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Spatial Distribution of Heavy Metals in Different Soils Particle Size Fractions at the Deposit Site and in the Vicinity of Mkuju Uranium Mine in Tanzania

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Abstract

This study has been conducted with the aim of benchmarking the levels of selected heavy metals in different particle fractions and their spatial distribution in the soil of Mkuju River deposit area and adjacent human settlements before the commencement of uranium extraction industry in the area. The soil samples were randomly collected from these areas and measurements were undertaken for bulk and particle fractions concentrations of Ba, Cr, Cu, Pb, Sr, Zn, Hg and Cd using Energy Dispersive X-ray Fluorescence (EDXRF). The total concentrations of Ba, Cr, Cu, Pb, Sr, Zn, Cd, and Hg in mg/kg; in the bulk soil samples for both sites ranged from; 153.54 - 2140.89, 23.37 - 138.74, 2.47-161.73, 6.84 - 45.2, 25.22 - 420.25, 17.78 - 356.88, 0.18 - 47.62, and 0.06-0.51 correspondingly. The dominant soil fraction size for both areas was 300-150 µm, while the finest fraction represented 13% and 12% for Mkuju River and human settlements respectively. The relatively high heavy metals concentrations were observed in fine soil fractions than coarse soil fraction, which signify potential high risk from air pollution through atmospheric dispersion, considering that fine particle are easily dispersed, and remain suspended for a long period of time as compared to coarse particles. Cr had high distribution factor of 5.33 and 3.28 for Mkuju River and human settlements respectively. The observed baseline results prior to commencement of mining activities in the study areas reveal that, most of heavy metal concentrations in the areas are within the permissible limits provided by USEPA, mean world values, Tanzania Bureau of Standards (TBS), and that the observed heavy metals distribution characteristic is a results of natural processes.

Keywords

Heavy Metals, Mkuju River, Uranium, Air Pollution

1. Introduction

Tanzania is currently in a process of starting uranium mining activities at the Mkuju River in the Southern part of the country. This shall be the first uranium mining in Tanzania and the East Africa Region. It is thus important to define the baseline levels of heavy metals in the environment before the commencement of mining activities. It is anticipated that the uranium mining and milling activities at Mkuju River will increase the background levels of various heavy metals in the vicinity the mining and processing sites. According to Navarro [21] the anthropogenic activities such as mining activities can lead to an increase in the concentrations of heavy metals in the environment that could lead to severe environmental problems such as impacting human health as well as damaging the biodiversity and ecosystem. Different studies in the world have reported the

presence of elevated concentrations in the environment of different heavy metals resulting from uranium mining, milling and wastes disposals from mining and processing activities. The elevated heavy metal levels, contaminate the soil with subsequent negative impacts on soil quality as well as human and biological systems [15 and 10]. Normally, when disposed wastes are not well covered; wind and water erode their surface and materials with heavy metals, especially fine particles, are transported long distance. Fine particles have high specific area, that retains high amount of heavy metals [30]. These kind of particles are easily transported and dispersed in the air and water media [28]. Thus, understanding of the distribution characteristics of heavy metals in the soil before mining activities commences is essential and is central for hazard appraisal and soil remediation practices [18].

The accumulation of the high level of heavy metals in the soil can affect the plant growth on the soil, thus restricting the use of soil for different purposes [25], [9], [4]. Contamination of soil by heavy metals appear to be virtually permanent, as heavy metals can be transformed from one compound to another through chemical and biochemical reactions, but cannot be destroyed. On other hand, the human uncontrolled intake or chronic exposure to heavy metals can lead to several human and animals risks. For example, the excess amount of Lead and Cadmium in the human body causes lung cancer, prostatic proliferative lesions, kidney dysfunction, bone fractures, damage of nervous system, and suppress of immune systems [32]. The exposure effects are more dangerous for heavy metals with the radionuclide characteristics such as Uranium, Strontium and Thorium. The decay of Strontium releases sulphides and oxide particles to environment and on inhalation by human beings, the decay products can cause bone cancer [8]. Likewise, the transfer of Thorium even at low concentration from the soil to plant leaves, has been reported to increase cancer risk to the human being [7].

Some researches in Tanzania have been conducted to determine the levels of heavy metals in the soil [20], [13], [3], [5]. However, most of the previous studies focused on assessing the heavy metals in bulk soils and none of them assessed the spatial distribution and its particle size fractions especially from uranium mining. Normally, the size fractions have different composition and properties, which can affect the behaviour of pollutants in the soil and air microenvironment [1].

This study was carried out to investigate the spatial distribution of heavy metals in the bulk soil and size fractions of the soils occurring naturally on earth surface. The study will provide the baseline data and background information prior to uranium mining in the cited area. The heavy metals analyzed in this study were Strontium (Sr), Barium (Ba), Mercury, Cadmium (Cd), Chromium (Cr), Lead (Pb), Copper (Cu) and Zinc (Zn). These heavy metals are associated with uranium mining and milling activities.

