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Distribution, contamination and ecotoxicological risk assessment of trace metals in stream sediments of Mari area, Betare-Oya gold district (Neoproterozoic Fold Belt)

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Abstract

Mari area is located within the pull apart syn-depositional Neoproterozoic volcano-sedimentary basins in Cameroon. The main aim of this study was to evaluate sources, contamination and ecotoxicological risk of trace metals in sediments of Mari area. Stream sediment samples were collected and analyzed using inductive couple plasma mass spectrometry (ICP-MS). Sediment samples show high content in Mn (868 - 2950 ppm), Pb (20.90 – 2950 ppm), Th (50.70 – 200 ppm), Zn (77 – 255 ppm), Cu (8.60 – 222 ppm) and depleted in the rest of trace elements. The contamination index (CI: 169.07 - 671.38), modified contamination index (mCI: 12.08 - 47.96), pollution load index (PLI: 0.94 - 3.21), nemerow integrated pollution load index (NIPI: 0.15 - 296), geo-accumulation index (Igeo: -5.32-21.76), potential environmental risk factor (Er: 0.14 - 333.33) and ecotoxicological risk of trace metals indicate sediments contamination and metal pollution of the ecosystem. Sediments exhibits high probability toxicity index for trace metals and have biological effect on biota. The lithology, mining activities, agricultural practices and domestic activities are responsible for sediments contamination and metal pollution in Mari area. Constant monitoring and implementation of environmental laws and regulation in mining areas is of great important to protect the environment and biota for sustainable development.

Keywords: Mari area, neoproterozoic, trace metals, stream sediment, contamination, ecotoxicological risk

Introduction

Environmental contamination is a serious challenge in recent years due to industrialization, urbanization and agricultural practices. Rivers and streams in mining communities in Cameroon are under serious threat due to an exponential increase urbanization, industrialization and intensive agricultural practices which have a propensity for large amounts of waste and toxic metals discharged into them, resulting in sediments contamination and metal pollution. The significant environmental issue that affects human health, others biotas and marine life is the contamination of sediments with trace metals. Trace metals if released into aquatic environment with low solubility have a tendency to settle in the bottom sediments (Briffia *et al*., 2020) [6], potentially becoming a source of harmful metals to the water column. The major source of trace metals in aquatic ecosystem is sediment (Zahra *et al*., 2014) [43]. Sediments release metals in the water ecosystem by acting as sink and source for those metals (Fernandes and Nayak, 2012) [10]. The chemical composition, the grain size and the total organic matter content of sediments have an impact on the distribution of trace metals within them (Ali *et al*., 2018) [3]. Sediments is widely used as an environmental index to assess metal contamination in the water ecosystem (Islam, 2015) [16]. They offer useful data regarding the level of environmental contamination and pollution (Rahman and Islam 2010) [34]. Natural sources of metals include weathering, volcanism, rock, erosion, forest fires, and volcanic eruptions. Anthropogenic input includes agricultural activities, electronic waste, municipal and household waste, sewage, agrochemicals and mining operations (Lemnyuy *et al*., 2022; Mandeng *et al*., 2019; Noa Tang *et al*., 2021) [18, 23, 29]. Mining therefore has great consequences to the environment and is responsible for the release of massive amounts of hazardous metals into the surrounding environments (Ahmad *et al*., 2014; Ameh *et al*., 2014) [1, 2]. Trace metals, with their immutable characteristics of non-biodegradability, toxicity and bioaccumulation, have a negative impact on the health of humans, as well as ecosystems and therefore, their occurrence in streams and rivers needs to be strictly controlled (Liu *et al.*, 2016) [25]. Also, metal pollutants pose potential harmful effects on human health and the whole ecosystem (Wei *et al*., 2016) [42]. Mining and agricultural activities constitute a major source of income for the local population in the Mari area that they cannot do without. However, the negative effect of these mining activities and agricultural practices remains a major challenge today in the study area as hazardous substances are been discharge into the water ecosystem which is detrimental to humans and biota. Mining and agricultural practices has been on going in the Mari area for decades and this area requires investigation to identify the negative impact of mining and agricultural activities. The main aim of this research is to evaluate the sources, contamination and ecotoxicological risk of trace metals in sediments of Mari area.

Geologic setting

Regional geology

The Pan-African orogeny in Central African include the Central African Fold Belt, which traverses a sizeable portion of Nigeria, Chad, Central African Republic, and Cameroon and extends eastwards to Sudan and Uganda, is part of the Pan-African orogeny in central Africa (Toteu *et al.*, 2006)

[39]. The Trans-Saharan Belt in western Africa and the Brasiliano Orogen in northeastern Brazil are both important Neoproterozoic Orogen connected by the Pan-African fold belt (Castaing *et al*., 1994; Neves *et al*., 2006; Fig. 1) [7, 28]. The Pan-African fold belt in Cameroon also called Neoproterozoic fold belt or North-Equatorial fold belt (Poidevin, 1983; Nzenti *et al*., 1988) [33, 30] is made up of three structural units (Toteu *et al*., 2004) [38]: (i) the Western Cameroon domain (WCD) which is situated west of the Tcholliré–Banyo shear zone (Fig 1, TBSZ). Rocks of the Western Cameroon domain are interpreted as products of deposition in an opening back arc basin between 830 and 665 Ma (Penaye *et al*., 2006; Toteu *et al*., 2006; Bouyo *et al*., 2015) [31, 39, 5], (ii) The Yaoundé domain (YD) is a syntectonic basin, U–Pb dating of detrital zircons revealed a depositional age of ca. 625 Ma (Fig 1; Toteu *et al*., 2006) [39]. The metasediments consist of epicontinental deposits and were deposited to a passive margin (Nzenti *et al*., 1988) [30], (iii) The Adamawa Yade domain (AYD), where the study area is found (Fig. 1) is situated between the Tcholliré-Banyo fault and Sanaga fault. This domain is made up of variety of granitoids, these granitoids placement is governed by the tectonic regime 610 – 585 Ma and the 640 - 610 Ma crustal thickening phase (Fozing *et al*., 2021) [11]. According to the reconstruction of pre- drift Gondwana, the AYD is characterized by significant NE-striking transcurrent shear zones that are thought to be extensions of major shear zones of northeast Brazil (Castaing *et al*., 1994) [7].

