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Reliability of stored river water as an alternative for consumption in Ekpoma, Nigeria: a human health risk assessment

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ABSTRACT

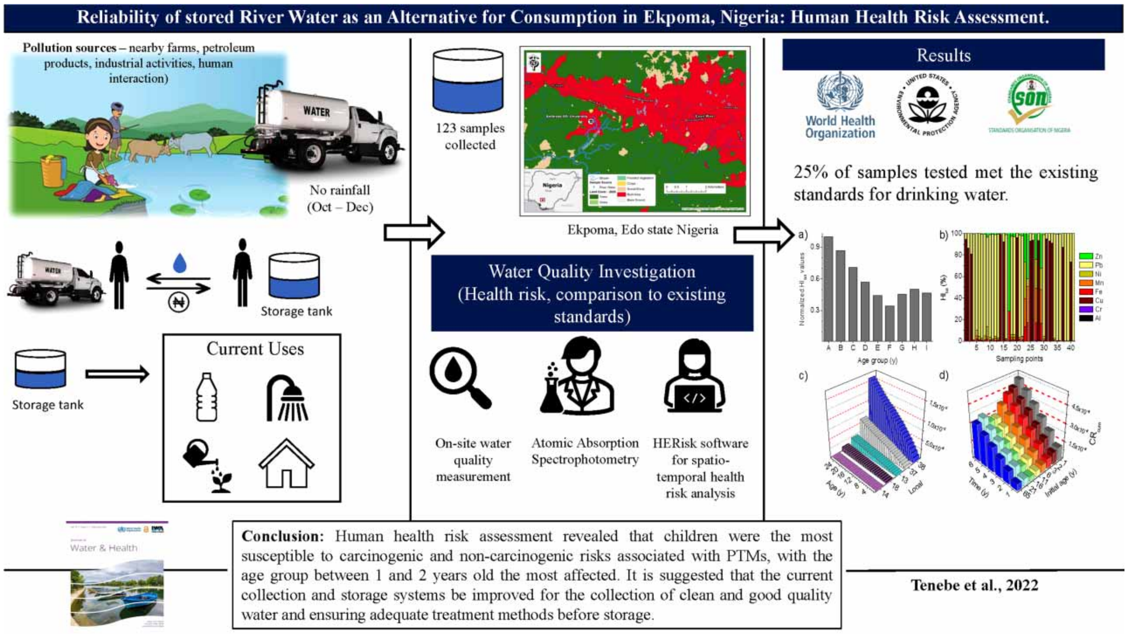
With looming global water-related issues, the monitoring of water quality for household and industrial consumption has become more pertinent. Rivers in nearby towns serve as primary water sources for Ekpoma town. 123 samples of stored river water were collected from 41 sampling locations and physical properties – pH, electrical conductivity (EC), salinity, temperature, and total dissolved solids (TDS) – were measured in situ using the Hanna edge® Multiparameter EC/TDS/Salinity Meter-HI2030. Atomic absorption spectrophotometry (AAS) was used to detect and measure the concentration of potentially toxic metals (PTMs): Al, Cr, Cu, Fe, Mn, Ni, Pb, and Zn. The measured concentrations were compared to the WHO, US EPA, and NSDWQ regulatory standards, and a spatiotemporal health risk analysis was performed using HERisk software. Twenty-five percent of the tested samples contained PTM concentrations within the allowable regulatory limits. Spatiotemporal health risk analysis showed that 98.8% of the cumulative carcinogenic risks (CRcum) were entirely from Pb contamination via oral ingestion. PTM concentrations in the samples suggest the degradation of river water quality due to agricultural activities, crude oil exploration activities, and soil composition in the region. Best management practices (BMPs) and treatment processes for the removal of detected contaminants are recommended to improve water quality.

Key words: chemical elements, drinking water, pollution, public health, risk assessment, river water

HIGHLIGHTS

- The water quality and human health risks from stored river water were examined.
- Eight chemical elements were detected and analyzed from the collected samples.
- Twenty-five per cent of the samples tested contained PTM concentrations within allowable limits.
- Risk analysis showed that 98.8% of the cumulative carcinogenic risks (CR_{cum}) were derived entirely from Pb contamination via oral ingestion.
- BMPs and treatment processes were recommended.

GRAPHICAL ABSTRACT



Conclusion: Human health risk assessment revealed that children were the most susceptible to carcinogenic and non-carcinogenic risks associated with PTMs, with the age group between 1 and 2 years old the most affected. It is suggested that the current collection and storage systems be improved for the collection of clean and good quality water and ensuring adequate treatment methods before storage.

1. INTRODUCTION

Industrial activities have increased in the last century, resulting in an exploration of the Earth's resources (Briffa *et al.* 2020). Improper management of these resources and the disposal of residual products have led to an increase in environmental pollution. Some by-products of these processes are organic, inorganic, radioactive, gaseous, and organometallic compounds. Water, one of Earth's resources, is a known medium for the transport of biological, organic, inorganic, and radioactive contaminants from the environment to the human body. Water contamination could be due to naturally occurring or anthropogenic matter (Zeng *et al.* 2020). The quality of freshwater in many parts of the world has degraded over the past few years and attracted global attention. This is because many human activities currently depend on freshwater sources. Contamination of these water bodies poses a risk to human health and ecology (Emenike *et al.* 2020). A recent study found that microplastics found in aquatic environments can absorb metals, resulting in a change in their chemical structure, thus posing a greater risk to human and environmental health (Fred-Ahmadu *et al.* 2022a, 2022b). One major source of anthropogenic contaminants in surface water bodies is urban and agricultural stormwater. Research suggests that potentially toxic metals (PTMs) can be found in cultivated and uncultivated soils from fertilizer overapplication and natural processes, such as weathering and leaching of rocks (Xie *et al.* 2022; Yang *et al.* 2022). Dore *et al.* (2014) and Suvarapu & Baek (2017) found that air is a medium for transporting PTMs into soil and river networks. These PTMs also settle in water and form sediments in river networks, thus affecting aquatic life and biota (Zhang *et al.* 2010; Briffa *et al.* 2020; Zeng *et al.* 2020). Anake *et al.* (2020) investigated dust samples for particulate matter and discovered the presence of silver, aluminum, cadmium, and other metals in the samples tested. Another study tested roadside dust particles and discovered ten different metals in the samples, including As and Cu (Emenike *et al.* 2019).

Metals and metalloid ions are divided into three categories. The metals in the first group include mercury, cadmium, and lead, all of which are toxic at low concentrations. The second group of metals is less dangerous (bismuth, indium, thallium, and antimony), and the third group includes essential metals, such as zinc, cobalt, copper, iron, and selenium, which are involved in the body's chemical or biochemical functions and are toxic at high concentrations (Zaynab *et al.* 2022). Trace elements such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni), selenium (Se), vanadium (V), and zinc (Zn) pose major threats to human health and the environment at varying exposure levels (Dore *et al.* 2014). These trace elements in their organic forms can be absorbed by plants, transferred to the food chain, and have drastic effects on wildlife when critical limits are exceeded (Spurgeon *et al.* 2013). Some PTMs have also been found to affect DNA, organs, and cells, and as such, are classified as carcinogens (Zeng *et al.* 2020). The presence of

PTMs in the atmosphere is intensified in areas with industrial and vehicular emissions and secondary aerosols (Suvarapu & Baek 2017; Zeng *et al.* 2020). Other sources of PTMs in the aquatic environment include leaching from landfills, agricultural fields, and weathering. The presence of PTMs in air, soil, and rainwater can directly affect their presence in surface water bodies (Souza *et al.* 2016).