2. Materials and Methods

2.1. Description of Study Area

The study areas is located in the Southern part of Tanzania between latitudes 10° 03' 00" S and longitudes 36° 53' 00" 'E within the Namtumbo district, in Ruvuma Region (Figure 1). The study area encompasses Mkuju River uranium deposit and its three surrounding human settlements villages namely Likuyu, Mandela and Nambecha. These settlements are located at an average of 54 km from the Mkuju River Deposit. The settlements are located at the Likuyuseka ward with estimate of total population of 10,811 [29]. The deposits site is not currently occupied by people except for a camp occupied by staff.

Mkuju river hosts a viable uranium deposits of sandstone type of about 25, 200 t U, with an estimated production of 1,600 tU per year at its maximum capacity, over a minimum of 12 years [16]. The uranium mineral at Mkuju River occurs at the shallow depths, which justify the use of the open pit mining method. The method will generate large amount of wastes in form of dusts, which is easily transported by wind and water erosion to the settlements around the deposits. The study areas have tropical climate with average monthly rainfall of about 90.5 mm and heavy rainfall extending from January to April. The maximum wind speed in the area is 11.6 ms^{-1} which dominantly blows from Southwest (SW) to Northeast (NE). The ambient temperature varies from 18.8°C to 28.2°C with an average of 22.3°C.

2.2. Sample Collection and Preparation

The soils investigated in this study were sampled from Mkuju River uranium deposit site and the adjoin settlements villages of Likuyu, Mandela and Nambecha. The sampling points were randomly selected and a total of 28 soil samples were collected. Sixteen (16) sampling points were at Mkuju River and 12 sampling points were located at human settlements near the deposit (Figure 2). The sampling points were georeferenced using hand held Geographical Positioning System (GPS). The samples were collected from the surface layer based on the fact that, this layer is more affected by re-suspension [2] and at each sampling point, three samples of approximately 500g each were collected and later mixed in the laboratory to obtain homogenous soil samples. Each soil sample was packed in polythene bags, well labeled to indicate the site name, sample number and sampling date. The soil samples were transported to Ministry of Water, soil laboratory, in Dar es Salaam where the dry sieve analysis was performed. The fractionation of bulk soils into five particle size fractions was conducted using sieves with the following sizes: 600, 425, 300, 150 and 75 μm. The sieved soil samples were well packed in the polythene bags and transported to the Tanzania Atomic Energy Commission (TAEC) laboratory in Arusha City for analysis. Heavy metals concentrations in the soil were measured from bulk soil samples, without dry sieving for the five soil particles fractions.

