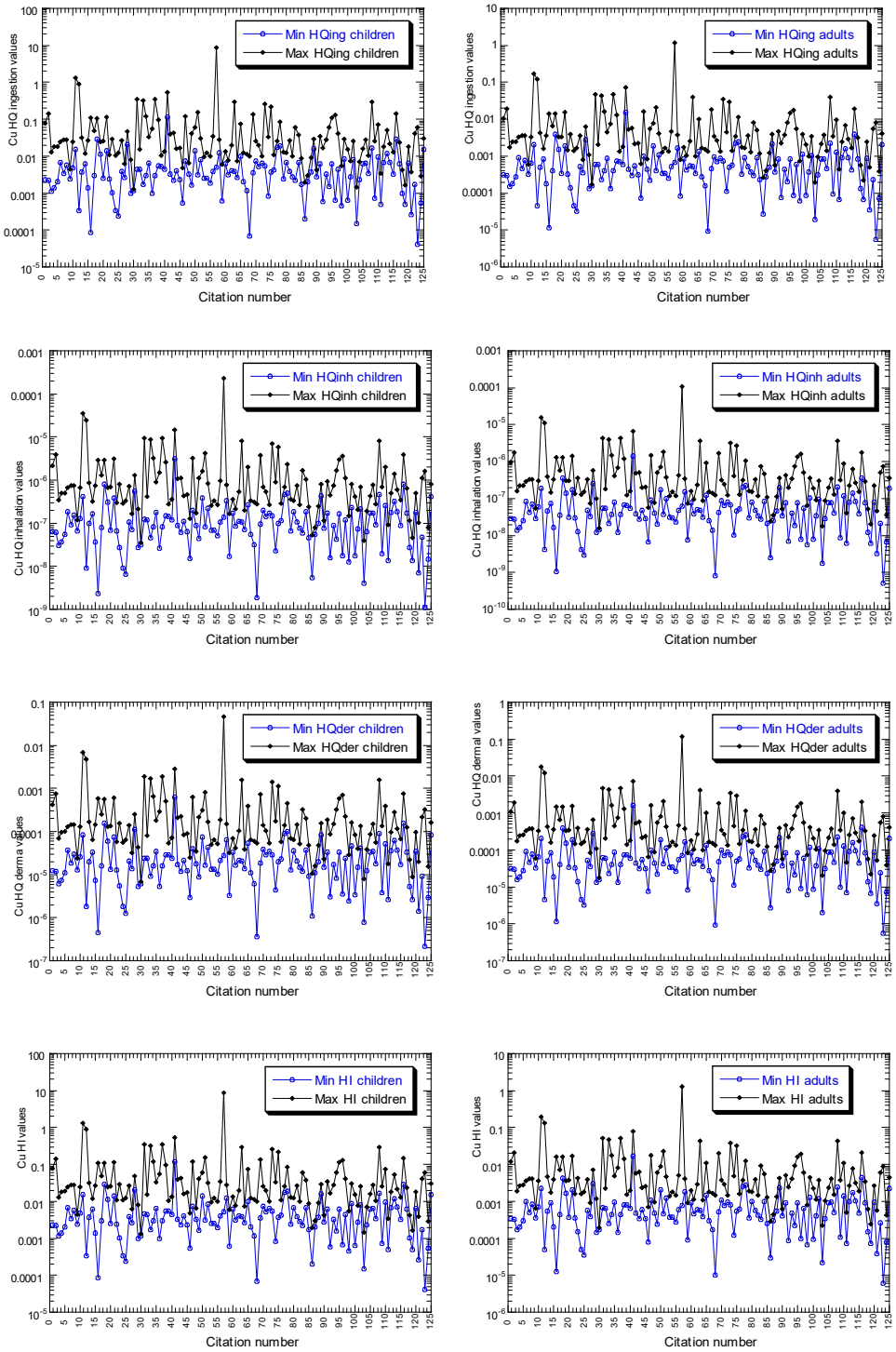


Figure 4. The values of hazard quotient ingestion (HQ_{ing}), hazard quotient inhalation (HQ_{inh}), hazard quotient dermal contact (HQ_{der}), and hazard index (HI) for adults and children based on the Cu data ranges (minimum and maximum) in the sediments cited from 125 papers in the literature published between 1980 and 2022. The citation numbers follow the detailed reference numbers presented in Tables S1 and S2.

For children, the values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI ranged from 4.10×10^{-5} to 8.54 (mean: 7.16×10^{-2}), 1.12×10^{-9} to 2.32×10^{-4} (mean: 1.95×10^{-6}), 2.19×10^{-7} to 4.55×10^{-2} (mean: 3.82×10^{-4}), and 4.12×10^{-5} to 8.58 (mean: 7.19×10^{-2}), respectively.

