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Flow Regime Impacts on Chemical Pollution in the Water and Sediments of the Moopetsi River and Human Health Risk in South Africa

Abraham Addo-Bediako *, Thato Matita and Wilmien Luus-Powell

Department of Biodiversity, University of Limpopo, Sovenga 0727, South Africa
* Correspondence: abe.addo-bediako@ul.ac.za

Abstract

Many effluents from human activities discharged into freshwater ecosystems cause chemical pollution. Chemical pollution in rivers is a serious threat to freshwater ecosystems due to the associated potential human health risks. This study determined the extent of chemical pollution, identified potential sources of pollution and assessed human health risk in the Moopetsi River, an intermittent river in the Limpopo Province of South Africa. Chemical analyses were conducted on water and sediment samples collected during high-flow, low-flow and intermittent-flow regimes. The findings showed seasonal variations in the chemical pollution levels in the sediments and the highest contamination was measured during intermittent flow. The enrichment factor and geoaccumulation index values identified chromium and nickel as major contributors to sediment contamination. The mean arsenic, chromium and nickel levels exceeded the established guideline values. An evaluation of human health risk was conducted using ingestion and dermal absorption pathways. The results showed that ingestion has greater non-carcinogenic and carcinogenic risks than dermal exposure, especially for children during intermittent flow. The elements of great concern for non-carcinogenic risk were chromium, manganese and nickel and for carcinogenic risk, they were arsenic, chromium, nickel and lead. The outcome of this study is useful for waste management and conservation to reduce environmental degradation and human health risk.

Keywords: chemical elements; contamination; hazard index; hazard quotient; health risk

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1. Introduction

Globally, the growing human population has led to increasing demand for food and services, putting stress on the environment and on resources as a result. Many freshwater ecosystems are so seriously degraded that they can no longer be used for their intended purpose [1]. The pollution of freshwater systems worldwide is largely caused by the discharge of untreated waste from human activities into the environment [2,3]. These effluents contain, among other pollutants, chemical elements and may cause a disruption of the ecological balance [4,5]. Unfortunately, chemical elements are not biodegradable and are persistent in the environment and accumulate in the water, sediment, the aquatic biota and humans [6,7]. Chemicals can cause severe threats to the environment and to aquatic biota and humans [4,5].

Chemical elements are often distributed between the water column and sediments, but most of them are in the sediments [8]. Thus, sediments usually function as sinks for

chemical elements and have the potential to function as a secondary contamination source in the water [9,10]. The degree of contamination in water is governed by physicochemical parameters such as pH and temperature [9,11]. Human exposure to chemicals is linked to many health issues such as cancer and other diseases affecting the brain, liver, bones and kidneys [12].

In many developing countries, chemical pollution is aggravated by a lack of proper treatment of waste before discharging into freshwater environments [13,14]. Monitoring of toxic chemicals in freshwater is increasingly becoming important to determine environmental and human health risks [15,16]. This type of assessment is crucial for environmental management and conservation.

South Africa faces water scarcity, and it is predicted that the demand for water will soon surpass the supply due to the increasing economic activities in the country [17]. South Africa has an average annual rainfall of 450 mm, far lower than the global value of 870 mm [18]. Furthermore, many rivers have been polluted because of intensive agricultural and industrial activities. The Moopetsi River forms part of the Olifants River Basin. The Moopetsi River is currently facing serious degradation due to intensive mining and construction, threatening the ecosystem as a result. It experiences intermittent flow during the dry season. This river provides drinking water, a source of protein (fish), cultural (e.g., spiritual cleansing) and recreational activities for many rural communities, that are vulnerable to pollution-related challenges, hence there is a need to investigate the extent of pollution. This study aimed to assess the variations in the levels of nine elements—arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn)- in the water and surface sediments of the Moopetsi River during different flow regimes. It also evaluated the possible health risks faced by communities relying on the water from the river. The findings of this study provide crucial information for proper waste management and for the implementation of measures to mitigate the impact of chemical contamination on the river and on humans.

2. Materials and Methods

2.1. Study Site

The Moopetsi River forms part of the Olifants River System in South Africa. The Moopetsi River catchment is about 820 km^2 and has an elevation of 656 m. The area is characterized by parallel belts of rocky ridges and mountains, with heavily eroded valleys. The summer and autumn seasons are typically warm with mean daytime temperatures ranging from 19 °C to 22 °C. The winter season is usually mild and the mean daytime temperatures range from 13 °C to 19 °C. The area has an average annual rainfall from 600 mm to 1000 mm and rainfall occurs mainly during the summer and autumn (December to April) [19]. The Moopetsi River provides water for domestic purposes, local irrigation of crops and livestock watering. The main activities around the Moopetsi River include chrome, platinum and sand mining which may contaminate the water. The sampling sites consist of rocks, stones and cobbles, with a few grasses and shrubs at the riverbanks.

2.2. Water and Sediment Sampling

Samples of water and sediments were taken from seven selected points along the river, covering the upper, middle and lower reaches of the Moopetsi River (Figure 1), during high-flow (February), low-flow (July) and intermittent-flow (October) regimes. Water samples were taken and kept in 500 mL acid-rinsed plastic bottles at 4 °C, before chemical analysis. Surface sediment samples were collected using a hand trowel. The samples were kept frozen until chemical analysis. Sediment samples were divided into smaller portions in glass vials and desiccated at 60 °C for 24 h. Nylon with a mesh size of 0.2 mm was then

used to sieve the samples. Using a nitric acid (HNO_3) and hydrochloric acid (HCl) mixture at a 3:1 ratio, 0.1 g of each sediment was mineralized. Chemical element determinations of water and sediments were conducted in parallel using three separate batches. Analyses of elements in both water and sediments were performed using an inductively coupled plasma-optical emission spectrophotometer (ICP-OES, Perkin Elmer, Optima 2100 DV, Markham, ON, Canada), at a certified laboratory (WaterLab) in Pretoria, South Africa. Analytical accuracy was obtained using approved guidelines, and recoveries were within 10% of standard values.