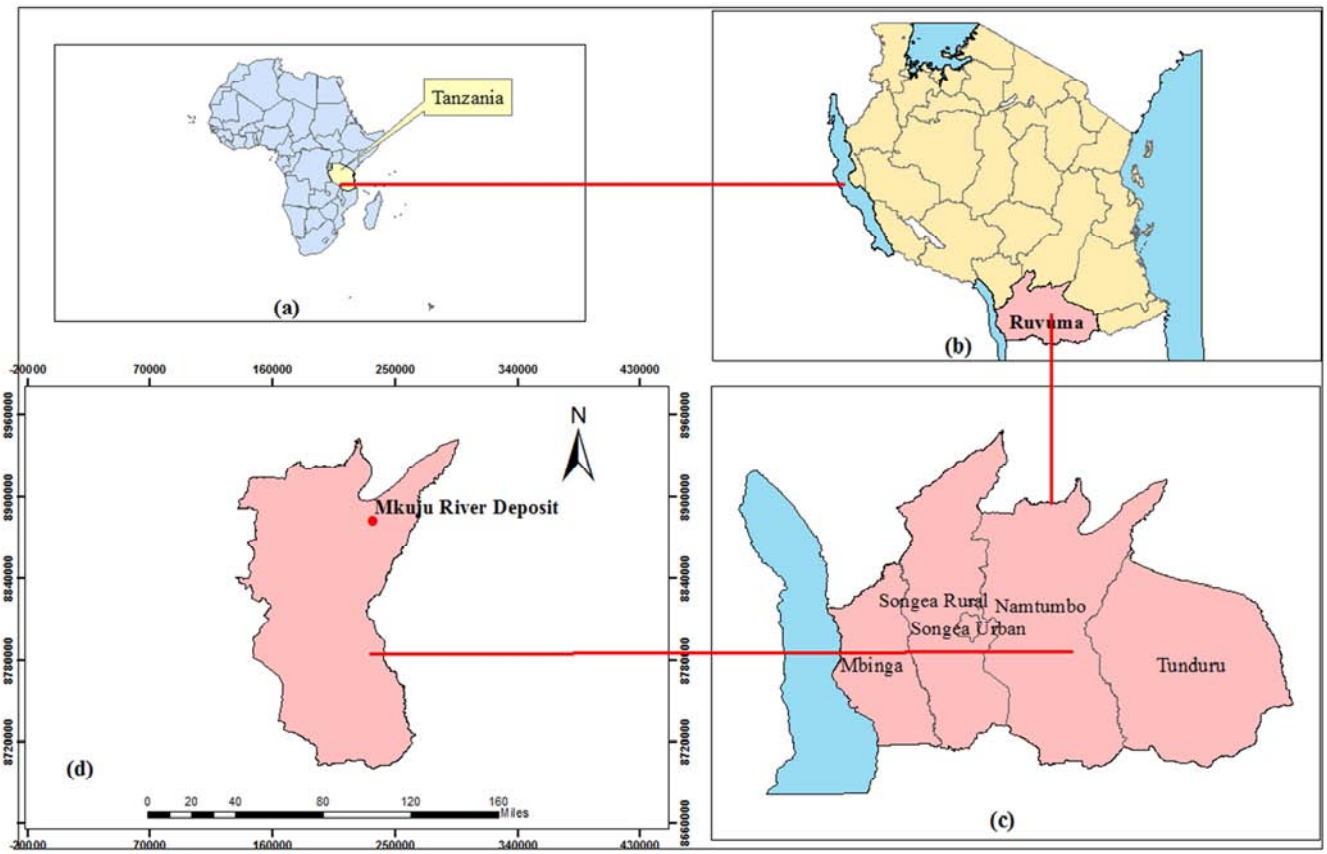

Figure 1. *Location of the Ruvuma Region, Namtumbo District and Mkuju River Uranium Deposit.*

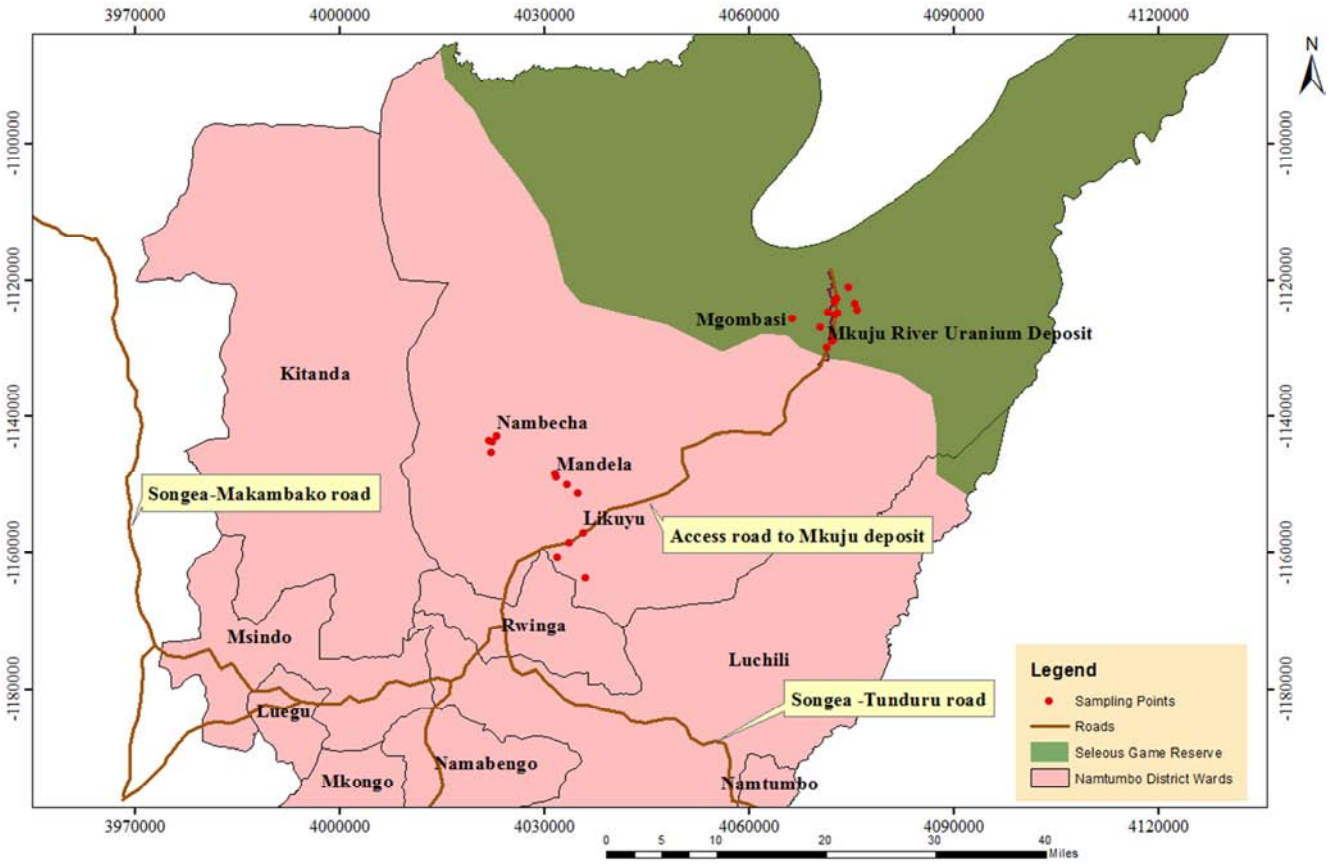

Figure 2. *Soil Sampling Points.*

2.3. Analytical Methods

The soil samples were individually dried to reduce moisture contents and attain the constant weight in an oven at a temperature of 100°C for 24 hours. To reduce the particle size and attain the conditions for homogeneity of the sample, each sample was grounded into fine powder using an agate mortar and pestle and then sieved through 2 mm stainless steel sieve. Prior to analysis with EDXRF, 4 g of each sieved sample was mixed with 0.9 g cereox binder materials manufactured by Fluxana company in Germany and then homogenized in a pulverizer to attain the homogenous mixture of sample. The homogenized mixture of sample was then pressed at about 15 tons to obtain cylindrical pellets of 32 mm diameter using a die pellet maker.