Local geology

The Mari area in the Betare-Oya gold district is located in the Lom basin which is a pull apart Neoproterozoic syn depositional basin in Cameroon (Ngako *et al*., 2003) [27]. This pull-apart basin economic potential, particularly gold, is accounted for by the reworking and remobilization of an Archean to Paleoproterozoic basement (Toteu *et al*., 2004) [38]. The Lom basin is composed mainly of meta-volcanic and meta-sedimentary rocks (Ngako *et al*., 2003) [27]. In this Neoproterozoic basin, meta-tuffs, volcanoclastic rocks and sedimentary rocks make up the bulk of the lithology (Toteu *et al*., 2004, 2006) [38, 39]. The gold bearing quartz veins of the Lom basin are spatially connected to P and P' of the Riedel model and were created during transpressional and transtensional processes (Fils *et al*., 2020) [12]. The majority of mineralized zones in the Lom basin were quartz veins and surrounding altered wall rocks which are frequently made up of quartz, carbonates, chlorite, muscovite and sulphides (Azeuda Ndonfack *et al.*, 2021) [4]. The Lom basin is characterized by intensive artisanal and semi-mechanized alluvial gold mining. Primary gold in the Betare-Oya district occur as dissemination hosted in quartz veins that truncate meta-sedimentary rocks in the vicinity of small local granitoid intrusions (Vishiti *et al*., 2017) [41]. Apart from mining activities, agricultural practices and hunting are visible in the mining communities for their livelihood.

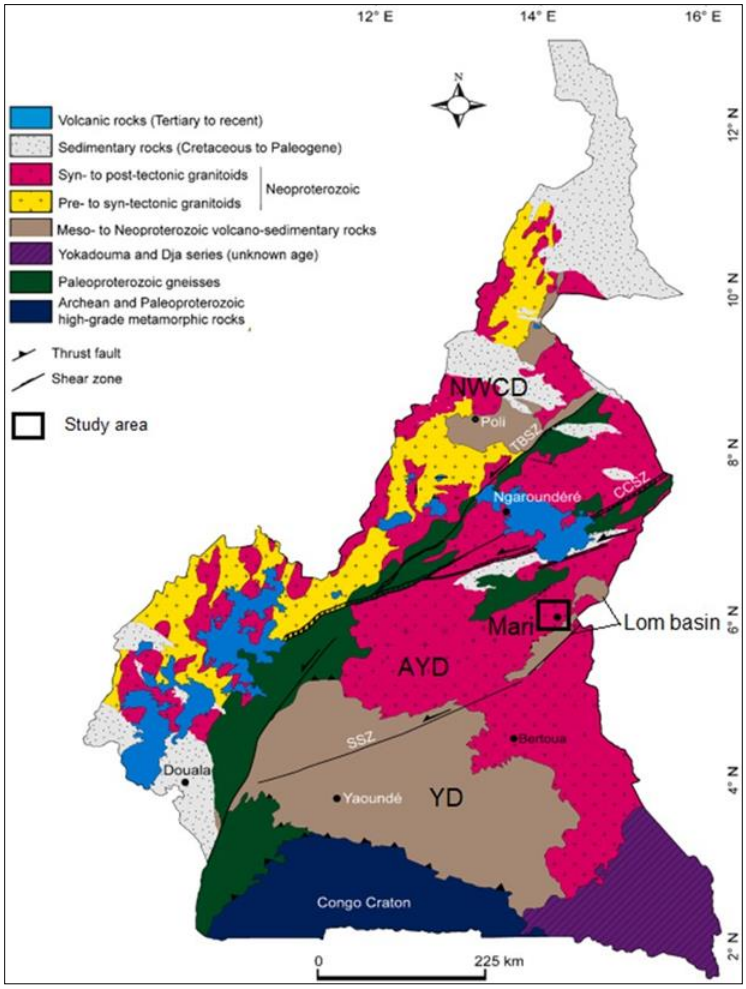

Fig 1: Geological map of Cameroon showing the three structural domains of the Central African fold (Azeuda Ndonfack *et al.*, 2021) [4]. NWCD: Northern western Cameroon Domain, YD: Yaoundé Domain; AYD: Adamawa Yade Domain.

Materials and methods

Twenty (20) representative stream sediment samples were collected during the fieldwork in the study area. In Mari area, stream sediment samples were collected from both active and abandoned mines. To avoid humus content, sediments were collected using a hand-held digger from a depth of 0.20 to 85 cm. Chemical analysis was done using the Inductive Coupled Plasma Mass Spectrometry (IC-PMS) to obtain the concentrations of trace elements at Activation Laboratories in Ancaster, Canada. Stream sediment samples were subjected to pre-analysis preparation by drying them (60°) and sieving (177µm). 0.5g of sample is then digested in aqua regia (a combination of HCl and nitric acid to leach sulphides, some oxides and silicates) at 90° in a microprocessor-controlled digestion block for 2 hours. Then the digested samples are diluted and analyzed by a Perkin Elmer Sciex ELAN 6000, 6100/9000. ICP-MS represents the most sensitive and flexible method for the analysis of mass fractions of trace elements in geo materials (rocks, mineral, water). Its advantage is in its nearly complete ionization of many elements achieved by the use of argon plasma at temperature of up to 7000°C and fast data acquisition. To guarantee the accuracy of the findings, precautions and quality assurance practices were implemented. In the laboratory, the experiment was conducted using deionized wate, analytical-grade chemicals, and proffessionally cleaned and rinsed glassware.