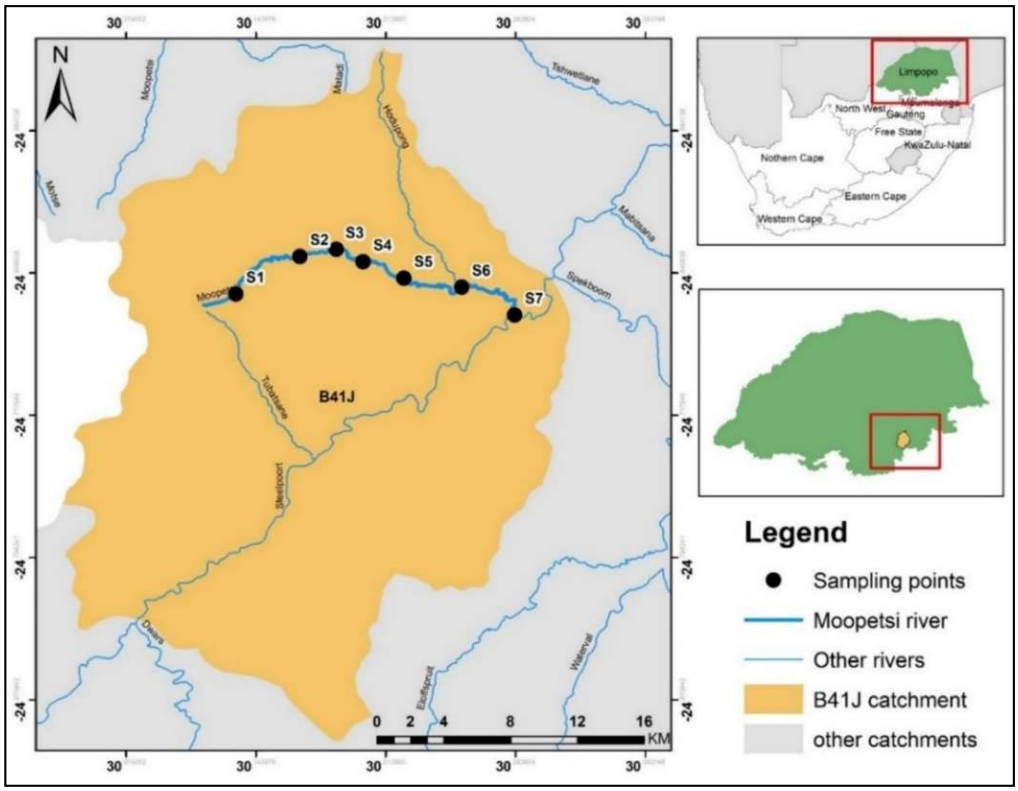


Figure 1. Map of the Moopetsi River showing sampling sites.

2.3. Statistical Analysis

One-way ANOVA was used to determine variations in concentration levels in the sediments and water among the flow regimes, using Statistica (Version 12).

2.4. Sediment Risk Evaluation Indicators

2.4.1. Enrichment Factor (EF)

The presence and pollution levels in the sediments were computed using the enrichment factor [20,21] as follows:

$$EF = [Cn/(Fe)] / [(Baseline\ Cn)/(Baseline\ Fe)],$$

where *Cn* is the concentration of the element. Iron was used as a reference element. The reference values for the enrichment factor are presented in Table 1 [22].

Table 1. Sediments classification using EF and Igeo.

EF Classes	Enrichment Level	I_{geo} Value	I_{geo} Class	Contamination Level
EF < 1	No enrichment	$I_{geo} \leq 0$	0	Uncontaminated
EF = 1–3	Minor enrichment	I_{geo} = 0–1	1	Uncontaminated/moderately contaminated
EF = 3–5	Moderate enrichment	I_{geo} = 1–2	2	Moderately contaminated
EF = 5–10	Moderately severe enrichment	I_{geo} = 2–3	3	Moderately/strongly contaminated
EF = 10–25	Severe enrichment	I_{geo} = 3–4	4	Strongly contaminated
EF = 25–50	Very severe enrichment	I_{geo} = 4–5	5	Strongly/extremely contaminated
EF > 50	Extremely severe enrichment	$I_{geo} > 5$	6	Extremely contaminated

2.4.2. Geoaccumulation Index (Igeo)

In order to determine the chemical contamination caused by natural (geological) process and anthropogenic activities [23], the geoaccumulation index was used:

$$Igeo = log2\,(Cn/1.5Bn)$$

Cn is the concentration of the element in the sediment, B_n is the geochemical background concentration of element 'n', and 1.5 is used as the background matrix correction factor due to lithogenic effects. The geoaccumulation index classes [23] and contamination levels [22] are presented in Table 1.