The heavy metal composition of each pellets were determined by using the polarized Energy Dispersive X-Ray Fluorescence (EDXRF) Spectro Xepos model with serial number 4R0138. The EDXRF equipment has an inbuilt Turboquant (Tq 9232) algorithm for matrix effect correction which increase the excitation sensitivity of elements (Schramm and Heckel, 1998). The instrument use three secondary targets to increase the excitation sensitivity of elements, the light energy elements from Sodium (Na) to Vanadium (V) use High Oriented Pure Graphite (HOPG), the middle energy elements from Chromium (Cr) to Zirconium (Zr) and Praseodymium (Pr) to Uranium (U) were excited using Molybdenum secondary target, a Barkla Aluminum Oxide (Al_2O_3) target is used to excite the elements with high energy from Yttrium (Y) to Cerium (Ce) [24].

Measurement was done by loading each prepared pellet into a cleaned sample holder and then inserting the same in the X-ray excitation chamber. The EDXRF measurements mechanism involved the irradiation of the sample with X-rays and measurements of the energy (Wavelength) and intensity of the generated fluorescent X-rays to determine the type and content of the elements in the sample. In practice EDXRF used in this study is calibrated by the manufacturer, however the validity of manufacturer calibrations is achieved by performing the routine multichannel analyzer recalibration once in a week. The analysis time was constant and equal to 16 minutes per reading for each sample. The heavy metal data presented in this paper is the mean of the three measurements taken with ED-XRF instrument.

In order to maintain the quality of the output data, the accuracy provided by EDXRF and the significant contamination or interference in the analysis process was conducted. The accuracy provided by the EDXRF technique was evaluated by using the Montana soil 2711A Standard Reference Material (SRM) obtained from National Institute of Standards and Technology, USA. The standard soil was prepared and analysed by using similar experimental conditions as the unknown sample. The average measured concentration of SRM were compared with certified values of the same element in a sample, which facilitate the establishment of the level of agreement between the measured and certified values.

The significant contamination or interference in the analysis process was controlled by analyzing the binder material and assess if there are any contamination of the sample by binder which may lead to reporting the high level of metal concentrations from the field soil samples. The binder material preparation employed the same procedure as soil sample and Soil Reference Material. The results of the measured binder concentrations revealed that there was insignificant contamination or interference for each analyte.

2.4. Heavy Metal Distribution Factor

The distribution factor is the term used to indicate in which size fraction the heavy metal is preferentially enriched. The distribution factor has been widely used in studies of metal distribution in different soil particle size fraction [2], [1]. The distribution factor is normally calculated as a ratio between the metal concentration in particular particle size fraction and the metal concentration in the bulk soil. The equation given by [1] has been used in this study to determine distribution factor of each analyzed metal in each of the particle fractions. The distribution factor was calculated by using Eq. (1) below.

$$DF_x = \frac{X_{fraction}}{X_{bulk}} \quad (1)$$

Where, X fraction and X bulk are composition in mg kg^{-1} of heavy metal in a given fraction and bulk sample respectively. The heavy metal is assumed to be distributed in this fraction, if distribution factor is greater than 1.

3. Results and Discussion

3.1. Physical Characteristics

The physical characteristics of Mkuju River and adjacent human settlements soils were measured and presented in Table 1, while Figure 3 shows the mass fraction in percentage of the soil particles in the surface soil from Mkuju River and the Human Settlements area. It has been observed that the Mkuju River and the human settlements area have similar soil characteristic where sand soil is dominant over silt and clay soil. The mean sand content (wt/wt) were 72.64% and 70.00% for Mkuju River and human settlements area respectively. Similarity the observed mean specific density of soil for Mkuju River and human settlements area were 2.36 g/m^3 and 2.46 g/m^3 respectively. The soil in both area is not reach in organics and friable clay. The p-value was 0.626 which indicated that the difference between specific density values is not significant at $p<0.05$.

Fractionation of bulk samples into various particle sizes revealed different characteristic between the two study areas. In human settlements area, the dominant particle size fraction was 300-150 µm with a mean composition of 28% (wt/wt) (Figure 3); the smallest size fraction (<75µm) ranged from 7% to 19% (mean 12%) while the largest size fraction (600-425

µm) ranged from 5% to 29.4% (mean 12%). On other hand, Mkuju River samples showed a heterogeneous particle size distribution with dominant fraction at 300-150 µm (mean 37%); while smallest fraction (<75µm) represents 13% of the total soil and the largest fraction (600-425 µm) varied from 4% to 29%, with mean value of 17%.