Results and discussion

Trace metals distribution

Table 1 displays the studied trace metal contents in sediments from the Mari area. The sediments had significant concentrations of Mn (868 - 2950 ppm, av. = 1836 ppm), Pb (20.90 – 2950 ppm, av. = 200.88 ppm), Th (50.70 – 200 ppm, av. = 167 ppm), Zn (77 – 255 ppm, av. = 118.31 ppm) and Cu (8.60 – 222 ppm, av. = 31.94 ppm) compared to the value of the Upper Continental Crust by Rudnick and Gao (2003) [35]. Hg (4.00 – 10 ppm), U (4.10 - 45.50 ppm) display moderate values. The rest of elements such as Cd (0.010– 0.47 ppm), Fe (2.77 – 14.90 ppm), and Ni (1.70 – 34.90 ppm) exhibit low contents when compared to the standard. The high content of Cu, Mn, Pb, Th and Zn could be attributed to severe weathering of the parent rocks, ore and metals processing, agricultural activities (application of phosphate fertilizers, usage of herbicides, fungicides, pesticides) and exhausts emissions in Mari area (Tehna *et al*., 2019; Manga *et al*., 2017; Doumo *et al*., 2022; Lemnyuy *et al*., 2022; Ndema Mbongué *et al*., 2023) [37, 20, 8, 18, 26]. The leaching of the source rocks during weathering is the cause of low mean level of other metals. According to the findings, high metal concentrations in sediments from Mari area may have a deleterious impact on aquatic ecosystems and human health through the food chain. The computed standard deviation values were high due to the concentration of trace metals with high variable heterogeneity in the study area. The high variance of Cr (302.42), Cu (2466.65), Mn (269534.52), Pb (419997.28), Th (2380.79) and Zn (1636.73) indicates a non-homogeneous variability of trace metals and clearly indicating that the possible human-related influence.

Table 1: Composition of trace metals in stream sediments of Mari area

Sample	Cd	Co	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Sc	Th	U	Zn	Zr
MA1	0.06	11.3	18	18.6	4.16	10	2740	4.6	24.4	2.5	93.5	8.4	151	31.2
MA4	0.03	5.1	12	12.4	3.43	5.19	1630	3.3	28.7	3	200	21	108	3.2
MA5	0.05	4.9	21	14	4.03	10	2250	3.5	43	2.2	173	14.3	133	9.4
MA6	0.03	4.8	22	11.4	3.88	10	2170	3.5	30.2	3	194	16.1	89.4	17.3
MA7	0.07	5.4	17	25.3	4.26	7.64	2950	3.9	21.3	2.4	50.7	4.1	153	20
MA8	0.01	4.5	15	15.8	4.06	5.11	1900	4.2	20.9	2.8	97.3	8.9	89	14.4
MA9	0.03	11.4	21	25	3.85	5.11	1850	6.1	44.4	2.3	167	13.6	102	8.5
MA10	0.09	3.9	21	11.1	3.19	8.13	1530	4.3	32.1	2.7	200	27.1	86.8	4.2
MA11	0.02	2.6	11	8.6	3.32	5.49	1630	2.1	137	2.4	200	23.6	114	3.3
MA12	0.09	3.1	17	17.1	4.03	10	2090	2.7	50.4	2.1	200	21.7	146	10.1
MA13	0.06	3.9	22	107	4.07	4.36	2010	3.8	84.6	2.3	200	17.5	126	5.7
MA14	0.07	2.6	21	13.3	3.12	10	1550	2.2	46.9	2.8	200	18.9	125	6.6
MA18	0.04	2.9	23	14.1	3.37	9.86	914	3.6	43.2	2.2	200	45.5	88.9	1.1
MA19	0.02	3.2	54	16.8	4.18	7.45	868	5.1	100	3.1	200	34.7	77	1.9
MA22	0.03	3.8	32	44.6	2.77	4	1790	2.6	71	3.9	99.2	7	93.6	16
MA23	0.09	4.3	13	11.9	3.34	8.36	1720	1.7	56	1.9	200	22.4	147	8.8
MA24	0.02	4	17	12.8	4.58	7.67	2340	3.7	22.6	3.1	103	8.2	93.8	18.6
MA26	0.47	16.6	85	222	14.9	10	1240	34.9	2950	1.5	200	18	84.6	3.9
MA28	0.21	3.3	41	12.6	3.49	4.09	1750	3	66.9	1.9	200	18.6	103	6.8
MA29	0.04	4	17	24.4	3.89	10	1800	3.4	144	1.9	171	14	255	10
Min	0.01	2.60	11.00	8.60	2.77	4.00	868.00	1.70	20.90	1.50	50.70	4.10	77.00	1.10
Max	0.47	16.60	85.00	222.00	14.90	10.00	2950.00	34.90	2950.00	3.90	200.00	45.50	255.00	31.20
Mean	0.08	5.28	25.00	31.94	4.30	7.62	1836.10	5.11	200.88	2.50	167.44	18.18	118.31	10.05
Variance	0.01	12.95	302.42	2466.65	6.44	5.51	269534.52	50.25	419997.28	0.31	2380.79	96.32	1636.73	57.01
St Dev	0.10	3.60	17.39	49.67	2.54	2.35	519.17	7.09	648.07	0.56	48.79	9.81	40.46	7.55

Trace Metal Contamination

Contamination index (CI), modified contamination index (mCI), pollution load index (PLI), nemerow integrated pollution load index (NIPI), Enrichment factor (EF) and Geo-accumulation index (Igeo)

The sum of the contamination factors for various heavy metals is known as contamination index (CI) after Hakanson (1980) [15].

$$CI = \sum_{i=1}^{8} CF$$

Where C_I less than 6 denotes low contamination degree; 6 < C_I < 12 suggests moderate contamination degree; 12 < CI < 24 indicates contamination degree and CI > 24 signals significant contamination degree. The computed contamination index (CI) of the stream sediments is presented in Table 2. The CI varies from 169.07 (in sample MA22) to 671.38 (in sample MA26). The stream sediments generally indicate high contamination degree (CI > 24) and it is attributed to high anthropogenic input sources within the Mari area.