For adults, the values of HQ_{ing}, HQ_{inh}, HQ_{der}, and HI ranged from 5.50×10^{-6} to 1.15 (mean: 9.62×10^{-3}), 5.03×10^{-10} to 1.05×10^{-4} (mean: 8.79×10^{-7}), 5.58×10^{-7} to 1.16×10^{-1} (mean: 9.73×10^{-4}), and 6.06×10^{-6} to 1.26 (mean: 1.06×10^{-2}), respectively.

Based on Figure 4, the HI values for children from Lake Pamvotis of Greece (24,985 mg/kg; [92]), and Victoria Harbor in Hong Kong (3790 mg/kg; [36] exceeded 1.00. For adult HI values, only the HI value from Lake Pamvotis of Greece exceeded 1.00. This indicates a non-carcinogenic risk of Cu. These HI values were mainly due to the HQ_{ing} value (> 90%) when compared to HQ_{inh} and HQ_{der}.

4. Comments on the Hazard Quotients of Children

From the present estimation of the children's HRA based on Cu levels in the sediments, it is rather far from reality. The ingestion pathway was included in this study with two assumptions that (1) children spend more time on the beach (or muddy sediment areas in the coastal areas), and (2) the sediment-bound metal pollutants could be introduced into children's bodies via the direct ingestion of small particles by hand-to-mouth action. The first assumption is arguable as the definition for children should be well defined. Most of the papers reviewed in this study did not clearly specify the age groups for their children's HRA. Children aged 1–2 years old are different from those who are 10–12 years old. The resistance and sensitivity of the children's bodies are very different between these two groups of ages. Therefore, if those aged 2 to 12 years old are all considered as children, erroneous assumptions could be reached, and as a result, the conclusions would be invalid. Perhaps specifying the body weight of children could reduce the error. However, a similar body resistance and maturity of children between 5 and 12 year old, with a similar body weight of 40 kg, can be assumed. Since obesity among children has become an issue nowadays, the estimation of HQ through the ingestion pathway in a child is somewhat questionable.

The present ecological risk of Cu indicated the median concentration of Cu is generally low and not considered a polluted or a low ecological risk. The localized Cu contamination of the 11 papers exceeding 500 mg/kg dry weight is focused on in this commentary section, as shown in Table 4.

Table 4. Comments on the 11 papers with Cu concentrations over 500 mg/kg dry weight from 125 papers in the literature.

No.	Locations	Cu > 1000	Sources	Comment	Reference
1	Lake Pamvotis, Greece	24,985	Industrial activities; urban stormwater runoff; agriculture, livestock, and domestic sewage.	The report offered baseline data for future research on the anthropogenic influences on the protection and management of Lake Pamvotis, which have been a concern of city officials for decades. Cu must be continuously monitored for ecological and health risks.	[92]
2	Victoria Harbor, Hong Kong	3790	Traffic due to its vicinity to the airport runway and the industrialized area.	Evidently, the increasing levels of toxic Cu in Hong Kong were attributable to the escalating population density, rapid industrialization, and land reclamation. This study can therefore offer a significant source of information about Cu mitigation and pollution management in Victoria Harbor.	[46]