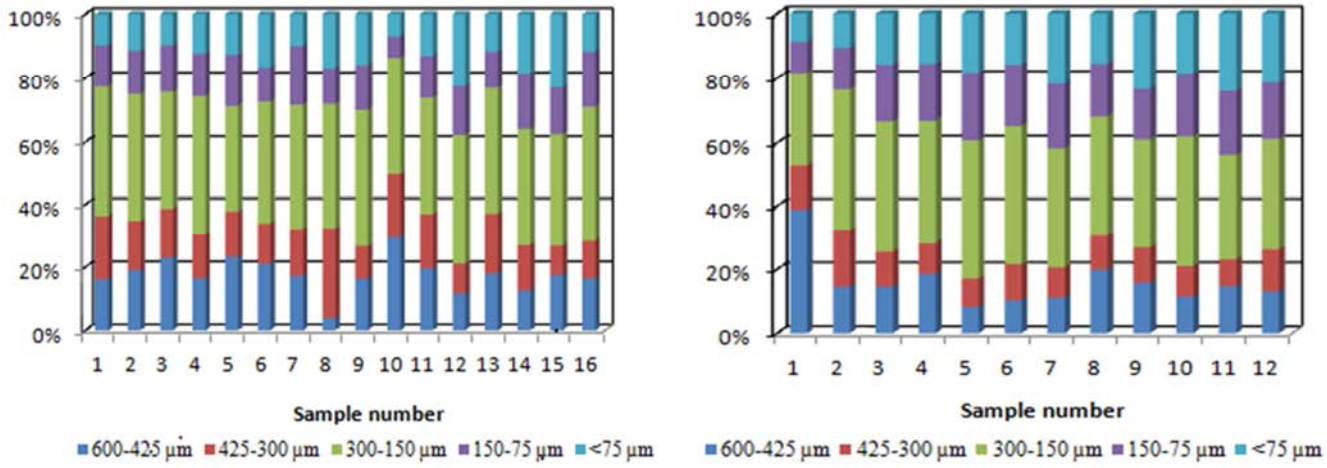

Figure 3. *Composition in percentage of five particle size fractions in surface soil from Mkuju River (left) and Human Settlements area (right).*

Table 1. *Mean Physico-chemical characteristics of Mkuju River and human settlements soils.*

Sampling Area		Specific Density (g/m^3)	Gravel (%)	Sand (%)	Silt (%)	Clay (%)
Mkuju River	Mean	2.36	4.4	72.64	13.33	9.57
	Min	0	0	31.15	3.67	1.73
	Max	2.97	29.13	90.15	61.77	22
Human Settlements	Mean	2.46	1.77	70	14.76	13.46
	Min	2.01	0	29.2	2.89	1.7
	Max	2.65	17.4	91.87	36.7	34

3.2. Heavy Metals Distribution in the Bulk Soil

The basic statistics of the measured heavy metals concentrations, including minimum and maximum values, average, median and standard deviation (SD), and mean world values are presented in Table 2. The spatial distribution of heavy metals concentrations in the bulk soil samples were measured and compared with the earth metal concentrations in the earth's crust, Tanzania Bureau of Standards (TBS) and EPA permissible levels [27], [26].

Table 2. *Mean (mg/kg), Standard Deviation (STD), Minimum and Maximum concentrations of Elements at Mkuju River and Human Settlements Areas compared to Permissible Levels from TBS and EPA.*

Sampling Areas		Cr	Cd	Cu	Zn	Sr	Ba	Pb	Hg
Mkuju River	Mean	56.37	3.00	31.55	46.29	214.08	1375.17	31.37	0.24
	STDEV	3.5	2.2	0.7	0.6	0.6	19.1	0.4	0.1
	Min	23.37	3.00	31.55	26.34	74.82	364.72	6.84	0.12
	Max	108.39	23.55	161.73	130.29	420.25	2140.89	40.32	0.51
	Median	53.02	1.87	16.37	39.48	206.32	1356.87	32.92	0.22
Human Settlements	Mean	74.25	5.52	23.32	108.92	85.47	672.97	27.20	0.19
	STDEV	3.10	1.99	0.92	0.83	0.62	12.03	0.45	0.50
	Min	34.88	0.18	2.47	17.78	25.22	153.54	11.78	0.06
	Max	138.73	47.62	47.13	356.88	153.02	1496.38	45.21	0.31
	Median	61.77	1.60	22.33	41.29	81.70	534.00	26.81	0.20
	Mean World Conc.	100	0.06	30	50		340	10	0.03
	Max TBS		1	200	150			200	
	Max USEPA	11	0.43	270	1100			200	

The comparisons of the spatial distribution of the bulk soil samples, showed that; Cu, Ba, Sr, Pb and Hg have higher mean concentrations at Mkuju River deposit than the human settlements as presented in Table 2. The possible reason for