The modified contamination index (mCI) aids in determining how heavily contaminated the sediments samples are overall heavy metal. It is determined as:

$$mC_i = \frac{\sum CF}{n}$$

In which n is the number of elements that were analyzed and CF is the contamination factor of those elements (Hakanson 1980). mCI < 1.5, nil to low; 1.5 < mCI < 2, low; 2 < mCI < 4, moderate; 4 < mCI < 8, high; 8 < mCI < 16, very high; 16 < mCI < 32 extremely high and mCI > 32, ultra-high contamination. Modified contamination index (mCI) varies between 12.08 (MA22) to 47.96 (MA26; Table 2). Sample MA8, MA9, MA13, MA22 and MA28 show very high contamination (mCI < 16) comprising 25%; samples MA1, MA4, MA5, MA6, MA7, MA10, MA11, MA12, MA14, MA18, MA19, MA23, MA24 and MA29 are in the class of extremely contaminated (mCI = 16 - 32) explaining 70%; sample MA26 is ultra-highly contaminated (mCI > 32) explaining for 5% of the total samples. This indicates a significant human activities metal input into stream sediments and due attention should be paid to monitoring metals entering the various environmental compartments due to mining activities and agricultural practices in the study area (Mimba *et al*., 2018; Kamga *et al*., 2017; Tehna *et al*., 2019; Lemnyuy *et al*., 2022) [24, 17, 37, 18].

The pollution load index (PLI) serves to determine the cumulative pollution effect at various constitutes in sediments at different locations (Tomlinson 1980) [40].

$$PLI = (CF_1 \times CF_2 \times . \times CF_n)^{1/n}$$

PLI > 1 implies pollution; PLI < 1, shows no pollution and PLI = 1, denotes perfection. Pollution load index (PLI) across Mari area is displays in Table 2. PLI ranges from 0.94 (in sample MA8) to 3.21 (in sample MA26). Sample MA4, MA9 and MA18 present no pollution (PLI >1), accounting for 15%, while the other samples are highly polluted (PLI > 1) explaining 85% of the Mari area. The heavily pollution levels of PLI in samples is linked to the impacts of anthropogenic activities (mining operations).

The nemerow integrated pollution load index (NIPI) evaluates amount of metal pollution and determine the impact of various metals on the sediments and environmental quality. According to Gao *et al*. (2016) [13], the index is calculated using the following formula:

$$NIPI = \frac{\sqrt{\left(\frac{1}{m}\sum_{i=1}^{m} P_i\right)^2 + (P_{imax})^2}}{2}$$

NIPI < 0.7, denotes unpolluted (Clean), 0.7 ≤ NIPI < 1.0, indicates little pollution (warning limit); 1.0 ≤ NIPI < 2.0, implies slight pollution (light pollution); 2.0 ≤ NIPI < 3.0, indicates moderate pollution (medium pollution); NIPI ≥ 3.0, implies serious pollution heavy/high pollution. The nemerow integrated pollution load index (NIPI) values for Mari area are in the trend: Hg > Pb > Th > U > Cu > Cd > Zn > Fe > Mn >Zr > Co >Cr > Ni >Sc (Table 2). It can be observed that the stream sediments are enriched in Cr, Co, Ni and Sc (NIPI < 0.7) are unpolluted; sediments enriched in Fe, Mn, Zn, and Zr (NIPI = 1) are slightly polluted whereas samples enriched in Cd, Cu, Hg, Pb, Th and U (NIPI ≥ 3.0) are heavily polluted. The high level of NIPI (Fig 3) values measured probably resulted from the various industrial and anthropogenic activities. It is believed that the presence of trace metals in sediments could be traced to mining activities and agricultural practices (Mimba *et al*., 2018; Kamga *et al*., 2017; Tehna *et al*., 2019; Lemnyuy *et al*., 2022) [24, 17, 37, 18].

Enrichment factor is used to assess the level of heavy metals contamination in sediments. According to Tariq *et al*. (2018) [36], it is expressed as follows:

$$EF = me/Fe_2O_3)_{sample} / (me/Fe_2O_3)_{sample\ background}$$

The enrichment factors are displayed in Fig 2a. Hg (716.88), Pb (120.77) and Th (76.81) have the highest EF value. In every Samples do not exhibits any enrichment (EF < 1) in Cd, Co, Cr, Cu, Mn, Ni, Zn and Zr. Zn is in the category of minimal enriched (EF < 2) in the samples. Hg, Pb, Th and U are in the class of extreme enrichment (EF = 20 - 40). Anthropogenic sources such as mining activities, agrochemicals, and agricultural practices are responsible for the severely enrichment of several trace metals in the studied area. High Hg enrichment is likely due to the processing of the metal ores, identified as one of the anthropogenic activities responsible for the accumulation of Hg in the area. Pb enrichemnt could be derived from the oxidation and dissoluion of sulphide minerals (galena) at the mining sites (Matias, 2008; Mimba *et al*., 2018; Doumo *et al*., 2022) [21, 24, 8] while Th and U coud be attributed to pesticides, fingicides, agrochemicals and phosphate fertilizers usage by the local population for high crop yeild. These findings suggest that sediemnts is more likely to contain reasonable amounts of trace metals.

The geo-accumulation index (Igeo) is developed to measure the amount of metal accumulation in sediments.

$$Igeo = Log_2 \left(\frac{Cn}{1.5 \times Bn}\right)$$

Where, Cn refers to the measured metal ion in samples; Bn represents to element's background content and 1.5 is the background matrix correction factor resulting from lithogenic effects (Rudnick and Gao, 2003) [35]. The Geo-accumulation index (Igeo) shows a variation from -5.32 (Mn) to 21.76 (Mn; Fig 2b). Cd in the Mari area show Igeo values less than zero which was practically unpolluted. This suggests that anthropogenic Cd pollution has no effects on the sediments and that sediments levels are at background levels. Meanwhile, sediments enriched in Fe and U are strongly polluted (Igeo = 3 - 4), while sediments with high Sc content are moderately to severely polluted (Igeo = 4 - 5). Moreover, highly polluted (Igeo > 5) sediments are

enriched in Th, Zn, Hg, Cr, Cu, Co, Mn, Ni, Pb, and Zr. It is assumed that the primary human activities responsible for the study area's Hg, Cu, Zn and Pb pollution is the processing of the metal ores, while intense alteration, agricultural activities and other anthropogenic inputs are responsible for pollution Co, Cr, Mn, Ni, Th and Zr.