2.5. Evaluation of Health Risks

2.5.1. Chronic Daily Intake (CDI)

The potential health risks of exposure to chemical elements through oral ingestion and dermal contact were assessed using a quantitative method [24,25].

$$CDI_{ingestion} = \frac{C_w \times IR \times EF \times ED \times CF}{BW \times AT} \quad (1)$$

$$CDI_{dermal} = \frac{C_w \times \mathrm{SA} \times K_p \times \mathrm{ET} \times IR \times EF \times ED \times CF}{BW \times AT} \quad (2)$$

where $CDI_{ingestion}$ denotes the average chronic daily intake by ingestion ($mg{\cdot}kg^{-1}{\cdot}day^{-1}$); CDI_{dermal} is the chronic daily intake by dermal absorption ($mg{\cdot}kg^{-1}{\cdot}day^{-1}$); C_w is the average concentration of the chemicals in water (mg/L), K_p represents the dermal permeability coefficient in water (cm/h): 0.001 for As, Cu, Fe and Mn, 0.002 for Cr, 0.004 for Ni and Pb, and 0.006 for Zn [26]. All other constants in the equation are shown in Table 2.

Table 2. Parameters used in the formula for health risk assessment.

Parameter	Unit	Child	Adult
Exposure frequency (EF)	Day/year	365	365
Body weight (BW)	kg	15	70
Ingestion rate (IR) or daily intake (DI)	L/day	1.8	2.2
Exposure duration (ED)	Years	6	70

Table 2. *Cont.*

Parameter	Unit	Child	Adult
Skin surface area (SA)	cm^3	6600	18,000
Exposure time (ET)	Hours/day	1	0.58
Conversion factor (CF)	L/cm^3	0.001	0.001
Average time (AT)	Days	365 × 6	365 × 70
Particular emission factor (PEM)	Days m^3/kg	1.3×10^9	1.3×10^3

Note(s): Adapted from USEPA [27].

2.5.2. Non-Carcinogenic Risks

The hazard quotient (HQ) through ingestion ($HQ_{ingestion}$) and dermal contact (HQ_{dermal}) were calculated for the non-carcinogenic risk. The HQ is the average daily intake of individuals based on the reference dose (RfD) from ingestion and dermal exposure. The HQs were determined using

$$HQingestion/dermal = \frac{CDI\ ingestion/dermal}{RfDingestion/dermal} \quad (3)$$

where $RfD_{ingestion/dermal}$ is the ingestion/dermal toxicity reference dose ($mg \cdot kg^{-1} \cdot day^{-1}$). When HQ < 1, it indicates there is no non-carcinogenic effect and it is safe, but HQ > 1 indicates a non-carcinogenic risk effect to individuals exposed to the contaminants [28]. The probable non-carcinogenic effect of the various elements is the combined values of the HQs and expressed as the hazard index (HI) for each element. A value of HI > 1 is an indication that exposure may cause an adverse health effect on individuals [29].

2.5.3. Carcinogenic Risk (CR)

Carcinogenic risk from water ingestion or dermal contact was calculated using the equation below [27,30,31]:

$$CRingestion/dermal = CDIingestion/dermal \times CSF \quad (4)$$

where $CR_{ingestion/dermal}$ is the ingestion or dermal cancer risk of encountering chemical elements, *CSF* denotes the cancer slope factor. The CSF values for As, Cr, Pb and Ni ingestion are 1.5, 0.5, 0.0085 and 1.7, respectively. The CSF values for dermal contact for As and Cr are 3.66 and 20, respectively [26]. The overall risk of cancer linked to contaminant exposure in water was taken to be the sum of the incremental risks from each element (ΣILCR). The acceptable cancer risk should be between 1×10^{-6} and 1×10^{-4} [32,33].

3. Results and Discussion

3.1. Chemical Elements in Sediments

There were differences among flow regimes in the mean chemical element concentrations in the sediments (Table 3). Chemical elements are often distributed in varying concentrations in the water and sediments, but more in the sediments [8]. The sediments' concentrations recorded were substantially higher than that of the water column. Higher concentrations of chemical elements in sediments than in the overlying water have been reported in other studies [34,35]. The higher concentrations in the sediments are mainly due to chemicals that are attached to organic or inorganic particles that are later settled in the sediments. Once contaminants are absorbed or adsorbed to a particle, they can be in the sediments for an extended period [36] and potentially become a secondary source of contamination in the water [37,38].

Table 3. Mean concentrations of the elements in sediments measured during different stream flow regimes in the Moopetsi River.

Elements (mg/kg)	High Flow	Low Flow	Intermittent Flow	SQG *	Average Shale Value
	Mean ± SD	Mean ± SD	Mean ± SD		
As	0.608 ± 0.3	0.802 ± 0.2	1.39 ± 0.99	5.9	13
Cr	5619 ± 2362	3176.7 ± 648	4000 ± 2070	37.3	90
Cu	9.46 ± 4.9	10.71 ± 2.9	20.55 ± 15.5	35.7	45
Fe	38,469 ± 8017	35,267.9 ± 2866	34,362 ± 3317	-	47,200
Mn	1183.1 ± 171	1273.3 ± 239	1124.3 ± 323	-	850
Ni	306.71 ± 40	319.3 ± 81.4	263.0 ± 84	-	68
Pb	4.09 ± 1.65	3.62 ± 1.7	6.59 ± 4.1	35	20
Zn	52.14 ± 12	46.43 ± 8.4	48.7 ± 9.2	123	95

Note(s): * Sediment Quality Guideline Guidelines (CCME) [39].