higher mean concentrations of these metals at Mkuju River compared to human settlements area could be geogenic characteristics of the area. The magnitude of heavy metals concentration at Mkuju River and Human settlements area followed the following sequence: Ba> Sr> Cu> Zn> Cr>Pb>Cd>Hg and Ba>Cr>Sr>Zn>Pb>Cu>Cd>Hg respectively. Barium had highest concentration while mercury had minimum concentration in both study sites. The Ba mean concentrations value were 1375.17 mg/kg and 672.97 mg/kg for Mkuju river and Human Settlements respectively. The mean Ba concentrations for both study sites exceed the background level of 550 mg/kg in the earth [14]. Barium in nature is widely distributed in the igneous rocks, sandstone and shale, which is typically the geological characteristic of Mkuju River and surrounding human settlements area [14], [16]. This might be the reasons for higher values of Barium compared to other elements for both sites. The measured levels of Zn, Cu and Pb for both sites were below the TBS maximum permitted heavy metals in the soil. The mean Mercury values for both areas were above the mean world values of 0.03 mg/kg [14], [19]. The mean Strontium concentration levels for Mkuju River and human settlements were within the desirable level of 350 mg/Kg in the earth crust. This might be the reasons of higher values of Barium than other elements at both sites. However, the measured levels of Zn, Cu and Pb for both sites were below the desirable limit as per TBS maximum permitted heavy metals in the soil. Mercury mean values for both sites were above the mean world values of 0.03 mg/kg [14], [19]. The mean Strontium concentration levels for Mkuju River and human settlements were within the desirable level of 350 mg/Kg in the earth crust.

The levels of Cr, Cd and Zn were higher in the human settlements area compared to their levels at Mkuju River deposit. The leading heavy metal at the human settlements area is Zn with a mean value of 108.92 mg/kg, followed by Cr with the mean 74.25 mg/Kg, and lastly Cd with mean value of 5.52 mg/Kg. High concentration of Zn at the human settlements area might have been contributed by farming system of the area. The soil type in the area is sand which characterized with low plant available Zn, which encourage the use of the artificial fertilizers containing Zn Sulphate for agricultural activities. This lead to increased Zn levels in the area. Although Zn has a leading concentration in the human settlements area with mean value of 108.92 mg/kg, its level is still below the TBS allowable limit of 150 mg/Kg (Table 2).

The Cadmium mean concentrations in the soil were 3.00 mg/Kg and 5.52 mg/Kg for Mkuju River and human settlements area respectively. These values were higher than the Tanzania Bureau of Standards maximum allowable concentration of 1 mg/kg [26]. For Chromium, the mean values in the soil were 56.37 mg/kg and 74.25 mg/kg for Mkuju River and human settlements area respectively which were higher than the maximum allowable level of 11 mg/kg specified by USEPA. The observed higher values of the Cd and Cr at the human settlements area than Mkuju River vicinity could be due to the application of the phosphate fertilizers in farms within the vicinity of the human settlement areas. It was observed that, agricultural activities in the human settlement area utilize a significant amount of fertilizer including phosphate fertilizer to facilitate plant growth and increase the productivity. The phosphate fertilizers contains trace amount of the heavy metals including Cd and Cr, as impurities, which, after continued fertilizer application may significantly increase their content in the soil [12]. In general, most of the measured heavy metals concentrations were below the EU and TBS required standard, which means that, any future increase of concentration of these heavy metals, may be due to the mining activities.

3.3. Heavy Metal Distribution in the Soil

Heavy metals distribution in soil of the Mkuju River and human settlements was measured and presented in Figure 4. The heavy metal measured concentrations were compared with Tanzania Bureau of Standards (TBS), United State Environmental Protection Agency (EPA) and mean world average.

The comparison of heavy metal contents in different size fractions revealed highest concentrations in the fractions of less than 75μm in both study areas which can be described to the larger surface area of soil particles which in-effect render higher metal adsorbance [17]. The Chromium (Cr), Cadmium (Cd) and Mercury (Hg) concentrations in the size fraction less than 0.75 μm exceed the limit as outlined by EPA, TBS and mean world for both study areas. The reasons contribute to higher concentrations which exceed the limit include the geological natural occurrence of the area and also the application of fertilizer in the farming system in the vicinity settlements. The lowest concentrations of measured heavy metals were observed in 425-600mg/kg. In an essence, the heavy metal contents in the soils showed quite high values in the Mkuju river followed by human settlements, the condition can be attributed to the geological formation of an area. Zinc (Zn), Copper (Cu) and Lead (Pb) concentrations at finest fractions were below the EPA and TBS. Although there are is no international standard of soil particle partition, this research results have been similar to other research reported elsewhere [1], [6], [31], [23], [33]. The highest concentration in the fractions of less than 75 μm calls for scientific intervention to ensure safety. This will be even serious when major mining activities, commerce where heavy metals levels is expected to increase.