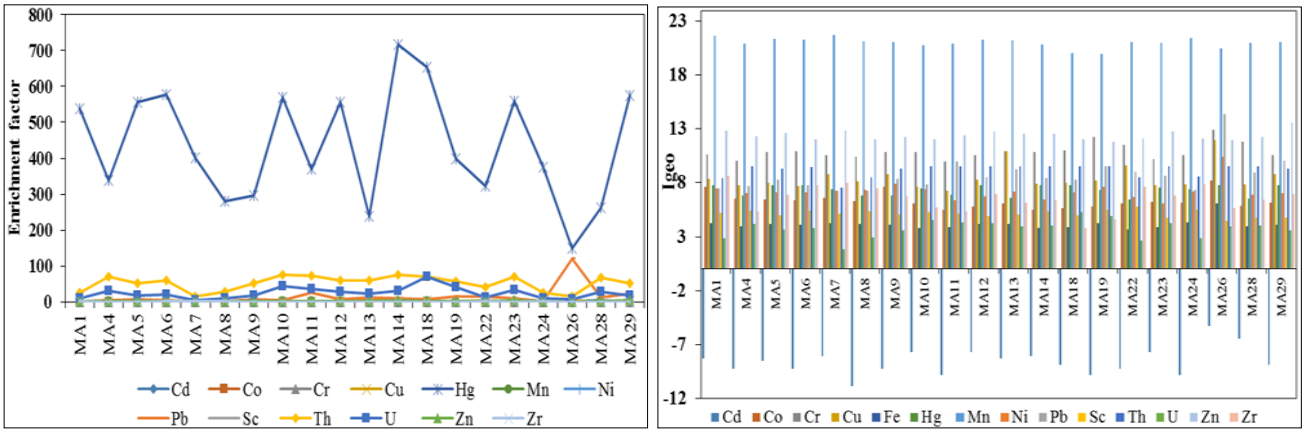

Fig 2: Pollution indices a) Enrichment factor (EF), and b) Geo-accumulation index (Igeo) of metals in sediments of Mari area

Assessment of potential ecological risks

The Environmental risk factor (Er) is used to quantify possible ecological risks connected to a certain specific pollutant (Hakanson *et al*., 1980) [15]. It is obtained by the formula:

$ErF = Tr^i \times CF^i$

Where Tr^i is the hazardous response factor of a metal (Pb = 5, Zn = 5, Cd = 10, Cr = 2, Ni = 5, Hg = 40, Cu = 5) and CF^i is the contamination factor. Er < 40, indicates low potential environmental risk; Er = 40 - 80 denotes moderate potential environmental risk; Er = 80 - 160, implies considerable potential environmental risk; Er = 160 - 320, indicates high potential environment risk and Er ≥ 320 denotes very high environmental risk. The Er varies from 0.14 (Ni) in sample MA23 to 1333.33 (Hg), in samples MA5, MA6, MA12, MA14, MA26 and MA29 (Table 3). Low Er (Er < 40) were present in sediments for Cd, Co, Cu, Cr, Ni, Mn Pb and Zn.This suggests that the environment may have less of an impact on living biota. Mari area generally exhibits moderate Er (Er: 40 - 320) for Pb (MA11, MA19, MA29) and Cd (MA28). Cd (in sample MA26) has high potential Er (Er = 160 - 320), while Hg and Pb (MA26) exhibits a very high potential Er (Er ≥ 320). Typically, the possible Er for Pb ranged from moderate to very high potential environmental risk factor (Er = 40 > 320) reflecting oxidation and dissolution of sulphides minerals (Matias, 2008; Mimba *et al*., 2018; Doumo *et al*., 2022) [21, 24, 8]. Due to Hg use by local miners and companies in the study area in the process of separating gold from sediment concentrate, Hg has a very high environmental risk factor. Research demonstrated both anthropogenic and natural pressure in the Mari area can have an impact on the release and bioavailability of these metals.

The Potential ecological risk index (PERI) accounts for cumulative impacts of the metals under consideration. It is the sum of all Er variables in a given study area (Hakanson *et al*., 1980) [15].

$PERI = (Er_1 + Er_2 + Er_3 \ldots + Er_n)$.

Where n is the number of elements under study and Er is the environmental risk factor. The results of potential ecological risk index (PERI) are presented in Table 3. PERI in the studied samples follows the trend: Hg >Pb > Cd > Cu > Zn > Mn > Ni > Co > Cr with values of 203280, 1826, 573, 188, 32.86, 20.40, 8.66, 7.94 and 7.41 respectively. Cr, Cu, Co, Mn, Ni and Zn shows low ecological risk index whereas Cd is in the category of considerable ecological risk index. In the study area, Hg and Pb displayed extremely high ecological risk indices. This suggests a higher danger of Cd, Hg and Pb contamination in the Mari area. This could be attributed to oxidation and hydrolysis of sulphides minerals from ore body rich in Cd and Pb (Matias, 2008; Mimba *et al*., 2018; Doumo *et al*., 2022) [21, 24, 8] and Hg used in gold separation from sediments concentrtate after panining. This result indicates anthropogenic metal input in the aquatic systems from different mining goldfields in Cameroon and the world.

Table 2: Contamination index (CI), modified contamination index (mCI), pollution load index (PLI) and newmerow integrated pollution load index (NIPI) of trace metals in sediments of Mari area

Sample	Cd	Co	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Sc	Th	U	Zn	Zr	PLI	Cd	mCd
MA1	0.75	0.42	0.13	0.69	0.62	333.33	1.52	0.08	2.22	0.11	16.70	6.46	2.10	1.64	1.35	366.78	26.20
MA4	0.38	0.19	0.09	0.46	0.51	173.00	0.91	0.06	2.61	0.14	35.71	16.15	1.50	0.17	0.97	231.87	16.56
MA5	0.63	0.18	0.16	0.52	0.60	333.33	1.25	0.06	3.91	0.10	30.89	11.00	1.85	0.49	1.22	384.97	27.50
MA6	0.38	0.18	0.16	0.42	0.58	333.33	1.21	0.06	2.75	0.14	34.64	12.38	1.24	0.91	1.19	388.38	27.74
MA7	0.88	0.20	0.13	0.94	0.63	254.67	1.64	0.07	1.94	0.11	9.05	3.15	2.13	1.05	1.12	276.58	19.76
MA8	0.13	0.17	0.11	0.59	0.61	170.33	1.06	0.07	1.90	0.13	17.38	6.85	1.24	0.76	0.91	201.30	14.38
MA9	0.38	0.43	0.16	0.93	0.57	170.33	1.03	0.10	4.04	0.11	29.82	10.46	1.42	0.45	1.24	220.21	15.73
MA10	1.13	0.15	0.16	0.41	0.48	271.00	0.85	0.07	2.92	0.12	35.71	20.85	1.21	0.22	1.14	335.27	23.95
MA11	0.25	0.10	0.08	0.32	0.49	183.00	0.91	0.04	12.45	0.11	35.71	18.15	1.58	0.17	0.94	253.37	18.10
MA12	1.13	0.12	0.13	0.63	0.60	333.33	1.16	0.05	4.58	0.10	35.71	16.69	2.03	0.53	1.28	396.79	28.34