In the sediments, the maximum levels of As, Cu and Pb were obtained during intermittent flow, those of Cr, Fe and Zn were found during high flow, while those of Mn and Ni were recorded at low flow. The mean Cr concentrations surpassed the permissible standard and the average shale value during the three flow regimes. The observed chemical concentration pattern suggests that the contamination is originating from different sources. The Mn and Ni mean concentrations were higher than their average shale values (850 and 68 mg/kg, respectively) during all the flow regimes. However, the mean concentrations of As, Cu, Fe, Pb and Zn were below the respective standard values and average shale values. Human activities in the area could be the leading cause of the high Cr, Ni and Mn levels in the river. For example, the Cr contamination is likely caused by chrome mining in the area. Furthermore, the high levels of Cr, Fe and Zn during high flow could be attributed to the hot summer season leading to the development of anaerobic conditions caused by dissolved oxygen depletion. These conditions accelerate the discharge of chemicals from bottom sediments into the overlying water [40]. In addition, inflows from the catchment area due to flooding may contribute to variations in chemical concentrations in the water and sediment. In contrast, some studies have reported lower concentrations of chemical elements during the wet season caused by the dilution of water, especially after heavy rainfall [41,42]. Conversely, high concentrations of As, Cu and Pb during the intermittent-flow period may be attributed to evaporation and the subsequent accumulation of chemicals in the standing waters.

3.2. Correlation of the Chemical Elements in the Sediments

The correlation analysis reveals strong relationships of the following elements, As-Cu-Pb, Cr-Fe-Mn-Ni-Zn, Cr-Zn, Fe-Zn, Mn-Zn, Ni-Zn (Table 4); for example, As had a strong positive relationship with Cu and Pb ($r = 0.750$, $p < 0.05$; $r = 0.595$, $p < 0.05$, respectively), Cr had a positive association with Fe, Mn and Zn ($r = 0.668$, $p < 0.05$; $r = 0.588$, $p < 0.05$ and $r = 0.764$, $p < 0.05$, respectively), Fe related positively with Zn, ($r = 0.670$, $p < 0.05$), Mn had significant correlations with Ni and Zn ($r = 0.928$, $p < 0.01$; $r = 0.749$, $p < 0.01$, respectively) and Ni related strongly with Zn ($r = 0.696$, $p < 0.05$). The high correlation observed among certain elements could be due to similar contamination sources and, therefore, their concentrations increasing or decreasing together. Secondly, physicochemical characteristics such as pH changes and redox reactions can influence parallel transport of elements like As, Cr, Fe and Mn. Thirdly, areas with similar land-use activities may introduce certain chemicals simultaneously. In the current study area, the major activities that could cause these high correlations could be runoff from mining, agriculture and domestic waste [43–45].

Table 4. Correlation among elements in the sediments of the Moopetsi River (bold indicates $p < 0.05$).

Element	As	Cr	Cu	Fe	Mn	Ni	Pb	Zn
As	1.000							
Cr	−0.333	1.000						
Cu	**0.749**	**−0.533**	1.000					
Fe	0.080	**0.668**	0.027	1.000				
Mn	−0.358	**0.589**	−0.175	0.426	1.000			
Ni	**−0.436**	**0.484**	−0.247	0.252	**0.928**	1.000		
Pb	**0.595**	−0.428	0.386	−0.071	**−0.771**	**−0.863**	1.000	
Zn	−0.069	**0.764**	−0.064	**0.670**	**0.749**	**0.696**	**−0.486**	1.000

3.3. Sediment Risk Evaluation Indicators

3.3.1. Enrichment Factor (EF)

The enrichment factor and geoaccumulation index are two indices normally employed to find the extent of contamination in sediments. The enrichment factor is a measure of the level and possible source of contamination in the environment. The enrichment factor has widely been applied to assess the extent of chemical pollution sources in river sediments [20,21]. The calculated EF values of the elements are shown in Table 5. According to the EF classifications, As, Cu, Mn, Pb and Zn exhibited deficiency to minimal enrichment (EF < 2) in the river. However, Ni and Cr showed moderately severe enrichment (EF > 5) and extremely severe enrichment, respectively, during all the flow regimes. The high Cr values are strongly linked to the high chrome mining and ferrochrome smelter in the basin. In addition, the high Ni enrichment could be caused by mining processes and fossil fuel combustion, especially by mining and other construction vehicles operating in the area. Some studies have reported various activities such as metallurgical processes, the combustion and incineration of fossil fuels and industrial and municipal waste discharge as primary source of Ni in the environment [46–48]. A previous study has reported substantial enrichments of Cr and Ni in four rivers in the Olifants River basin [49].

Table 5. Enrichment factor for elements in the Moopetsi River sediments.

Element	HF	LF	IF
As	0.057	0.083	0.147
Cr	76.60	47.239	61.05
Cu	0.258	0.319	0.627
Fe	1.000	1.000	1.00
Mn	1.71	2.005	1.817
Ni	5.79	6.28	5.313
Pb	0.251	0.242	0.453
Zn	0.673	0.654	0.704

3.3.2. Geoaccumulation Index (I_{geo})

The geoaccumulation index values for the three streamflow regimes are presented in Figure 2. I_{geo} is mainly used for the quantification of chemical element accumulation in sediments. Arsenic, Cu, Fe, Mn, Pb and Zn had values below 1 (<1), and they were in class '0', while the Cr and Ni values were above 1 (>1) for the three flow regimes; Ni showed moderate contamination ($I_{geo} < 2$) and Cr showed extreme contamination ($I_{geo} > 5$), and they were in class 2 and class 6, respectively. The Cr and Ni concentrations are of major environmental concern. The high Cr and Ni concentrations, and EF and I_{geo} values of Cr and Mn, suggest that both geochemical processes and human activities contribute to the river's contamination.