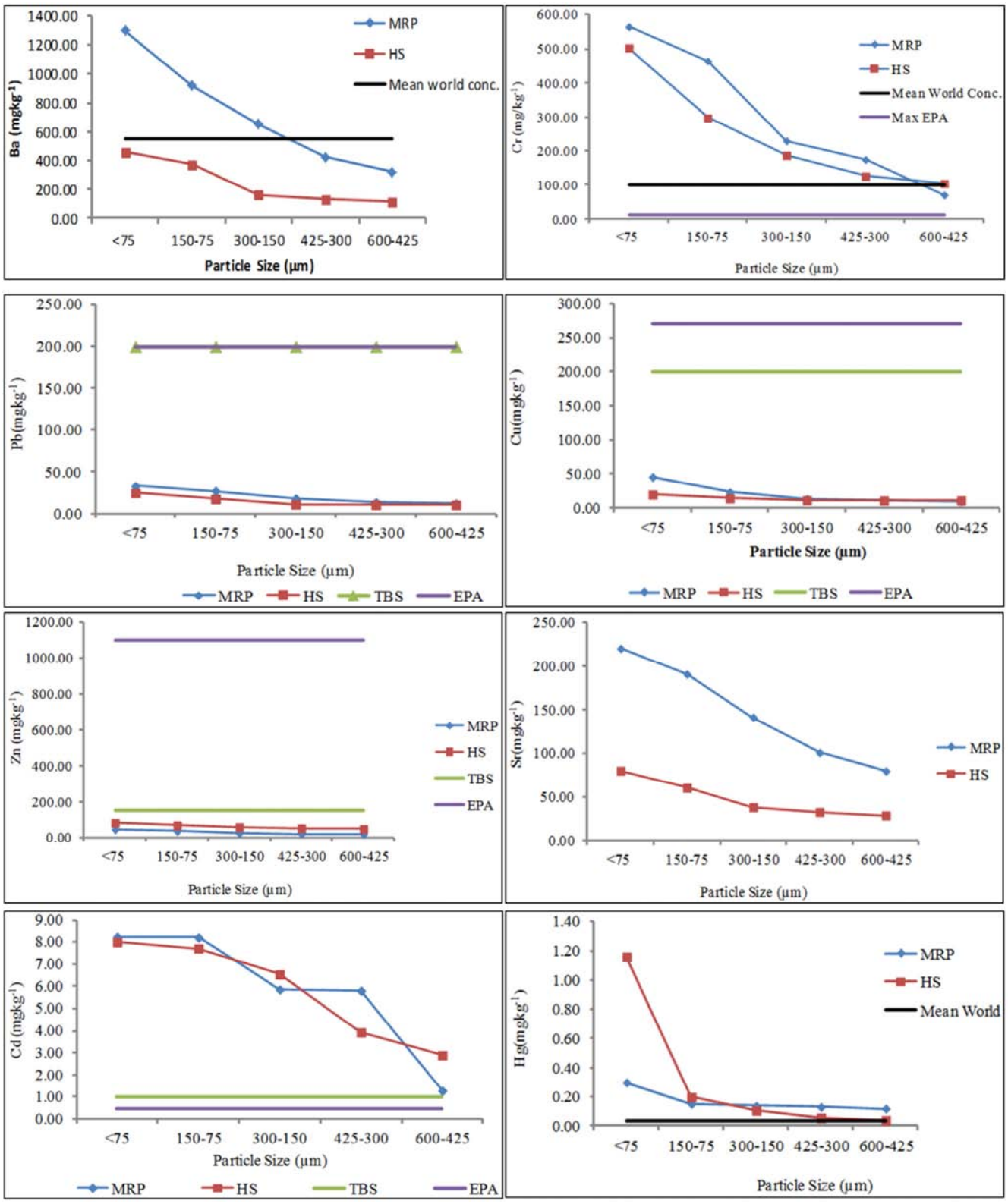

Figure 4. *Mean concentrations (mg/kg) of heavy metals in the five particle size fractions of the soils from Mkuju River Project (MRP) and Human Settlements (HS).*

3.4. Distribution Factor Analysis (DFx)

The distribution factors (DFx) of analyzed metals for Mkuju River and Human Settlements are shown in Figure 4. The DF_X show that the heavy metals distributed more in the finer fractions than in the coarser fractions, mean that the distribution of heavy metals in soils increased with decrease of particle size of the soil. The smallest faction (<75µm) for Mkuju River had the highest mean DF_x of 10.02, 2.75, 1.43,

1.25, 1.04, 1.03, 0.94 and 0.93 for Cr, Cd, Cu, Hg, Pb, Sr, Ba and Zn respectively and human settlements distribution factors was 6.78, 6.22, 1.45, 0.93, 0.90, 0.82, 0.72 and 0.62 for Cr, Hg, Cd, Sr, Pb, Cu, Zn and Ba respectively. The lowest distribution factor was on observed in coarse particle fraction (600-425 µm) for both sites. Cr was a leading heavy metal distributed to both study areas with overall distribution mean value of 5.33 and 3.28 for Mkuju river and human settlements respectively. The presence of Cr is due to natural occurrence and anthropogenic activities.