The hydrological dynamics of a river affect the transformation and accumulation of PTMs in the water (Zeng *et al.* 2020). Chaturvedi *et al.* (2015) and Essa *et al.* (2018) found that aqueous PTMs solutions (Hg, Cd, Pb, Ag, Cu, and Zn) were subjected to the volatile metabolic products of *Escherichia coli* Z3 for 24 h under aerobic laboratory conditions, and the toxicity of the metals was removed by immobilizing the metal ions. This shows that in water bodies with adequate dissolved oxygen and good microbial balance, PTMs would be a reduced concern. However, there has been an observed increase in the pollution load in surface water bodies. Poor effluent management introduces high levels of contaminants into rivers and lakes, particularly in industrial regions (Emenike *et al.* 2018; Samuel *et al.* 2018a, 2019). River flow is relatively slow (Zeng *et al.* 2020); thus, activities in areas close to the water body can negatively affect water quality at a faster rate than the self-purification rate of the river. PTMs in rivers can be transferred from one basin to another (Zhang *et al.* 2021, 2022).

This study examined the presence of PTMs in river water samples collected from storage tanks in Ekpoma, south-–south Nigeria, and attempted to estimate the health risks associated with the consumption of water from these sources, which is a major drinking water source in the community (Tenebe *et al.* 2020). The geological formation of Ekpoma makes access to the groundwater supply very difficult (Okeke *et al.* 2011). This has led communities to source water from rainwater catchments and purchase water from nearby rivers (Tenebe *et al.* 2020). A study examining the microbiological quality of the main sources of water in the environment revealed that there was heavy pathogenic contamination of water. Furthermore, the study analyzed the catchment (harvesting and storage) conditions of the water in the Ekpoma community and showed that the water is stored in concrete wells and covered with corrugated roofing sheets, which in some cases have rusty parts (Tenebe *et al.* 2020). It is also common knowledge that the exposure of concrete to water results in leaching (Hitomi n.d.; Khayan *et al.* 2019; Yuan 2018) which then introduces trace quantities of PTMs into water (Hohberg & Schiessl 1997; Magnus 2011). However, in Ekpoma, water is constantly stored in these concrete wells, and as such, there is a potential increase in the concentrations of PTMs in the water (Dic-Ijiewere *et al.* 2018). Omoigberale *et al.* (2013) found that rivers in the Ehor community area are used by members of the community for activities such as bathing, washing clothes, etc. Many occupants in Ekpoma are farmers, which could mean a higher rate of PTMs in surface waters from runoff in agricultural fields. There is also the potential for dumping solid waste in drainage channels and surface water bodies, which is a common practice in the nation (Agunwamba *et al.* 2013; Olukanni *et al.* 2014; Tenebe *et al.* 2023). Therefore, it is pertinent to investigate the level of PTM contamination in river water resources to estimate, combat, and control public health risks (Samuel *et al.* 2018b).

This study aimed to investigate the PTM concentrations in river water samples collected from storage tanks and wells in Ekpoma, Edo State, Nigeria. The results from these samples were compared to the US EPA, NSDWQ, and WHO drinking water regulatory standards. A health risk analysis associated with both oral and dermal water consumption was carried out, and results showing the estimate of the most affected age group in the community were provided. Furthermore, this study sheds light on the importance of water quality assessment and provides insight into the health risks associated with the consumption of river water in the Ekpoma community. Suggestions on best management practices (BMPs) that can be implemented to improve water quality and consequently reduce PTM consumption in the region were offered in this study.

2. RESEARCH METHODOLOGY

2.1. Study site

Ibiekuma stream is a major surface water body with tributaries flowing into many towns in the Ishan region of Edo State. In the months with no rainfall, water merchants draw water from rivers in the Ibiekuma stream and sell it to communities in the Ishan region. The water purchased is stored in plastic and concrete storage tanks for later use. Stored river water samples were collected from Ekpoma, a town situated in the northern Ishan area of Edo State, South–South Nigeria. The Northern Ishan region covers about 150 km^2 (Figure 1) of land space and is stationed between latitudes 6° 41′ – 6° 5′ and longitudes 6° 00′ – 6° 30′ (Tenebe *et al.* 2022). In 2005, Ekpoma town was predicted to have a population of 834,750, with a 3% annual population growth rate (Okeke *et al.* 2011). The town is classified as a rural community, with most residents relying on small-to medium-scale agricultural techniques to make a living (Tenebe *et al.* 2020). During seasons of minimal rainfall,