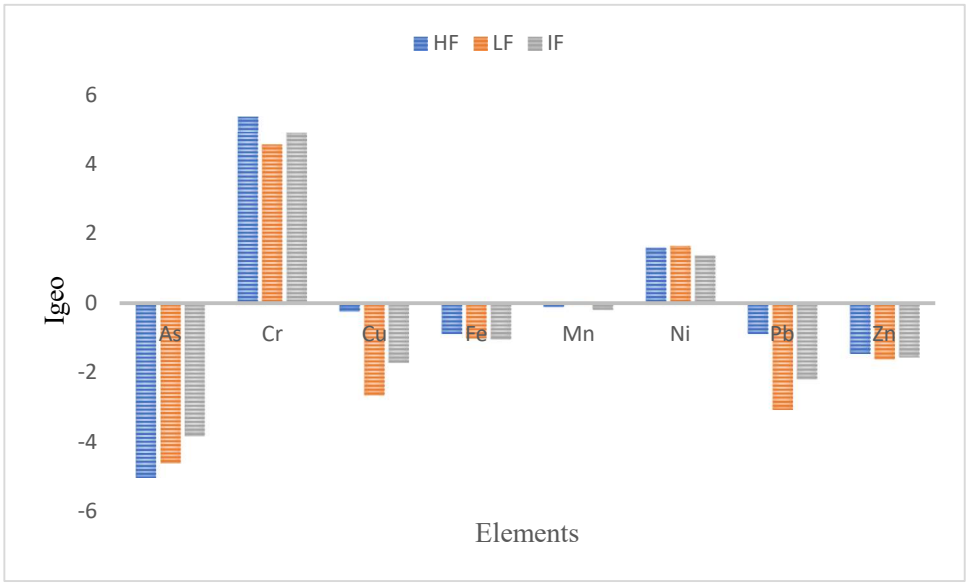


Figure 2. Geoaccumulation index of the elements in the sediments of the Moopetsi River.

3.4. Chemical Elements in Water

Similar to the sediments, there were variations in chemical levels in the water column. The mean concentrations of As, Cr, Cu and Pb were highest during intermittent flow, while the highest mean concentrations of Fe and Zn were recorded during high flow, and low flow had the highest concentrations of Mn and Ni (Table 6). The high levels of As, Cr, Cu and Pb could be due to evapoconcentration of these elements in the residual pools. However, the peak levels of Fe and Zn at high flow could be due to more surface runoff from the catchment, and the high Mn and Ni levels during low flow could be due to reduced dilution and baseflow from groundwater which can introduce certain elements based on the geology during this period. On the contrary, the low levels of most elements during low flow could be attributed to the fact that elements may bind to sediments and settle, leading to lowering of dissolved chemical levels in the water. The mean concentrations of Cr, Fe, Mn and Ni were above the standard values for all the flow regimes [50,51]. Thus, the concentrations of these elements in the water may impact negatively on aquatic biota and humans who consume water and food (fish) from the river. It has been reported that even low chemical concentrations in freshwater ecosystems, together with high nutrient levels may have a negative impact on the biota [52].

Table 6. Concentrations of chemical elements in water at different flow regimes of the Moopetsi River.

Elements (mg/L)	High Flow	Low Flow	Intermittent Flow	SANS	USEPA	WHO
As	0.002 ± 0.0	0.002 ± 0	0.003 ± 0.002	0.01	0.05	0.01
Cr	8.06 ± 5.9	7.95 ± 1.62	10.01 ± 5.19	0.05	0.1	0.05
Cu	0.024 ± 0.013	0.027 ± 0.007	0.05 ± 0.04	2.0	1.3	2.0
Fe	96.3 ± 20.0	88.3 ± 7.5	86.0 ± 8.3	0.3	0.3	0.3
Mn	2.96 ± 0.43	3.19 ± 0.6	2.81 ± 0.8	0.01	0.3	0.04
Ni	0.77 ± 0.1	0.799 ± 0.2	0.66 ± 0.21	0.07	0.1	0.02
Pb	0.01 ± 0.004	0.009 ± 0.001	0.016 ± 0.01		0.015	0.01
Zn	0.13 ± 0.03	0.12 ± 0.021	0.12 ± 0.023	5.0	5.0	3.0

Note(s): SANS [51], USEPA [26], WHO [53].