MA13	0.75	0.15	0.16	3.96	0.61	145.33	1.12	0.06	7.69	0.11	35.71	13.46	1.75	0.30	1.38	211.17	15.08
MA14	0.88	0.10	0.16	0.49	0.46	333.33	0.86	0.04	4.26	0.13	35.71	14.54	1.74	0.35	1.13	393.05	28.07
MA18	0.50	0.11	0.17	0.52	0.50	328.67	0.51	0.06	3.93	0.10	35.71	35.00	1.23	0.06	0.99	407.07	29.08
MA19	0.25	0.12	0.40	0.62	0.62	248.33	0.48	0.09	9.09	0.14	35.71	26.69	1.07	0.10	1.14	323.73	23.12
MA22	0.38	0.14	0.24	1.65	0.41	133.33	0.99	0.04	6.45	0.18	17.71	5.38	1.30	0.84	1.14	169.07	12.08
MA23	1.13	0.16	0.10	0.44	0.50	278.67	0.96	0.03	5.09	0.09	35.71	17.23	2.04	0.46	1.16	342.60	24.47
MA24	0.25	0.15	0.13	0.47	0.68	255.67	1.30	0.06	2.05	0.14	18.39	6.31	1.30	0.98	1.02	287.89	20.56
MA26	5.88	0.62	0.63	8.22	2.22	333.33	0.69	0.59	268.18	0.07	35.71	13.85	1.18	0.21	3.21	671.38	47.96
MA28	2.63	0.12	0.30	0.47	0.52	136.33	0.97	0.05	6.08	0.09	35.71	14.31	1.43	0.36	1.25	199.37	14.24
MA29	0.50	0.15	0.13	0.90	0.58	333.33	1.00	0.06	13.09	0.09	30.54	10.77	3.54	0.53	1.35	395.20	28.23
NIPI	4.21	0.46	0.46	5.87	1.63	296.38	1.37	0.42	190.07	0.15	32.94	26.65	2.76	1.22			

Table 3: Environmental risk factor and potential ecological risk index of trace metals in stream sediments of mari area

Sample	Cd	Co	Cr	Cu	Hg	Mn	Ni	Pb	Zn
MA1	22.50	0.85	0.27	3.44	13333.33	1.52	0.39	11.09	2.10
MA4	11.25	0.38	0.18	2.30	6920.00	0.91	0.28	13.05	1.50
MA5	18.75	0.37	0.31	2.59	13333.33	1.25	0.30	19.55	1.85
MA6	11.25	0.36	0.33	2.11	13333.33	1.21	0.30	13.73	1.24
MA7	26.25	0.41	0.25	4.69	10186.67	1.64	0.33	9.68	2.13
MA8	3.75	0.34	0.22	2.93	6813.33	1.06	0.36	9.50	1.24
MA9	11.25	0.86	0.31	4.63	6813.33	1.03	0.52	20.18	1.42
MA10	33.75	0.29	0.31	2.06	10840.00	0.85	0.36	14.59	1.21
MA11	7.50	0.20	0.16	1.59	7320.00	0.91	0.18	62.27	1.58
MA12	33.75	0.23	0.25	3.17	13333.33	1.16	0.23	22.91	2.03
MA13	22.50	0.29	0.33	19.81	5813.33	1.12	0.32	38.45	1.75
MA14	26.25	0.20	0.31	2.46	13333.33	0.86	0.19	21.32	1.74
MA18	15.00	0.22	0.34	2.61	13146.67	0.51	0.31	19.64	1.23
MA19	7.50	0.24	0.80	3.11	9933.33	0.48	0.43	45.45	1.07
MA22	11.25	0.29	0.47	8.26	5333.33	0.99	0.22	32.27	1.30
MA23	33.75	0.32	0.19	2.20	11146.67	0.96	0.14	25.45	2.04
MA24	7.50	0.30	0.25	2.37	10226.67	1.30	0.31	10.27	1.30
MA26	176.25	1.25	1.26	41.11	13333.33	0.69	2.96	1340.91	1.18
MA28	78.75	0.25	0.61	2.33	5453.33	0.97	0.25	30.41	1.43
MA29	15.00	0.30	0.25	4.52	13333.33	1.00	0.29	65.45	3.54
PERI	573.75	7.94	7.41	118.30	203280.00	20.40	8.66	1826.18	32.86

Assessment of ecotoxicological risks

Probability of toxicity index (mERMQ) and multiple probable effect concentration quality (mPECQ)

The probability of toxicity index (mERMQ) serves as a tool for identifying heavy metals detrimental effects on the soil environment of heavy metals (Pejman *et al*., 2015) [32]. In order to evaluate the potential consequences of various metals, mERMQ is proposed to determine the potential effects of metal in sediment and it is calculated by the equation:

$$\text{mERMQ} = \frac{\sum_{i=1}^{n} \frac{C_i}{\text{ERMi}}}{n}$$

Ci= concentration of the metal i, ERMi is the ERM value of metal i, and n is the number of metals: Low priority sites are designated as mERMQ < 0.1; mERMQ = 0.1 - 0.5, low to medium priority sites; mERMQ ranges between 0.5 to 1.5 indicates medium to priority high sites and mERMQ > 1.5 refers to high priority sites. mERMQ was used to assess the ecological contamination levels of trace metals in each sample. The results of probability of toxicity index are displays Table 4. The entire study area apart from sample MA26 were all less than 1.5, indicating a high to medium priority sites and sample MA26 was in the class of high priority in the study area. This is a clear indication that the sediments have 100% probability toxicity in the entire study area. This exhibits considerable pollution of metals in sediments of Mari area. It is believed that the occurrence of trace metals in sediments could be traced to mining activities, atmospheric deposition and agricultural practices (Tehna *et al*., 2019; Lemnyuy *et al*., 2022; Ndema Mbongué *et al*., 2023) [37, 18, 26].