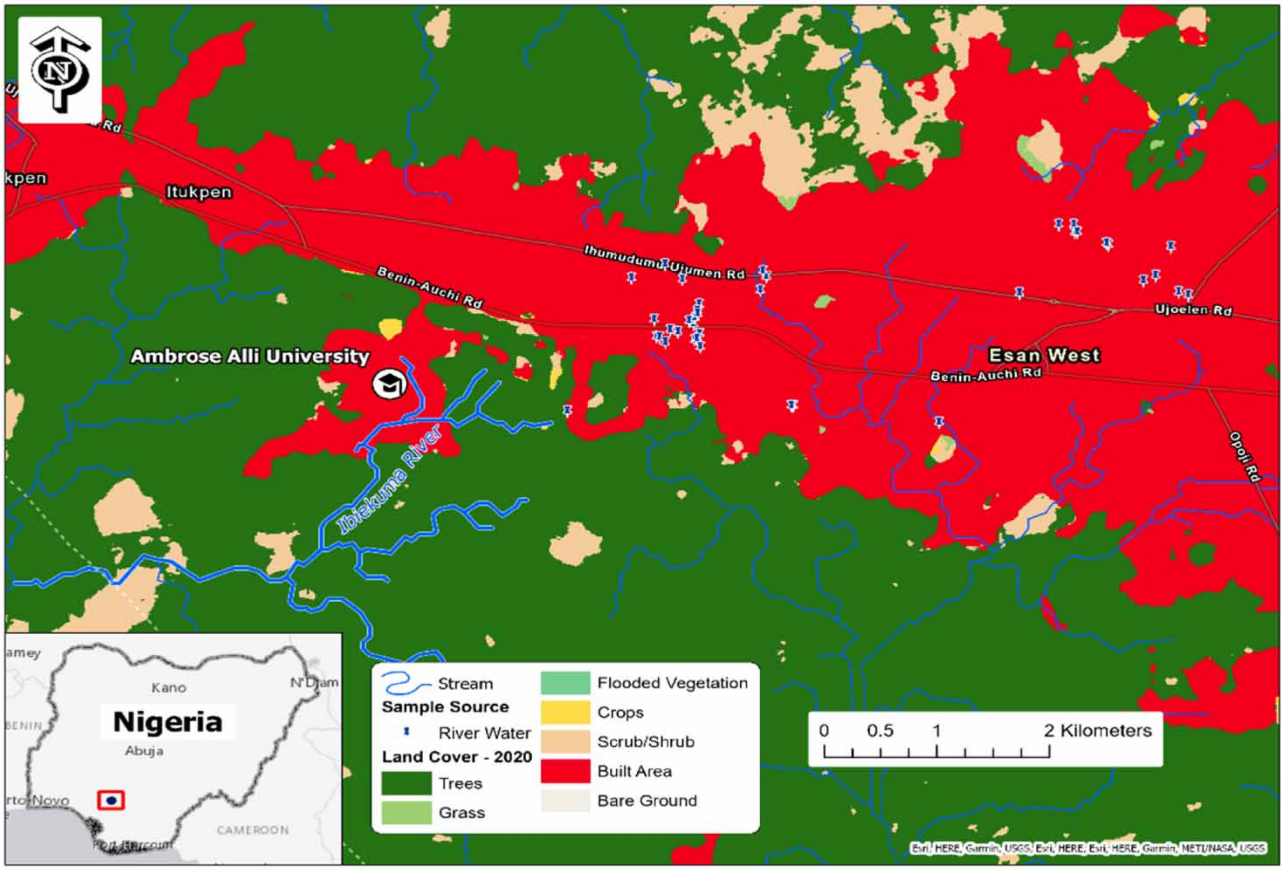


Figure 1 | River water sampling locations in Ekpoma, Edo State.

25 L of water was sold for 50–60 naira as of 2020. This water is sourced from surface water bodies in nearby towns and stored in roof-harvested rainwater during the wet season. Tenebe *et al.* (2020) claimed that the water sold to community members comes from storage tanks containing potentially dangerous bacteria and perhaps poisonous chemical components. The people of Ekpoma live on a daily income of less than one dollar and must deal with the ever-increasing prices of water sold in the town. The limited availability of water during times of little rain fosters a bidding war to access water sourced from rivers. Water merchants sell to the highest bidder and people in accessible areas, as some regions are characterized by poor road infrastructure, and thus demand extra time to supply those regions.

The surface water collected and sold during periods of minimal rainfall is sourced from rivers that are used for agricultural irrigation, washing, bathing, and industrial processes. The dumping of wastewater in these rivers is not controlled and water collection and storage processes are not monitored. This necessitates evaluation and health risk assessment to determine the potential dangers posed by the consumption of this water and to suggest required regulatory actions to mitigate these risks (Tenebe 2020; Ogarekpe *et al.* 2020).

2.2. Experimental procedure

2.2.1. Samples

A total of 123 samples were obtained from 41 sampling locations in the community and were analyzed between October and December. The sampling points were separated into five regions: Idumegbo, Ihumudumu, Ujoelen, Emando, and Ukpenu Extension. Three samples were collected from the storage tanks at each sampling point to provide a more accurate representation of the conditions being assessed. Samples were obtained from storage tanks and not from rivers to provide a full representation of the condition of water being consumed by the community. Obtaining water samples from rivers would not provide a representation of contamination because of the water collection practices and possible combinations of water from multiple rivers. The samples were collected in sterile high-density polyethylene (HDPE) bottles, capped, stored in ice at 4 °C, and transported to the laboratory for analysis.

The tankers used for the collection of water from the river are metal tankers. Currently, no information is available on the quality of the tankers, the coating on the inside and outside and how these directly affect the water quality. The tanks used for storage are PVC storage tanks, while others are made purely out of concrete. The concrete tanks in this community are not coated with any paint, thus paint from the storage tank is very unlikely to impact the water quality.

2.2.2. Reagents

All the reagents used were of analytical-grade. Concentrated HNO_3 (nitric acid) and 30% (m/v) H_2O_2 were used for the digestion of all samples. Distilled water (18.3 MΩ cm^{-1}) was obtained from the Barnstead™ Easypure™ II water purification system. Glass and plastic apparatus used for experimental procedures were cleaned using 10% nitric acid and rinsed several times with distilled water.

2.2.3. Sample preparation procedures

Conventional wet acid digestion was used to prepare the samples for elemental analysis. 25 mL of sample was combined with 15 mL of concentrated HNO_3 in a 150 mL glass beaker. The resulting solution was heated on a hot plate at 85 °C for about 3 h. Once the solution had been cooled to ambient temperature, 10 mL of H_2O_2 was added. The solution was reheated until a clear solution was obtained. Following this, the solution was reduced to about 2 mL, quantitatively transferred into a 25 mL volumetric flask, and made up to the 10 mL volume with distilled water. The samples were stored at 4 °C prior to being analyzed. Three representative samples were prepared for each analysis as a quality control measure, and after each 12-sample analysis, a blank sample with three standards was analyzed. Standard solutions of all eight elements were prepared using analytical-grade ingredients, reagents, and a dilution factor of 1,000 mg L^{-1}.

2.2.4. Apparatus

A Hanna H198130 probe attached to a waterproof Hanna edge® Multiparameter EC/TDS/Salinity Meter-HI2030 was used to measure the physical parameters of the samples, such as pH, electrical conductivity (EC), salinity, temperature, and total dissolved solids (TDS) at the collection points. The physical characteristics of water samples should always be measured because they provide insight into the potential sources of contaminants in the samples tested. The multiparameter instrument was standardized and calibrated using buffer solutions with pH values of 4.0 and 7.01 and potassium chloride (EC) (Tenebe *et al.* 2022). Under standard operating conditions, the UNICAM 969 atomic absorption spectrometer (AAS) – SN. 12083 was used to quantify the eight chemical species (Al, Cr, Cu, Fe, Mn, Ni, Pb, Zn). The analysis was conducted using a 3111A alpha standard and N–A and A–A flames in the spectrophotometer. To detect each metal, the samples were subjected to UV wavelengths ranging from 217.0 to 357.9 nm. The AAS measurement time was 4 s. The instrument had detection limits for the elements as follows: Pb and Mn (0.001 mg L^{-1}); Ni, Cr, Zn, and Cu had (0.005 mg L^{-1}); Fe (0.020 mg L^{-1}), and Al (0.100 mg L^{-1}).

Method detection and quantification limits (Limit of Detection (LOD) and Limit of Quantification (LOQ)) were calculated in the following equations:

$$\mathrm{LOD} = 3 * \frac{\sigma}{S} \tag{1}$$

$$\mathrm{LOQ} = 10 * \frac{\sigma}{S} \tag{2}$$

where σ is the standard deviation of analytical blank measurement and S is the slope of the calibration curve ($y = mx + b$) (Ahmad *et al.* 2021; Feng *et al.* 2022; Wan *et al.* 2022).