Table 4. *Cont.*

No.	Locations	Cu > 1000	Sources	Comment	Reference
3	Scheldt Estuarine, Netherlands	2600	Industrial discharges.	According to the study, the Cu load in the Scheldt estuary has decreased dramatically for three decades (1960–1990). The extent to which these alterations represent a shift in manufacturing strategies, or a purification of industrial waste is unknown. Cu must be regularly evaluated for environmental and health hazards.	[47]
4	Old Nakagawa River, Japan	1565	Industrialization, urbanization, deposition of industrial wastes and others.	The paper stated that, in order to monitor the trend of Cu contamination, industrial establishments, the municipal council, and/or the government of Japan should reevaluate the current waste treatment and disposal methods for urban sediments or introduce more effective ones.	[76]
5	Mvudi River, South Africa	1027	Release of partially treated wastewater effluents from the Thohoyandou wastewater treatment plant, runoffs from agricultural soil, landfill sites very close to the river and other non-point sources, such as atmospheric deposition	The paper emphasized that higher Cu concentrations in river sediments could potentially have deleterious impacts on aquatic life. Cu must be continuously monitored for ecological and health risks.	[66]
6	Polluted drainage sediments from Peninsular Malaysia	1019	Untreated urban wastes; industrial effluents.	The paper highlighted the importance of treating effluents in this drainage basin. In order to limit unlawful discharges and dumping into drainages, it is necessary to increase public awareness. Cu must be continuously monitored for ecological and health risks.	[72]
7	Kaoshiung Harbor, Taiwan	946	Industrial and municipal wastewaters were discharged from the neighboring industrial parks and river basins. The Cu area was severely affected by untreated or partially treated industrial effluents and municipal sewages.	The paper shed light on the properties and mechanisms of metal distributions in Kaohsiung Harbor sediments. The data would aid in the creation of more effective watershed and harbor management methods to minimize metal discharges into the harbor, as well as a strategy for the cleanup of polluted sediments.	[68]
8	Serbia	870	Not specifically mentioned.	The study indicated that the river sediments examined were highly polluted with Cu. Cu must be continuously monitored for ecological and health risks.	[98]
9	Korbevačka River, Serbia	859.9	Mining and processing of metal ore/ smelting.	The paper stressed the need for a human exposure risk assessment of Cu for screening purposes in order to identify significant exposure pathways and establish the urgency of sediment cleanup measures.	[143]
10	Kaohsiung Harbor, Taiwan.	760	Derived from the polluted Canon River and the Love River, Salt River, and Jen-Gen River.	The paper provided harbor management departments with a great deal of important information, particularly on the Cu derived from the four major contamination sources, allowing for the future control of Cu according to the severity of contamination in sediments.	[108]
11	Shima River, China.	630	Industrial effluents; receiving discharges from Huizhou City.	The paper showed that the Cu bound to sedimentary particles may be resuspended and migrate from the upper and medium reaches to the lower reaches, endangering the safety of the local water supply. Therefore, improving sediment quality necessitates the control of pesticide application, the reduction in industrial wastewater discharge, and the implementation of a river channel dredging project.	[110]

It can be synthesized that, based on Table 4, there is always a need for mitigation, and the management of an exposure risk assessment of Cu that may be utilized effectively for screening purposes in order to identify significant human health exposure routes. Consequently, all Cu-polluted sediments necessitate immediate sediment cleanup measures. Lastly, there is a need for the ongoing monitoring of Cu's ecological and health risks in the future.

5. Conclusions

The present review on Cu concentrations in the sediments based on 125 publications revealed three interesting points. First, there were 11 papers (8%) documenting Cu levels of more than 500 mg/kg dw while China was the country with the highest number (26%) of papers published between 1980 and 2022, out of 37 countries. Second, given that the Cu data cited from the literature were not normally distributed, the maximum Cu level was higher than all the established guidelines in Table 3. However, the median Cu concentration (22.95) was lower than most of all the established guidelines. The I_{geo} median value indicated a status of 'unpolluted', while the CF median value indicated a status of 'moderate contamination', and the ER median value indicated a status of 'low potential ecological risk'. However, ecological risks for Cu could be based at present on the above mentioned 8% of the literature in the present study. Third, all calculated HI values were found to be below 1, indicating no potential chance of Cu non–carcinogenic effects in both adults and children, except for children's HI values from Lake Pamvotis of Greece, and Victoria Harbor in Hong Kong.

It can be synthesized that, based on Table 4, there is always a need for mitigation, and the management of an exposure risk assessment of Cu that may be utilized effectively for screening purposes in order to identify significant human health exposure routes. Consequently, all Cu-polluted sediments necessitate immediate sediment cleanup measures. Lastly, it is recommended that regular monitoring of the ecological–health risks of Cu in sediments should be carried out in view of the expansion of domestic wastes related to urbanization, and the mismanagement of effluents related to industrialization. All these issues are complicated since they are a blend of socio-economic and environmental factors. It is difficult to decide which one is responsible in the first place, and which one will be the priority from the governing body's point of view.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/pollutants2030018/s1, Table S1. The minimum range of Cu concentrations (mg/kg dry weight) in the sediments cited from 125 papers the literature published between 1980 and 2022, and their calculated values of geoaccumulation index (Igeo), contamination factor (CF), ecological risk (ER), hazard quotient ingestion (HQ_{ing}), hazard quotient inhalation (HQ_{inh}), hazard quotient derma contact (HQ_{der}), hazard index (HI) for adults and children based on the cited Cu data; Table S2. The maximum range of Cu concentrations (mg/kg dry weight) in the sediments cited from 125 papers the literature published between 1980 and 2022, and their calculated values of geoaccumulation index (Igeo), contamination factor (CF), ecological risk (ER), hazard quotient ingestion (HQ_{ing}), hazard quotient inhalation (HQ_{inh}), hazard quotient derma contact (HQ_{der}), hazard index (HI) for adults and children based on the cited Cu data.

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