The multiple probable effect concentration quality (mPECQ) evaluates the ecological risks levels of trace metals in sediments by the following relation:

$$\text{mPEC}_Q = \frac{\sum_{i=1}^{n} \frac{C_i}{\text{PEC}_i}}{n}$$

Where Ci = content of element i in sediments (mg/kg). PECi stands for probable effect concentration of individual metal based on consensus. The PEC values of Cr, Ni, Cu, As, Cd, Pb, Zn, and Hg were 111, 48.6, 149, 33, 4.8, 129, 459 and 1.06 mg/kg respectively, n is the number of heavy metals. mPECQs < 1, non-toxic (toxicity incidence is only about 25%); mPECQs = 1 – 5 means the incidence of toxicity is 25 – 75% and mPECQs ≥ 5 indicates that toxicity incidence is greater than 75% (Li *et al*., 2019) [21]. mPECQ in sediment ranges from 0.74 to 4.29 (Table 4). Samples MA4, MA8, MA9, MA11 MA13, MA22 and MA28 were all less than 1 indicating no toxic, the incidence of toxicity is relatively low (mPECQ < 1) which is (< 25%) and the rest of the sample shows medium incidence of toxicity (mPECQ < 1-5) which is (25 - 75%) of the research area. The variation in metal concentrations in Mari area required more focus and attention.

Contamination severity index (CSI) and modified hazard quotient (mHQ)

The Contamination severity index (CSI) is informative in terms of the concentration of trace metals in the sediments. It was introduced by Pejman *et al*. (2015) [32] by the relation

$$CSI = \sum_{i=1}^{w} W_t \left[\left(\frac{C_i}{ERL_i} \right)^{\frac{1}{2}} + \left(\frac{C_i}{ERM_i} \right)^{2} \right]$$

W_t = target heavy metal i, Ci is its measured content i, ERLi is its ERL value i, ERMi is the ERM value of target metal i, and n is the number of selected metals. CSI is classified as follows: < 0.5 (uncontaminated), 0.5 - 1 (very low severity contamination), 1 - 1.5 (low severity), 1.5 - 2 (low to moderate severity), 2 - 2.5 (moderate severity), 2.5 - 3 (moderate – high severity), 3 - 4 (high), 4 - 5 (very high severity) and >5 (ultra-high severity contaminated severity). Contamination severity index is a new technique used in evaluation of sediments ecological risk proposed by Pejman *et al*. (2015) [32]. The CSI follow the trend: Pb > Zn > Ni > Cr > Cu > Cd with values 190, 1.79, 1.75, 1.56, 0.91 and 0.55 respectively (Table 5). Cd and Cu exhibits minor severity of contamination (CSI < 0.5 - 1) in sediments, Cr and Ni are in the category of low severity of contamination (CSI: 1 - 1.5). Zn shows low to moderate severity of contamination (CSI: 1.5 - 2) and Pb display ultra-high severity of contamination (CSI > 5) across the study area. Contamination of Pb across the study area is due to intense oxidation and hydrolysis of sulphides mineral (galena), mining activities, automobile exhaust and atmospheric deposition.

Modified hazard quotient (mHQ) is used to evaluate biological impact in sediment-related chemical states on benthic species (MacDonald *et al*., 2000; Emenike *et al*., 2020) [22, 9].

$$MHQ = \left[C_i \left(\frac{1}{TEL_i} + \frac{1}{PEL_i} + \frac{1}{SEL_i} \right) \right]^{\frac{1}{2}}$$

Where Ci stands for the measured concentration of metal i, TELi is the TEL value of the target metal i, PELi is the PEL ratio of the target metal i, and SELi is the SEL value of the target metal i. The following classification is adopted for mHQ values: MHQ > 3.5 corresponds to extreme severity of contamination, 3.0 < MHQ < 3.5, very high severity contamination; 3.0 < MHQ < 2.5, high severity contamination; 2< MHQ < -2.5, considerable severity contamination; 1.5 < MHQ < 2, moderate severity contamination; 1.5 < MHQ < 1, low severity of contamination; 0.5 < MHQ < 1, very low severity of contamination and MHQ > 0.5, nil severity of contamination. The mHQ varies from 0.505 (Cu) in sample MA10 to 9.18 (Pb) in sample MA26 (Table 5; Fig 3). Cd, Cr, Cu and Ni are in the category of low severity of contamination, Cd (in sample MA26), Cr (in sample MA19 and MA26), Cu (in sample MA12 and MA22), Ni (in sample MA26 and Pb and Zn are in the class of low severity of contamination, Cu (in sample MA26) and Pb (in sample MA29) are classified moderate severity of contamination and Pb is in extreme severity class in the study area. Pb with extreme severity of contamination could be attributed to anthropogenic inputs which have a key contribution for the enrichment of metals in stream sediments (Harikrishnan *et al*., 2018) [16].

Table 4: Probability of toxicity index (mERMQ) and multiple probable effect concentration quality (mPECQ) of trace metals in stream sediments of Mari area