2.3. Comparison of observed PTM concentrations with drinking water standards

Many developing and developed countries are struggling to preserve and monitor water quality because of the increasing degradation of groundwater and surface water quality. Because the quality of water from conventional sources has deteriorated, more people are considering alternative water sources, such as seawater, rainwater, reclaimed water, and brackish water. Water quality testing and monitoring were performed on the water obtained from these sources on a regular basis. To protect humans from the chronic and acute effects of drinking contaminated water, toxicologists, and regulatory agencies from various countries have collaborated to develop standards that serve as guidelines for limiting the concentration of potentially toxic species in drinking water, regardless of the water's primary source. The quantities of PTMs found in the river water samples were recorded and compared to the US EPA, NSDWQ, and WHO-established acceptable limits. The percentage deviation from the allowable limits was also recorded and the data were used to construct a human health risk assessment related to river water harvesting.

2.4. Human health risk assessment

A human health risk assessment was carried out considering eight chemical species (Al, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) as potential toxic metals (PTMs). The data used for the assessment are the mean values of triplicate samples recorded from each location. HERisk software was used to perform calculations. This software is essential for carrying out a spatiotemporal risk assessment, which provides a more detailed health risk assessment. In contrast to the previous version (HHRISK) and other codes, this software considers in its calculations the variation of a parameter related to residents' growth and, consequently, their changes in eating and behavioral habits (Neris *et al.* 2019a, 2021). More details about the methodology applied in the software and how it works can be found in Neris *et al.* (2021). The program allows users to provide different values for age-dependent parameters, such as body weight (BW), skin surface area (SA), and water intake rate (IR). In addition, the program contemporarily performs risk assessments for nine different initial ages (AI = 1, 2, 3, 6, 11, 16, 18, 21, and 65 years) of exposure to PTMs, based on these AIs, the code defines the age-dependent parameter values depending on the resident's age at a given time *t*. The code uses modified equations provided by the US EPA. Equations present in the HERisk code used to carry out the human health risk assessment and information on the parameters and values used for the exposure doses and elemental human health risk calculations are contained in the supplementary information provided by Tenebe *et al.* (2022) and Neris *et al.* (2019b).

The population of the studied region uses river water for drinking and bathing during the dry season (6 months). For this reason, the contamination routes considered were ingestion and dermal contact with water, with an exposure frequency (EF) of 183 days. During the rainy season, residents use rainwater for drinking and bathing. The daily intake dose and the dermal contact dose were calculated using equations provided by Tenebe *et al.* (2022). Health risk evaluation was carried out for residences and the experimental concentrations of chemical species were assumed to be constant over time to allow for the assessment of human health risks.

The possibility of a resident developing life-long cancer as a result of chronic carcinogenic species exposure (potential carcinogenic risk (CR)) and non-carcinogenic effects resulting from ingestion or dermal contact with potentially toxic species (non-carcinogenic hazard quotient (HQ)) (US EPA 2005, 2007) were evaluated using the following equations (Tenebe *et al.* 2022; US EPA 2004).

$$\mathrm{HQ}^{\mathrm{IA}}(t) = \frac{\mathrm{D}^{\mathrm{IA}}(t)\cdot \mathrm{BAF}}{\mathrm{RfD}} \tag{3}$$

$$\mathrm{CR}^{\mathrm{IA}}(t) = \mathrm{D}^{\mathrm{IA}}(t)\cdot \mathrm{SF}\cdot \mathrm{BAF}\cdot \mathrm{ADAF} \tag{4}$$

where IA is the dose at time *t* for initial age IA (mg kg^{-1} day^{-1}), BAF is the chemical species dose fraction absorbed by the organism (bioavailability factor), RfD is the reference dose of the chemical species (mg kg^{-1} day^{-1}), ADAF is the age-dependent adjustment factor, and SF is the slope factor $(\mathrm{mg\ kg}^{-1}\ \mathrm{day}^{-1})^{-1}$, which converts the estimated daily doses averaged over a lifetime directly into the probability of a person developing cancer (Tenebe *et al.* 2022).

Because of the presence of two exposure routes, it was important to calculate the aggregated hazard index ($\mathrm{HI}_{\mathrm{agg}}$) and aggregated potential CR. To assess the overall risk, all exposure routes were incorporated, along with all chemical species (*n*), total hazard index ($\mathrm{HI}_{\mathrm{tot}}$), and the cumulative potential CR. Equations for this estimation can be found in Tenebe *et al.* (2022).

The classifications of non-carcinogenic and carcinogenic human health risks were extracted from the US EPA (1989) and Li *et al.* (2014), respectively. Non-carcinogenic risks (CRs): $\mathrm{HI}_{\mathrm{tot}} < 0.1$ (negligible); $1 > \geq 0.1$ (low), $4 > \mathrm{HI}_{\mathrm{tot}} \geq 1$ (medium), and $\mathrm{HI}_{\mathrm{tot}} \geq 4$ (high), while CR: $\mathrm{CR}_{\mathrm{cum}} < 1 \times 10^{-6}$ (negligible), $1 \times 10^{-4} > \mathrm{CR}_{\mathrm{cum}} \geq 1 \times 10^{-6}$ (low), and $\mathrm{CR}_{\mathrm{cum}} \geq 1 \times 10^{-4}$

(high). CR_{cum} values exceeding 1×10^{-4} imply that more than one in 10,000 residents presents the possibility of developing cancer.

3. RESULTS AND DISCUSSION

For the purpose of validating the experimental method, the LOD and LOQ for the test method were determined using Equations (1) and (2) and the results are shown in Table S4 of the supplementary document. The lower values of the detection limits indicate the sensitivity provided by the test method. The Relative Standard Deviation (RSD) % was calculated and the results ranged from 0.20 to 5.45 (Table S4).

3.1. Physical properties of stored river water samples

The physical properties of river water samples retrieved from storage tanks in the Ekpoma community revealed that the pH values in the study areas ranged from 8.0 to 9.2. The pH values may also be related to various human activities such as agricultural practices, washing, bathing, and using latrines near the source of water, in this case, rivers (Dey *et al.* 2021). The pH results were similar to those measured in rivers in other countries and 6.5–8.5 was specified for irrigation water and aquatic life (Anifowose & Oyebode 2019). Although a pH range of 6.5–9.0 is recommended for freshwater bodies, the US EPA lists industrial discharges, alkaline geology, soils, asphalt production or disposal, agricultural lime, oil and gas brines, industrial landfills, cement manufacturing, soap manufacturing, and limestone gravel roads as contaminant sources that contribute to high pH levels in the water. However, the TDS levels measured in the collected samples ranged between 0.06 and 89.6 mg L^{-1}. Excess dissolved solids in water have been associated with increased water temperature, inhibition of photosynthesis, and increased turbidity (Dey *et al.* 2021). The TDS values measured in the collected samples were lower than the regulatory guideline of 500 mg L^{-1}. The measured salinity values of 0–0.4 mg L^{-1} indicate that the water was sourced from freshwater. The total ionic concentration of water is often represented by measuring EC, and high conductivity values reflect higher water pollution. In this study, the EC values of the samples collected from different locations ranged from 0.15 to 227.3 μS/cm. The WHO set values for EC in drinking water at 400 μS/cm; hence, the EC measured in the samples falls within the acceptable limits. From the physical parameter analysis, it can be concluded that the river stored in the Ekpoma community is satisfactory in terms of EC, pH, salinity, and TDS values.