3.5. Evaluation of Human Health Risk

3.5.1. Non-Carcinogenic Risk—CDI Ingestion

The CDI ingestion values for As ranged from 2.4×10^{-4} to 3.6×10^{-4} mg·kg^{-1}·day^{-1} in children, and from 6.3×10^{-5} to 9.4×10^{-5} mg·kg^{-1}·day^{-1} in adults. The CDI of Cr ranged from 0.86 to 1.10 mg·kg^{-1}·day^{-1} for children and from 2.2×10^{-1} to 2.8×10^{-1} mg·kg^{-1}·day^{-1} in adults; the CDI values for Cu were between 2.9×10^{-3} and 6.1×10^{-3} mg·kg^{-1}·day^{-1} in children, and 8.5×10^{-4} and 1.6×10^{-3} mg·kg^{-1}·day^{-1} in adults; the values for Fe were between 2.20 and 2.40 mg·kg^{-1}·day^{-1} in children, and 5.8×10^{-1} and 6.2×10^{-1} mg·kg^{-1}·day^{-1} in adults; Mn values ranged from 3.8×10^{-1} to 3.8×10^{-1} mg·kg^{-1}·day^{-1} in children and from 8.8×10^{-2} to 1.0×10^{-1} mg·kg^{-1}·day^{-1} in adults; Ni ranged from 7.9×10^{-2} to 9.6×10^{-2} mg·kg^{-1}·day^{-1} for children and from 2.1×10^{-2} to 2.5×10^{-2} mg·kg^{-1}·day^{-1} in adults; for Pb, the values ranged from 1.1×10^{-3} to 1.9×10^{-3} mg·kg^{-1}·day^{-1} for children and from 2.8×10^{-4} to 5.0×10^{-4} mg·kg^{-1}·day^{-1} in adults; and finally the CDI values for Zn were between 1.4×10^{-2} and 1.6×10^{-2} mg·kg^{-1}·day^{-1} for children and from 3.6×10^{-3} to 4.1×10^{-3} mg·kg^{-1}·day^{-1} in adults. The highest CDI ingestion was obtained for Fe during intermittent flow in children (2.4 mg·kg^{-1}·day^{-1}) and the least for As during low flow and high flow (6.3×10^{-5} mg·kg^{-1}·day^{-1}) in adults. The CDI values for all of the chemical elements were greater in children than in adults. The highest CDI values of all the elements, except Mn and Ni, were obtained during intermittent flow (Table 7). The higher levels of Mn and Ni obtained during high flow compared to intermittent flow may be caused by effluents from activities in the area that enter the river during runoff. This is supported by the presence of high concentrations of these elements in the sediment at high flow.

Table 7. Estimated chronic daily intake of chemical elements in water from the Moopetsi River through ingestion pathway.

Dermal	RfD	High Flow	Low Flow	Intermittent Flow	High Flow	Low Flow	Intermittent Flow
	mg·kg^{-1}·day^{-1}	Adult	Adult	Adult	Child	Child	Child
As	0.0003	6.3×10^{-5}	6.3×10^{-5}	9.4×10^{-5}	2.4×10^{-4}	2.4×10^{-4}	3.6×10^{-4}
Cr	0.003	2.5×10^{-1}	2.2×10^{-1}	2.8×10^{-1}	9.7×10^{-1}	8.6×10^{-1}	1.10
Cu	0.04	7.5×10^{-4}	8.5×10^{-4}	1.6×10^{-3}	2.9×10^{-3}	3.2×10^{-3}	6.1×10^{-3}
Fe	0.7	6.2×10^{-1}	5.8×10^{-1}	5.8×10^{-1}	2.40	2.20	2.20
Mn	0.14	9.3×10^{-2}	1.0×10^{-1}	8.8×10^{-2}	3.6×10^{-1}	3.8×10^{-1}	3.4×10^{-1}
Ni	0.02	2.4×10^{-2}	2.5×10^{-2}	2.1×10^{-2}	9.2×10^{-2}	9.6×10^{-2}	7.9×10^{-2}
Pb	0.0014	3.1×10^{-4}	2.8×10^{-4}	5.0×10^{-4}	1.2×10^{-3}	1.1×10^{-3}	1.9×10^{-3}
Zn	0.3	4.1×10^{-3}	3.6×10^{-3}	3.8×10^{-3}	1.6×10^{-2}	1.4×10^{-2}	1.5×10^{-2}

3.5.2. Non-Carcinogenic Risk—CDI Dermal

Generally, the CDI dermal values for all the chemical elements were lower than those of CDI ingestion (Table 8). The As values ranged from 1.6×10^{-6} to 2.4×10^{-6} mg·kg^{-1}·day^{-1} in children, and from 6.6×10^{-5} to 9.4×10^{-5} mg·kg^{-1}·day^{-1} in adults. The CDI values for Cr ranged from 1.1×10^{-2} to 1.4×10^{-2} mg·kg^{-1}·day^{-1} for children and from 4.7×10^{-3} to 5.9×10^{-1} mg·kg^{-1}·day^{-1} in adults; the Cu CDI values were between 1.9×10^{-5} and 4.0×10^{-5} mg·kg^{-1}·day^{-1} in children, and 7.9×10^{-6} and 1.7×10^{-5} mg·kg^{-1}·day^{-1} in adults; the values for Fe were between 1.4×10^{-2} and 1.6×10^{-2} mg·kg^{-1}·day^{-1} in children, and 6.0×10^{-3} and 6.4×10^{-3} mg·kg^{-1}·day^{-1} in adults; the Mn values ranged from 2.2×10^{-3} to 2.5×10^{-3} mg·kg^{-1}·day^{-1} in children and from 9.2×10^{-4} to 1.0×10^{-3} mg·kg^{-1}·day^{-1} in adults; Ni ranged from 2.1×10^{-3} to 2.5×10^{-3} mg·kg^{-1}·day^{-1} in children and from 8.6×10^{-4} to 1.3×10^{-3} mg·kg^{-1}·day^{-1}

in adults; for Pb, the values were from 2.9×10^{-5} to 5.1×10^{-5} mg·kg^{-1}·day^{-1} for children and from 1.2×10^{-5} to 5.1×10^{-5} mg·kg^{-1}·day^{-1} in adults; and the CDI values for Zn were between 5.5×10^{-4} and 6.2×10^{-4} mg·kg^{-1}·day^{-1} in children and from 2.3×10^{-4} to 2.6×10^{-4} mg·kg^{-1}·day^{-1} in adults. Like CDI ingestion, the dermal values for all the chemical elements were found to be higher in children than in adults. Thus, children are at greater risk than adults.