Sample	Cd	Cr	Cu	Hg	Ni	Pb	Zn	mERMQ	mPECQ
MA1	0.007	0.12	0.05	7.69	0.09	0.22	0.56	1.25	1.54
MA4	0.006	0.14	0.04	3.99	0.07	0.39	0.49	0.73	0.86
MA5	0.003	0.15	0.03	7.69	0.07	0.27	0.33	1.22	1.52
MA6	0.008	0.12	0.06	7.69	0.08	0.19	0.57	1.25	1.54
MA7	0.001	0.10	0.04	5.88	0.08	0.19	0.33	0.95	1.17
MA8	0.003	0.14	0.06	3.93	0.12	0.40	0.38	0.72	0.85
MA9	0.010	0.14	0.03	3.93	0.09	0.29	0.32	0.69	0.82
MA10	0.002	0.08	0.02	6.25	0.04	1.25	0.42	1.15	1.33
MA11	0.010	0.12	0.04	4.22	0.05	0.46	0.54	0.78	0.91
MA12	0.007	0.15	0.27	7.69	0.08	0.77	0.47	1.35	0.64
MA13	0.008	0.14	0.03	3.35	0.04	0.43	0.46	0.64	0.74
MA14	0.003	0.08	0.03	7.69	0.07	0.26	0.40	1.22	1.51
MA18	0.004	0.16	0.04	7.58	0.07	0.39	0.33	1.23	1.51
MA19	0.002	0.37	0.04	5.73	0.10	0.91	0.29	1.06	1.25
MA22	0.003	0.22	0.11	3.08	0.05	0.65	0.35	0.64	0.73
MA23	0.010	0.09	0.03	6.43	0.03	0.51	0.54	1.09	1.31
MA24	0.002	0.12	0.03	5.90	0.07	0.21	0.35	0.95	1.18
MA26	0.052	0.59	0.57	7.69	0.70	26.82	0.31	5.25	4.29
MA28	0.023	0.28	0.03	3.15	0.06	0.61	0.38	0.65	0.73
MA29	0.004	0.12	0.06	7.69	0.07	1.31	0.94	1.46	1.67

Table 5: Contamination severity index (CSI) and modified hazard quotient (mHQ) of trace metals in stream sediments of Mari area

Sample	Cd	Cr	Cu	Ni	Pb	Zn	CSI
MA1	0.696	0.709	0.732	0.545	0.844	1.110	0.55
MA4	0.684	0.764	0.637	0.485	1.115	1.042	0.62
MA5	0.658	0.781	0.577	0.485	0.937	0.855	0.52
MA6	0.708	0.690	0.850	0.508	0.790	1.117	0.53
MA7	0.633	0.650	0.676	0.524	0.782	0.853	0.46

MA8	0.658	0.764	0.845	0.616	1.133	0.913	0.64
MA9	0.731	0.764	0.570	0.529	0.965	0.843	0.55
MA10	0.646	0.561	0.505	0.397	1.982	0.965	1.12
MA11	0.731	0.690	0.702	0.437	1.206	1.092	0.66
MA12	0.696	0.781	1.735	0.502	1.560	1.014	0.91
MA13	0.708	0.764	0.622	0.404	1.164	1.010	0.62
MA14	0.658	0.585	0.601	0.474	0.914	0.939	0.50
MA18	0.671	0.798	0.640	0.491	1.118	0.853	0.60
MA19	0.646	1.212	0.696	0.570	1.695	0.794	0.97
MA22	0.658	0.937	1.124	0.431	1.430	0.875	0.77
MA23	0.731	0.607	0.589	0.368	1.271	1.095	0.66
MA24	0.646	0.690	0.611	0.497	0.813	0.876	0.48
MA26	1.083	1.516	2.497	1.407	9.182	0.832	183.65
MA28	0.858	1.058	0.606	0.456	1.388	0.917	0.78
MA29	0.671	0.690	0.835	0.480	2.032	1.441	1.32

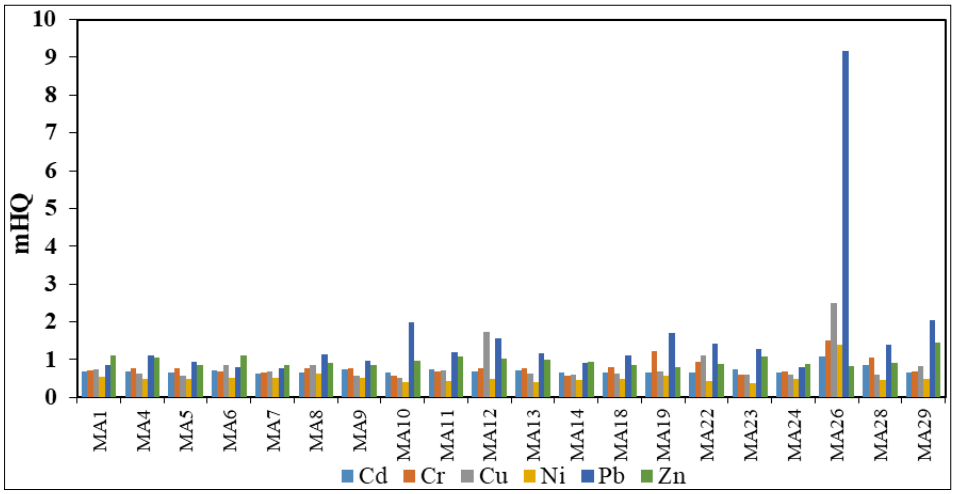

Fig 3: mHQ of trace metals in sediments of Mari Area

Conclusions

The present study investigated the distribution, sources, contamination and metal pollution using integrated pollution and ecotoxicological indicators. The findings have shown that: The stream sediments of Mari area exhibit high concentration in Mn, Pb, Th, Zn, Cu, Hg and U. The. The high content of trace metals could be attributed to intense weathering of the parent rocks, oxidation and hydrolysis of sulphide minerals associated with primary gold mineralization, mining activities, agricultural practices, surface runoff, exhausts emission, waste water irrigation and domestic activities. The other trace metals exhibit low concentrations due to their high mobilization and susceptibility during weathering. The Mari area is strongly contaminated by trace metals. Pollution indices revealed that sediments of Mari area are polluted by Cd, Hg, Th, Cu, Pb and U. The environmental risk factor (Er) and the potential ecological risk index (PERI) values for Cd, Hg and Pb was significant in the study area indicating high potential ecological risk index. The intense weathering of the source rocks, mining operations, ore and metals processing, agricultural practices and industrial emissions may be blame for high contents of Cr, Cu, Hg, Mn, Pb, Th, U, and Zn. Ecotoxicological indicators indicate high severity contamination and a high incidence of toxicity in sediments. The Mari area is facing a high risk of sediments contamination and metal pollution due to the high content of potential trace metals in stream sediments associated with the extensive development of artisanal gold mining and other anthropogenic activities. Attention should be paid to monitoring point sources of metals entering the various environmental compartments from mining activities, atmospheric deposition and agricultural practices in the area.

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