3.2. Comparison of observed water quality with drinking water standards

The river water samples collected from the 41 sampling locations were grouped into five regions, with three samples collected from each sampling point (Table 1). Drinking water regulatory standards consider water safe for domestic use when it contains the same or a lower concentration of a substance, as stated by regulatory agencies, such as the US EPA, WHO, and NSDWQ. Metal contamination of water is a concern affecting the environment. Even at low concentrations, these metals can be hazardous owing to their non-biodegradable nature (Zaynab *et al.* 2022). According to the WHO regulatory standard, Cu, Pb, Cr, Ni, and Al have set concentration limits but no limits for Fe, Mn, and Zn concentrations because these metals are rarely found in drinking water samples and thus are not considered threats to human health. The most recent US EPA regulations do not specify concentration limits for Ni because its presence in US drinking water is infrequent. As a result, Ni has become unregulated, and its regulatory limits are being revised. All the chemical elements described in this study have

Table 1 | River water sampling districts

District	Notable landmark	Samples in district	Total no. of samples in district
1	Along Benin-Auchi road, Near St. Nicholas Street, Onigbinde Cresent, Efe Block Road, Princeton hostel	1, 2, 3, 7, 9, 10, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41	18
2	Along Ihumudumu–Ujumen Road, Oshagale Street, Divine Royalty Hotel	4, 5, 6, 8	4
3	Ugoelen Extension Road	11, 12, 13	3
4	Emando, Close to Ihumudumu–Ujumen Road	15, 16, 17, 18, 19, 20, 22, 23, 24, 25, 26, 27, 28, 29	15
5	Ehimen Street	14	1

Table 2 | Average district PTM concentrations detected compared to regulated allowable limits

					Average values				
S/N	**Element**	**USEPA MCL (mg L^{-1})**	**WHO MCL (mg L^{-1})**	**NSDWQ MCL (mg L^{-1})**	**District 1**	**District 2**	**District 3**	**District 4**	**District 5**
1	Fe	0.300	–	0.300	0.438	0.182	0.323	0.492	0.375
2	Zn	5.000	–	3.000	0.067	0.014	0.062	0.134	0.101
3	Cu	1.300	2.000	1.000	2.319	ND	ND	2.669	1.847
4	Pb	0.000	0.010	0.010	0.800	0.139	0.310	0.291	0.017
5	Mn	0.050	–	0.200	0.440	0.098	0.310	0.426	0.359
6	Cr	0.100	0.050	0.050	0.154	ND	ND	0.198	0.259
7	Ni	–	0.070	0.020	0.110	0.044	0.060	0.053	0.039
8	Al	0.200	0.200	0.200	0.001	ND	ND	ND	ND

ND, not detected.

regulatory limits in the NSDWQ. The allowable limits provided in the regulatory standards were compared with the concentration values of the PTMs detected in this study (Table 2). The potential toxicity of the metals, without a basis for comparison across all standards, was evaluated in depth using human health risk assessment. This was crucial for assessing the danger of exposure to prolonged dermal and oral consumption at the measured concentration levels. Metals, which are present in food and the environment in small amounts, are essential for good health; however, larger concentrations can be toxic or detrimental. Their harmful effects on the blood, liver, kidneys, brain, lungs, and other crucial organs can lead to energy depletion. Long-term exposure eventually causes the degeneration of physical, tissue, and neurological processes that mimic diseases such as multiple sclerosis, Parkinson's disease, muscle dystrophy, and Alzheimer's disease (Tenebe *et al.* 2019a; Zaynab *et al.* 2022).

A study to examine the levels of Cd, Cr, Pb and Zn in water samples and fishes in the Ibiekuma stream was conducted in 2008. This study compared metal levels in the downstream and upstream segments of the river as well as the seasonal variations in metal concentration. The results revealed that Zn presented the highest levels of contamination in both water (1.12 ± 0.51 mg/L) and fish (45.5 ± 7.46 mg/kg) samples. Cd was also present in water (0.005 ± 0.002 mg/L) and fish (0.70 ± 0.22 mg/kg) samples, while Pb was only present in water (0.005 ± 0.002 mg/L) samples tested. Cr (0.02 ± 0.01 mg/L, 5.3 ± 2.45 mg/kg) and Cu (0.05 ± 0.03 mg/L, 19.0 ± 5.22 mg/kg) were present in concentrations exceeding the standards presented by the WHO in 1972. Metal concentrations in the river and fish were lower in the wet seasons and higher in the dry seasons (Obasohan 2008a). Another study examined the concentration of Fe, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn in the sediments of the Ibiekuma stream. The contamination levels of the respective metals were varied and the range of monthly mean values in μg/g were Fe (236.9–876.32), Cd (0.01–0.36), Cr (0.08–0.14), Cu (1.05–1.97), Mn (11.16–15.42), Ni (0.020–0.35), Pb (0.11–0.92), V (0.008–0.017), and Zn (44.96–55.89). Except for Fe and Mn, the seasonal mean values of the metals were not varied. The metal concentrations (with the exception of Cd) were lower than the average values for the continental crust and unpolluted African inland water sediments. It was inferred in the study that the measured metal contamination in the stream might not immediately pose a threat to the organisms present and to people who use the stream for drinking water and other domestic purposes. However, due to the slightly elevated Cd levels, close monitoring of metal pollution in the stream was advised in light of potential long-term concerns that Cd may present to the ecosystem (Obasohan 2008b).

In 2022, the water sources for consumption in Ekpoma and nearby towns will be withdrawn from the rivers in months with no rainfall. This implies that higher metal concentrations are taken from the rivers and sold to consumers. With increasing global pollution levels and more health studies, regulatory bodies have revised the allowable limits for some metals like Zn and Pb in drinking water. The results from this study show varying metal concentrations when compared to the results presented by Obasohan (2008a). Some districts have concentrations higher than those presented in the 2008 study and vice versa. This is also indicative of possible contamination during water collection and storage as well as the potential difference at different collection points along the river. Because this study does not consider sediment quality, the results cannot be compared to those presented by Obasohan (2008b). However, the concentrations of metals in the stream sediments are indicative of pollution levels and how the source water contributes to the quality of the water being sold in towns.