Table 8. Estimated chronic daily intake of chemical elements in water from the Moopetsi River through dermal absorption pathway.

Dermal	RfD	High Flow	Low Flow	Intermittent Flow	High Flow	Low Flow	Intermittent Flow
	mg·kg^{-1}·day^{-1}	Adult	Adult	Adult	Child	Child	Child
As	0.000123	6.6×10^{-7}	6.6×10^{-7}	9.8×10^{-7}	1.6×10^{-6}	1.6×10^{-6}	2.4×10^{-6}
Cr	0.000075	5.3×10^{-3}	4.7×10^{-3}	5.9×10^{-3}	1.3×10^{-2}	1.1×10^{-2}	1.4×10^{-2}
Cu	0.012	7.9×10^{-6}	8.9×10^{-6}	1.7×10^{-5}	1.9×10^{-5}	2.1×10^{-5}	4.0×10^{-5}
Fe	0.045	6.4×10^{-3}	6.0×10^{-3}	6.1×10^{-3}	1.6×10^{-2}	1.4×10^{-2}	1.5×10^{-2}
Mn	0.00096	9.7×10^{-4}	1.0×10^{-3}	9.2×10^{-4}	2.3×10^{-3}	2.5×10^{-3}	2.2×10^{-3}
Ni	0.0008	1.0×10^{-3}	1.0×10^{-3}	8.6×10^{-4}	2.4×10^{-3}	2.5×10^{-3}	2.1×10^{-3}
Pb	0.00042	1.3×10^{-5}	1.2×10^{-5}	2.1×10^{-5}	3.2×10^{-5}	2.9×10^{-5}	5.1×10^{-5}
Zn	0.06	2.6×10^{-4}	2.3×10^{-4}	2.4×10^{-4}	6.2×10^{-4}	5.5×10^{-4}	5.8×10^{-4}

3.5.3. Hazard Quotient (HQ) and Hazard Index (HI)

Generally, the HQ values for Cr, Fe, Mn and Ni through ingestion and dermal exposure exceeded 1, while the HQ of Cu, Pb and Zn through ingestion and dermal routes were less than 1.0 in both adults and children (Table 9). Chemical elements can have negative health effects on humans when the HQ value is greater than 1.0 [28]. The calculated HQ and HI values of Cr, Fe, Mn and Ni during high and low flows show evidence of non-carcinogenic risk at low flow, and there was evidence for non-carcinogenic risk based on the HQ and HI values (>1.0) for As, Cr, Fe, Mn, Ni and Pb during intermittent flow in the Moopetsi River (Table 9). In general, children may experience higher non-carcinogenic risks than adults, and on average the highest HI values were recorded during intermittent flow [31,54–56]. The differences in HI values observed between children and adults could be attributed to differences in sensitivity to chemical exposure and the amount of water consumption [57]. It is therefore important to continuously monitor and assess the physicochemical properties of the river to ensure good water quality [58].

Table 9. Estimated hazard quotients (HQ) and hazard index (HI) for adults and children of the Moopetsi River.

Element	Adult	Child	Adult	Child	Adult	Child
High Flow	HQ_{ing}		HQ_{der}		HI	HI
As	0.209524	0.8	0.0053	0.0129	0.2149	0.8129
Cr	10.550	40.30	70.50	170.0	81.05	210.30
Cu	0.0189	0.072	0.0007	0.0016	0.0195	0.0736
Fe	0.880	3.360	0.141	0.342	1.022	3.701
Mn	0.6645	2.5371	1.0117	2.442	1.6762	4.9791
Ni	1.2069	4.608	1.2601	3.0413	2.4668	7.6493
Pb	0.2245	0.8571	0.0312	0.0754	0.2557	0.9325
Zn	0.0137	0.0524	0.0043	0.0104	0.0180	0.0628

Table 9. *Cont.*

Element	Adult	Child	Adult	Child	Adult	Child
Low Flow						
As	0.2095	0.800	0.0053	0.0129	0.2149	0.8129
Cr	74.910	286.0	62.61	151.0	142.5	437.0
Cu	0.0212	0.081	0.0007	0,0018	0.0220	0.0828
Fe	0.821	3.140	0.133	0.322	0.953	3.472
Mn	0.7161	2.7343	1.0903	2.6318	1.8064	5.3660
Ni	1.2556	4.794	1.3108	3.1640	2.5664	7.9580
Pb	0.2020	0.7714	0.0281	0.0679	0.2302	0.8393
Zn	0.0122	0.0464	0.0038	0.0092	0.0160	0.0556
Intermittent Flow						
As	0.3143	1.200	0.008	0.0193	0.3223	1.2193
Cr	94.40	360.40	78.81	190.30	173.21	550.70
Cu	0.0401	0.153	0.0014	0.0034	0.0415	0.1564
Fe	0.840	3.190	0.141	0.331	0.980	3.521
Mn	0.6308	2.4086	0.9604	2.3183	1.5912	4.7268
Ni	1.034	3.948	1.0795	2.6057	2.1135	6.5537
Pb	0.3592	1.3715	0.0500	0.1207	0.4092	1.4921
Zn	0.0128	0.0488	0.004	0.0097	0.0168	0.0585

3.5.4. Carcinogenic Risk (CR)

Cancer risk usually depends on the degree of exposure and duration. The cancer risk estimates (CRing values) were calculated for As, Cr, Ni and Pb. For the ingestion exposure pathway, the estimated CRing values of these chemical elements in children were higher than in adults. The CR range for adults was from 2.40×10^{-6} to 1.42×10^{-1}, while in children, it ranged from 9.18×10^{-6} to 5.40×10^{-1} (Table 10). The highest CR_{ing} values for both adults and children were for Cr during intermittent flow, whereas the lowest values were for Pb during low flow. In all the elements, the CRing values exceeded the threshold (1×10^{-6} to 1×10^{-4}) in both children and adults [31].