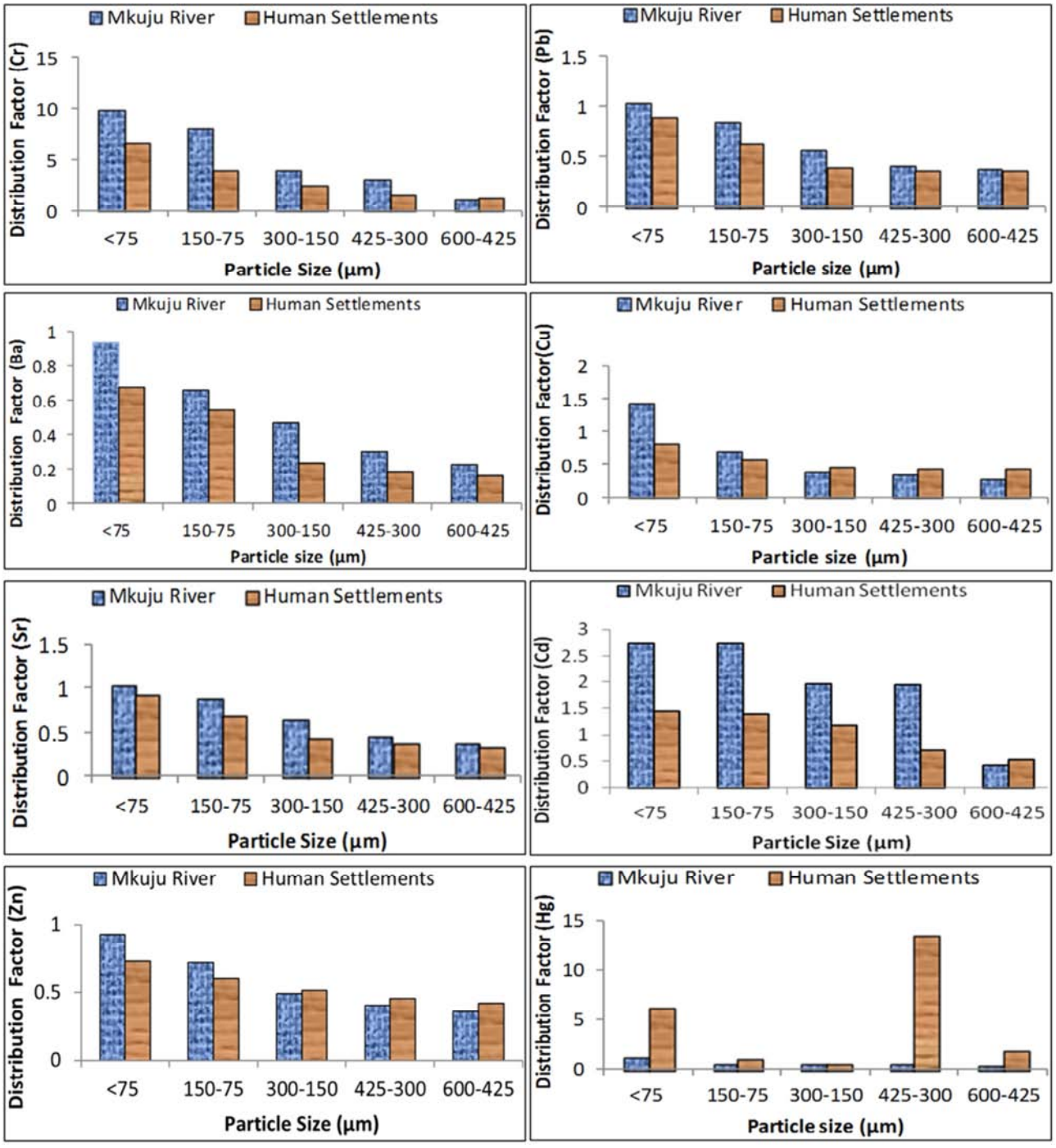

Figure 5. *The AFs for Cr, Pb, Ba, Cu, Sr, Cd, Zn and Hg in each soil particle size fraction.*

4. Conclusion

The pre-mining characterization of soils and heavy metals status data found here-in can be used as baseline values for monitoring of environmental contamination in the future, and during land reclamations after post-mining activities of the proposed uranium mining site. Results of this study will be used as benchmark during the implementation of environmental management plan and thus reduce possible negative impacts on ecosystem health.

The soil at the study areas is characterised with sand soil

type with dominant particle size fraction of 300-150 µm and the mean value of 37% and 28% for Mkuju River and human settlements respectively. Most of the measured heavy metals at Mkuju River and human settlements have concentrations below the TBS and EU allowable levels in the soil. The bulk soil samples indicated that Mkuju River have more heavy metals than the surrounding human settlements. However, the measured soil samples showed that Barium is a leading heavy metals with high mean values of about 1375.17 mg/kg and 672.97 mg/kg for Mkuju River and human settlements respectively. The levels are supported by geological nature of the area. Cadmium and Chromium showed higher concentrations than TBS and EPA standards respectively. This call for agent and proper monitoring of these metals even before the commencement of mining activities, since the mining activities is likely to increase the heavy metal concentrations in the environment, hence more exposure and effects to public.

The effect of particle size fraction on heavy metal distribution in the soil showed that higher concentrations of heavy metals occurred in finest fractions than other fractions, this is due to high surface area and negative charges associated with fine particles. Moreover, the Distribution Factor (DFx) for both study areas indicated that the heavy metals were distributed more in the finer fractions than in the coarser fractions. Cr was a leading heavy metal distributed to both study areas with overall fraction distribution mean values of 5.33 and 3.28 for Mkuju river and human settlements respectively. The presence of Cr is due to natural occurrence and anthropogenic activities. Fine particles, once blown to the air, remain suspended for a very long time, and as such are not deposited easily. Heavy metals in the area are thus likely to affect the environmental quality in general and the vulnerable population which include children and the elderly. In general, most heavy metals occurs naturally in the area, and the anticipated mining activities will contribute to the increase of environmental pollution in the area and beyond as the fine particles get transported by air as the transportation media.

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