Figure 2(a)–2(h) presents a graphical and individual element outlook of the information presented in Table 2. Cu, Cr, and Al were not detected in the samples collected from Districts 2 and 3 (Figure 2(b), 2(f) and 2h). The samples collected from District 5 had Cu levels higher than the US EPA and NSDWQ limits, whereas samples obtained from Districts 1 and 4 had higher Cu levels than the US EPA, WHO, and NSDWQ levels. The samples obtained from Districts 4 and 5 contained Cu levels higher than the US EPA and NSDWQ limits. All districts in Ekpoma had Pb levels above the NSDWQ limit (Figure 2(d)). Literature suggests that crude oil exploratory activities in the environment are a potential source of Pb and Cu contamination (Anake *et al.* 2020). The presence of Pb has also been linked to emissions from automobiles, electricity generators, lead-acid batteries, solder, and alloys (Anifowose & Oyebode 2019). Acute Pb exposure is known to cause fatigue, insomnia, arthritis, hallucinations, appetite loss, headaches, stomach discomfort, renal dysfunction, hypertension, and vertigo (Yang *et al.* 2022; Zaynab *et al.* 2022). Cu is a redox element useful for respiration in all living organisms. However, prolonged exposure to Cu has been associated with negative health implications. The use of pesticides by farmers is a major source of Cu contamination in rivers and lakes (Omoigberale *et al.* 2013; Krstić *et al.* 2018; Anifowose & Oyebode 2019).

The Fe concentration levels in the samples from District 2 were below the NSDWQ limit (Figure 2(b)). However, all the other districts had Fe levels above the NSDWQ limit. The Ekpoma community is characterized by high iron concentrations in the soil formation (Okeke *et al.* 2011; Tenebe *et al.* 2020) and this iron can be transported directly to the river, which ends up being harvested and stored in the tanks (Emenike *et al.* 2019). High iron concentrations in rivers have also been linked to iron concentrations in runoff from agricultural fields, as well as the leaching of corroded metallic objects (Anifowose & Oyebode 2019). Additionally, the collection time of water from the river and the hydrological mix can affect the concentration of pollutants in the collected water (Zhang *et al.* 2021, 2022).

In districts 2–4, no Al was detected (Figure 2) and the Al level in District 1 was well below the NSDWQ limit. The Zn concentration levels in all the districts were far below the US EPA and NSDWQ limits (Figure 2(d)), but the Cr level in Districts 1, 4, and 5 (Figure 2) was much higher than the US EPA and NSDWQ limits. Zn is a major component of the soil and rocks. Therefore, it is possible that soil formation near rivers is an important source of high Zn concentrations measured in stored river water samples. Other sources could include the dissolution of Zn from zinc-galvanized materials, especially because of the increased use of these materials in pre-urban settlements. The absence of Cr in samples from Districts 2 and 3 can be attributed to the nature of the storage tanks used in the districts. Unlike Districts 1, 4, and 5, which used concrete storage wells, Districts 2 and 3 used plastic storage tanks. Mn is considered an essential element in the human body; however, all the samples in the districts had Mn levels above the US EPA and NSDWQ limits. This implies that the concentration of Mn in water samples poses a potential risk to consumers. All samples from the five districts had Ni levels that exceeded the WHO and NSDWQ limits. Ni in the water samples tested can be linked to natural processes, such as chemical weathering of rocks and soil leaching since Ni forms a major constituent of rocks. The presence of Ni and Mn can also be associated with the overapplication of fertilizers in nearby farms that have been transported by precipitation events (Anifowose & Oyebode 2019; Tenebe *et al.* 2019a, 2019b).

3.3. Human health risk assessment

The results of the carcinogenic and non-carcinogenic risk assessments for all sampling points are shown in Figure 3 and Tables S4 and S5 (see Supplementary Material). As expected, children were the residents most affected by the PTMs quantified in river waters. The total non-carcinogenic risk (HI_{tot}) decreased by approximately 20% with each age group's advance until residents reached the adolescent stage at 16 years of age. From this age onward, the risks increased again by 30% and then stabilized. Almost all the calculated HI_{tot} values are derived from river water intake ($\geq$ 92%), the greatest risks for children can be explained by the ratio of water IR and BW (IR/BW) being higher than those of the adult age groups. The HI_{tot} values for the various age groups decreased in the order of $A > B > C > D > H > I > G > E > F$ (Figure 3).

The risk reduction up to age group F (16–18 years old) is evident in the non-carcinogenic risk values encountered at 22% of the sampling points. For localities 10, 12, 18, and 19, this age group was the only one with a negligible HI_{tot} value, while for the other inhabitants, the risks are low. Similarly, at sites 3, 38, 39, and 41, all age groups present medium risks, except for age group F which has low risks. Residents between 16 and 18 years old were also the only ones not to exhibit a high risk at sampling point (SP) 40, which proved to be the most harmful site in the studied area ($10.00 \geq HI_{tot} \geq 3.44$).

In addition to location 40, another 24% of the sampling points presented high risks for children aged between 1 and 11 years, with HI_{tot} values of up to 8.40 (SP 37, age group A). On the other hand, sites 1 and 2 showed medium risks for children up to 3 and 11 years old, respectively, while sites 7, 8, and 22 exhibited low risks for children up to 6 years old and negligible