Table 10. Carcinogenic risk of chemical elements through ingestion of water from the Moopetsi River.

Age/Flow Regime	As	Cr	Ni	Pb
Adult (High flow)	9.43×10^{-5}	1.22×10^{-1}	4.10×10^{-2}	2.67×10^{-6}
Adult (Low flow)	9.43×10^{-5}	1.11×10^{-1}	4.26×10^{-2}	2.40×10^{-6}
Adult (Intermittent flow)	1.41×10^{-4}	1.42×10^{-1}	3.52×10^{-2}	4.27×10^{-6}
Child (High flow)	3.60×10^{-4}	4.84×10^{-1}	1.57×10^{-1}	1.02×10^{-5}
Child (Low flow)	3.60×10^{-4}	4.29×10^{-1}	1.63×10^{-1}	9.18×10^{-6}
Child (Intermittent flow)	5.40×10^{-4}	5.41×10^{-1}	1.34×10^{-1}	1.63×10^{-5}

The cancer risk estimates (CR_{dermal} values) were calculated for As and Cr. Similar to the ingestion pathway, the CR_{dermal} values were greater for children than adults. The CR range for adults was from 2.40×10^{-6} to 1.18×10^{-1}, while in children, it ranged from 5.86×10^{-6} to 2.80×10^{-1} (Table 11). The highest CRing values for both adults and children were for Cr during intermittent flow, whereas the lowest were for Pb during low flow in children, and during low flow and high flow in adults. In both children and adults, the CR_{ing} values of all the elements were above the threshold (Table 11), but the risk factor in children was greater than that of adults for both ingestion and dermal pathways. The study also found that the carcinogenic risk is highest during intermittent flow. Similar studies have reported a higher risk in children than in adults to chemical contaminants in water [30,31,59,60]. The level of Ce in water samples during intermittent flow posed the

greatest carcinogenic risks. The findings suggest significant cancer risks for both adults and children from lifelong consumption of water and dermal contact. Carcinogenic risk values are grouped into three levels: (i) a CR value of less than 10^{-6} is at a negligible level; (ii) $10^{-6} < CR < 10^{-4}$ is an acceptable level; (iii) $CR > 10^{-4}$ indicates a high cancer risk to humans [32,33].

Table 11. Carcinogenic risk of chemical elements through dermal contact of water from the Moopetsi River.

Age/Flow Regime	As	Cr
Adult (High flow)	2.40×10^{-6}	1.06×10^{-1}
Adult (Low flow)	2.40×10^{-6}	9.40×10^{-2}
Adult (Intermittent flow)	3.60×10^{-6}	1.18×10^{-1}
Child (High flow)	5.86×10^{-6}	2.06×10^{-1}
Child (Low flow)	5.86×10^{-6}	2.20×10^{-1}
Child (Intermittent flow)	8.78×10^{-6}	2.80×10^{-1}

The chemical contamination in the river may be from different sources such as As from local mining and ore smelting, Cr from chromite ore extraction, Ni also from mining and smelting activities, as well as various industrial processes, and Pb from road transport, leaded fuels and industrial emissions. The extreme levels of some of the elements could be attributed to a lack of proper management of wastewater from human activities. Studies have linked Cr contamination to the mismanagement of wastewater discharges in developing countries [61,62]. The high concentrations of Mn and Ni could be from high-traffic-volume industrial and domestic effluents [63,64]. High levels of chemical pollution in rivers can lead to high health risks including cancer and neurological and reproductive abnormalities. In order to reduce the risk of contamination, waste management guidelines such as avoiding waste dumping in or near the river; ensuring that industrial waste, especially from mines, is pretreated to remove toxic chemicals and other harmful materials before they are discharged into the river; reducing runoff from agricultural fields; and educating communities on the proper disposal of household waste need to be implemented. These measures would reduce human exposure to these chemicals [65–67].

4. Conclusions

This study shows differences in the levels of chemical contamination in the riverine environment among the flow regimes. In the sediment, the major elements causing contamination were Cr and Ni, as supported by EF and I_{geo} values. In the water, the Cr, Fe, Mn and Ni concentrations were higher than their permissible standards. The highest non-carcinogenic risk (HI) was found during intermittent flow for Cr, Mn, Ni, As, Fe and Pb for children, and Cr, Mn and Ni for adults. For carcinogenic risk, the highest CR_{ing} values for Cr, Ni, As and Pb were also found during intermittent flow and the values were greater than the permissible values of between 1×10^{-6} and 1×10^{-4}. The highest CR_{dermal} values for As and Cr were also found during intermittent flow and the values were greater than the guideline values of between 1×10^{-6} and 1×10^{-4}. The CR_{ing} and CR_{dermal} values were greater in children than in adults. Oral ingestion of water and dermal contact of the surface water in the Moopetsi River pose non-carcinogenic risks, as well as carcinogenic risks, to the population living in the catchment. Although the sediments were not extremely contaminated enough to cause a high ecological and human health risk, continued monitoring and proper management are necessary to prevent further deterioration of the river.

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