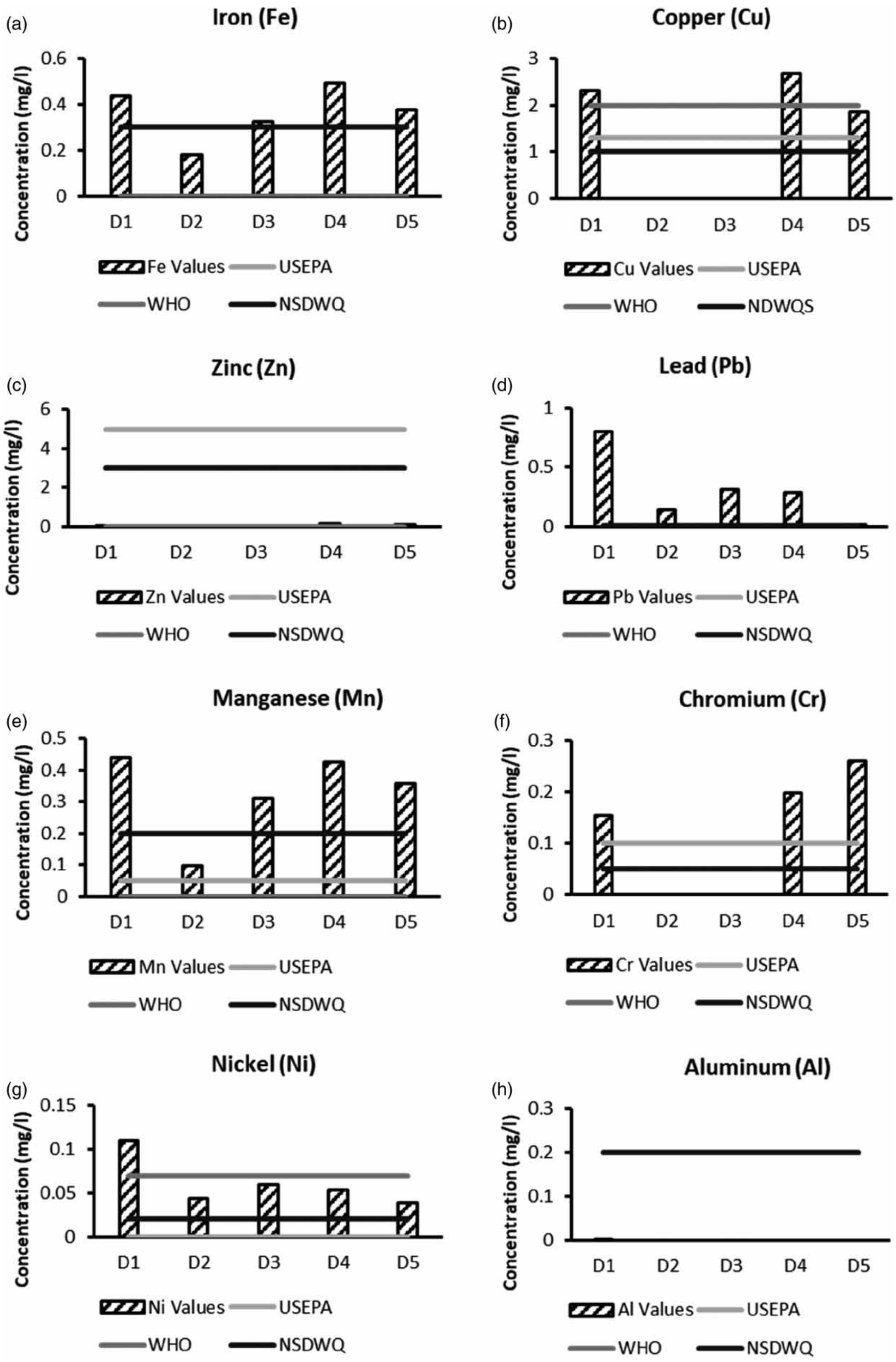


Figure 2 | (a–h) Graphical representation of PTM concentrations compared to regulatory standards (WHO, US EPA, NSDWQ).

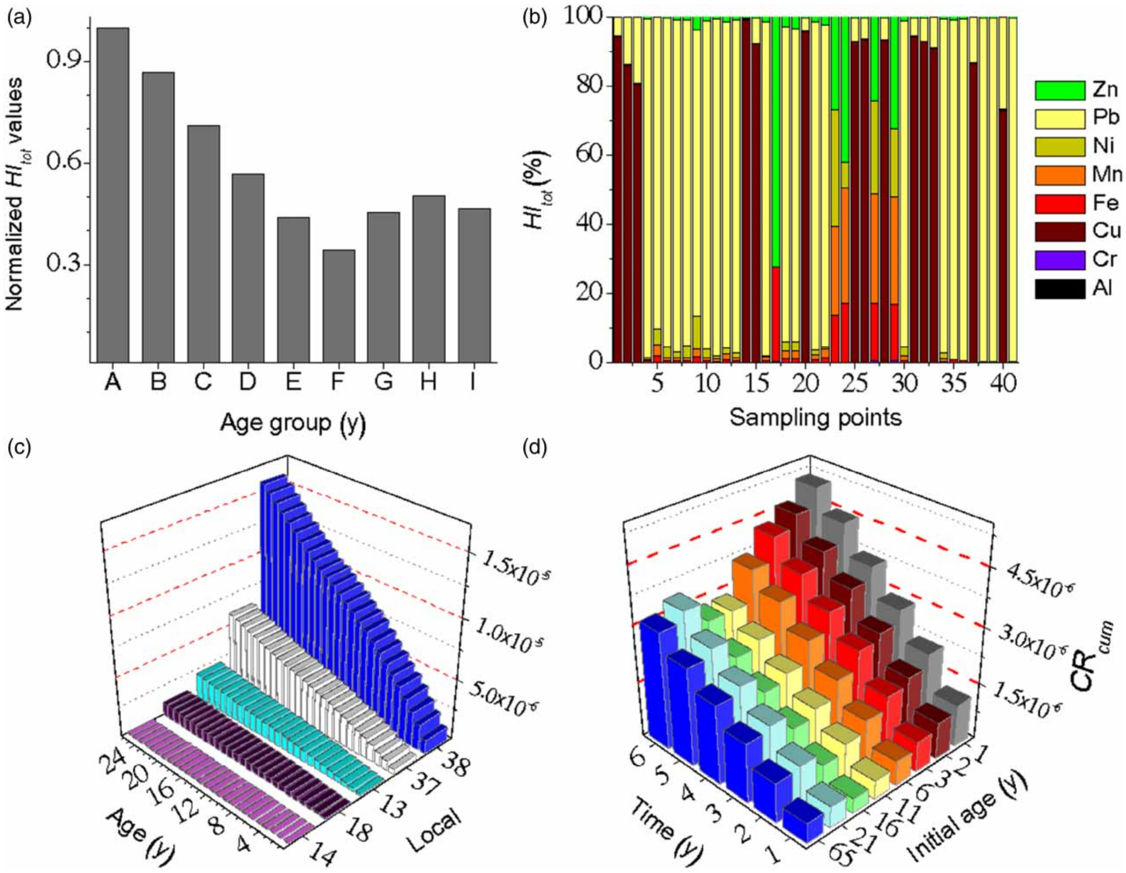


Figure 3 | (a) Normalized HI_{tot} values for each age group; (b) contributions of each chemical species for HI_{tot} values; (c) CR_{cum} spatiotemporal evaluation for five sites with different intensities of CR (IA = 1-year-old); and (d) comparison of spatiotemporal CR_{cum} evolution for different IA (SP 38).

HI_{tot} values for other residents. Among the locations, SP 17 proved to be the safest for the population ($HI_{tot} \leq 0.01$) and is part of the 17% of sites (SP 5, 9, 17, 23, 24, 27, and 29) that show negligible risks for any age group. Finally, the last 22% of sampling points presented low risks for any age group.

Analyzing the non-CR found for SP in each district, it was observed that none of the locations evaluated in the Ihumudumu and Ugoelen communities presented very significant risks ($HI_{tot} < 1.00$) for any age group. Of the 15 sites sampled in the Emando region, only 33% exhibited medium or high risks for all age groups, whereas 61% of the SP located in the Idumegbo district showed considerable hazards for some inhabitants. The only location sampled in the Ukpenu extension area (SP 14) presented medium or high HI_{tot} values (1.43–4.14) for the residents.

Among the PTMs analyzed, the contributions of Al and Cr to human health risks were negligible, representing less than 0.6% of the HI_{tot} values. In contrast, Cu and Pb contributed the most to the non-CR found at the sampling points (Figure 3(b)). Cu is responsible for 73.1–99.2% of the risks when present in concentrations greater than Quantification Limit (QL). In addition, all sites contaminated with Cu showed medium or high human health risks in some or all age groups. Similarly, Pb also presents extremely high contributions (up to 99.7%) when Cu is not present in the samples but drastically decreases in the presence of this PTM (0.5–26.4%). In this study, the chemical species Fe, Mn, Ni, and Zn did not contribute significantly to the HI_{tot} values, accounting for 2.1, 3.1, 4.6, and 3.6%, respectively. Some exceptions were localities 23, 24, 27, and 29, where the absence of Pb allowed these four PTMs to contribute between 7.3 and 72.2%, and location 9, whose Ni participation reached 9.5% even in the presence of Pb. Briefly, the decreasing chemical species mean contribution order for the HI_{tot} value is Cu > Pb > > Zn > Ni > Mn > Fe > Cr > Al.

The cumulative CR (CR_{cum}) calculated in this study was derived entirely from Pb, which is the only chemical species that presents carcinogenic effects by the oral route and is responsible for 98.8% of CR_{cum} values. For this reason, sites 17, 23, 24, 27, and 29, where Pb concentrations were below the QL, did not show CR to residents. Of the remaining 36 sampling points, the majority (69%) presented a low CR after 26 years of exposure to PTMs, independent of the residents' IAs, while only 22% exhibited negligible CR_{cum} values. At site 18, only residents whose contamination began at IAs of 1, 2, 3, 18, and 21 years old had a low probability of developing cancer during their lifetime, whereas, at locations 19 and 22, low CR was observed only for AIs of 65 and 1–2 years old, respectively. Site 38 presented the highest possibility for a resident to develop cancer over their lifetime, with an average probability of 1 in ~69,000 inhabitants. On the other hand, site 14 revealed a probability 148 times smaller (1 in ~10,200,000 residents) compared to location 38, which was the lowest among the sampling points with Pb concentrations above the QL.

The spatiotemporal risk assessment performed by HERisk showed that for residents of some locations, it took much less than 26 years of Pb exposure for CR_{cum} values to exceed the limit of 1.00×10^{-6}. When Pb ingestion began at IA of 1-year-old (Figure 3(c)), CR appeared even after 1 year of exposure to the contaminant at sites 38 and 40 ($CR_{cum} = 1.12 \times 10^{-6}$ and 1.04×10^{-6}, respectively). Similarly, at sampling points 39 and 41, it was necessary that residents drink the river water for only 2 years for the CR_{cum} values to be classified as low. At localities 37, 34, and 33, it was also necessary to have a few (3–5) years of exposure for the risks to no longer be negligible. Of the remaining locations with significant CR, 41% required 5–10 years of exposure, and the other 35% required more than 10 years for the probability of residents developing cancer during their lifetime to be greater than 1 in 1,000,000 inhabitants.

The spatiotemporal evaluation also revealed that at site 38 (highest CR), CR_{cum} values were higher than 1.00×10^{-6} after 1 year of exposure to Pb species only when contamination started at IA of 1-year-old (Figure 3(d)). For IAs of 2, 3, 6, 18, 21, and 65 years old, the inhabitants need to drink water from the river for 2 years to reach low CR, while for the other two AIs (11 and 16 years old), 3 years of oral exposure to Pb are required. These results suggest that the water from the Ibiekuma stream cannot be directly ingested by Ekpoma residents, and that purification treatments are necessary to make the water potable and free from risks to human health. PTM poisoning is known to harm the skeletal, neurological, endocrine, and immunological systems of humans when they are repeatedly exposed to PTMs through oral intake, cutaneous contact, and inhalation (Yang *et al.* 2022).

4. CONCLUSION

Environmental pollution is on the rise owing to the rapid agricultural and industrial development worldwide. Many rivers are affected by various degrees of PTM pollution. These metals are distinguished by their high stability, non-degradability, and continuous accumulation, which can cause irreversible damage to the surrounding environment. When the quantities of PTMs present in the samples examined were compared to the current standards, it was discovered that approximately 61.3% of the samples tested had PTM contamination levels above the permitted limits for these elements in drinking water. The US EPA considers Pb to be a carcinogenic element, and high levels of this element were found in the water samples analyzed. Cu concentrations were measured in some of the water samples examined in Districts 1, 4, and 5, and high concentrations are known to cause Cu toxicity when consumed. These findings suggest that long-term drinking of water may increase the risk of acquiring cancer and other ailments, such as liver damage, fatigue, insomnia, arthritis, hallucinations, appetite loss, headaches, stomach discomfort, renal dysfunction, hypertension, and vertigo. Environmental pollutants, such as organic contaminants in poorly maintained storage tankers and wells, may also have an impact on the stored water quality after retrieval from the river. Therefore, it is suggested that regulatory bodies or governments at all levels should enforce quality checks for tankers and storage wells as a part of BMPs. For tankers, quality checks should include but not be limited to non-toxic internal coating, cleaning before and after trips, and the use of non-toxic water collection accessories. Inspection of tankers should also be encouraged at collection points. For the storage wells, regulatory bodies should conduct inspections to ensure that the dimensions and quality of materials used in the construction of storage wells are regulated, as well as encourage proper maintenance practices.

According to the human health risk assessment, children are the most susceptible to the carcinogenic and non-CR associated with PTMs, with the age group between 1 and 2 years being the most affected. As children grow, HI_{tot} values decrease considerably until they reach the adult stage at 18 years old. The PTMs that contributed the most to the non-CR were Cu and Pb, while for the CR, Pb was the only contributor. The water ingestion route is accountable for almost all risks (91.8–99.2%). Site 38 showed the highest CR_{cum}, with a probability of residents developing cancer during a lifetime of 1 in approximately 69,000. The CR_{cum} spatiotemporal evaluation indicated that depending on the local and the residents' IAs, the CR can be significant even after 1 year of drinking Ibiekuma stream water.

Consumers in Ekpoma may be exposed to a variety of health risks, such as cancer and other diseases, because of their continued drinking of contaminated water from rivers. As a result, it is suggested that current river collection systems be enhanced to collect clean and high-quality water by ensuring adequate treatment methods before storage. This promotes the removal of organic, inorganic, and microbial contaminants, which may impair water quality. It is also recommended that covered storage tanks made of leaching-resistant materials should be used. These suggestions will almost certainly reduce the health risks for users who rely entirely on river water. BMPs such as constructed wetlands, detention ponds, vegetative filters, grazing management, and streambank stabilization should be implemented as cost-effective preventive measures for controlling the pollution of the river body. Also, continuous water quality monitoring, public education, and sensitization, using BMPs that meet regulatory requirements, monitoring and controlling illicit discharge and disposal into the river, proper disposal of household hazardous wastes, collection, and treatment of runoff before discharge into a surface water body will ensure water quality protection in the region. It is suggested that the microbial quality of the stored river water is investigated to ascertain the appropriate remediation and treatment strategies. Systems to optimize collection and storage processes to reduce contamination at collection points should be considered as areas for future research. Further studies can investigate the impact of runoff on the river water quality before it is collected, transported, and stored to monitor and control pollution at the source. Future research should also focus on investigating common water-related illnesses linked to metal consumption by collecting inpatient and outpatient health information in the Ekpoma community and determining the correlative relationship with river water consumption.

DATA AVAILABILITY STATEMENT

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

CONFLICT OF INTEREST

The authors declare there is no conflict.

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