Uranium contamination in drinking water and foodstuffs in Bahi District, Central Tanzania

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Abstract

A field survey was conducted in Bahi District in Central Tanzania to investigate uranium levels in drinking water and to evaluate its contamination in some foodstuffs as proxy to their safety for human and animal consumption. Regularly consumed cereal grains, cassava leaves, salt, soda ash, catfish, flamingo meat, surface and underground waters were randomly sampled in the district and analyzed using inductively coupled plasma - optical emission spectrometry (ICP-OES). The study found extremely high levels of uranium in surface and underground waters of up to 1233µg L^{-1}, a value that exceeds the World Health Organization standard of 30 µg L^{-1} by a factor of 41. Soda ash, which is locally consumed and some is exported, had a very high value of 1910 µgU kg^{-1}. Finger millet grains, catfish and flamingo had 32, 17.98 and 31.78 µgU kg^{-1}, respectively, values that were higher than the natural background level of 14 µgU kg^{-1} found in some common foodstuffs by the European Food Safety Authority. It was concluded that drinking water and consuming foodstuffs with high levels of uranium is endangering human and animal life in Bahi District. This may cause leukemia, brain disorder, kidney failure, lung damage and/or bone cancer. The public should therefore be informed about this risk and relevant authorities should undertake regular screening of food products from the affected district as a mitigation measure to avoid health problems in future.

Keywords: Foodstuffs; Soda ash; Tanzania; Uranium; Water

Introduction

Commercial superficial uranium (U) deposits have recently been confirmed in Bahi and Manyoni districts in Central Tanzania. The deposits occur in several seasonal wetlands with closed drainage system, which are surrounded by isolated hills of granites. The wetlands are fully utilized for growing rice, finger millet, sorghum, vegetables and other crops. In addition, these wetlands have for many years been used for grazing domestic animals, production of salt and soda ash, and fishing, without knowledge of the presence of superficial U deposits. Equally of concern is the fact that surface and ground waters, some of which might contain dissolved U compounds, are used for drinking, cooking and irrigation.

Granites in Bahi District are believed to be the primary source of this Uranium. In agricultural soils and ground water, U is enriched by weathering of parent rocks, particularly granites (Sasmaz and Yaman, 2008). Weathering releases the element from granites and makes it available in soils for plant uptake. A high level of U in agricultural soils enhances the chance of its assimilation and translocation in various parts of plants (Anke *et al*., 2009). According to Shahandeh and Hossner (2002), dicotyledonous plant species tend to accumulate more U than monocotyledonous species. Its bioavailability in soils for plants uptake depends on the interactions between physico-chemical and biological processes (Laroche *et al*., 2005).

Solubility and mobility of U in soils depends on the pH, redox potential, soil temperature, soil texture, organic and inorganic compounds, soil moisture and microbial activity (Rivas, 2005). For instance, in soils at pH < 6, U is available for plant uptake and translocation to various plant tissues as uranyl (UO_2^{2+}) cation (Ebbs *et al*., 1998). The soil pH determines U complexes that are formed in soils. For instance, where there are carbonate compounds in soils, soluble uranyl carbonate complexes are formed at a pH of 5.8 – 7.0, which enhances its ability to move in soils in the presence of soil moisture. In the presence of organic compounds, U may form soluble complexes which promote its mobility in soils. In contrast, soils with a lot of clay minerals, which are negatively charged may form complexes with U and thereafter inhibits its movement compared to sandy soils. Also soils under reduced conditions (redox potential [Eh] of less than 0.0V), reduces the soluble U (VI) species to the less-soluble U (IV), and thus, inhibit its movement. The solubility and mobility of U increase its ability to spread in agricultural soils and chance to be taken by many crops and grasses/shrubs used for grazing animals and/ or human consumption. However, U in soil does not often create a radiological hazard to humans, but it can cause biochemical toxicity, which could lead to leukemia, brain disorder, kidney failure, lung damage and /or bone cancer (Schnug *et al.,* 2005; Busby and Schnug, 2007; Bensoussan *et al*., 2009). The dangerous effect of U biochemical toxicity is about six times higher than those from its radioactivity (Schnug *et al.,* 2005).

Drinking surface and ground waters is among the possible pathways by which people and animals are exposed to U (Bensoussan *et al*., 2009). According to Duff and Amrhein (1996), U could be transported by water in a soluble form as uranyl ion (UO_2^{2+}), which is a complex formation with carbonates and phosphates. Closed drainage systems that feed wetlands in areas like Bahi and Manyoni in Central Tanzania might have significant accumulation of soluble U (Owen and Otto, 1995). Through the food chain, U can be transferred from the ground to humans and other living species. Knowledge about the extent of uptake and concentration of U within such a chain is lacking in most developing countries including Tanzania and, thus the need to establish prevailing levels for the safety of the people and animals.

Since U is taken up and accumulated in plant tissues and it can dissolve in water (Anke *et al*., 2009; Duff and Amrhein, 1996), there is likelihood that it is transferred from the ground to humans and other living species through the food chain in the study area. Thus, this study aimed to establish the levels of U contamination in drinking water and its uptake in food chain as a proxy to their safety for human and animal consumption.

Materials and Methods

Study area

The study was carried out in Bahi District at Bahi, Mpamantwa and Ilindi Wards. The district is situated in the central plateau of Tanzania between Latitude 4º and 8º South and Longitude 34º and 38º East. Topographically, Bahi District is characterized by a number of isolated hills of granites (granitic inselbergs) and lowlands which are flooded during the rainy seasons. It has a semi-arid tropical climate with unimodal and erratic rainfall of about 500 to 700 mm that falls between late November and mid-April (Swai *et al*., 2012). The district has an estimated population of 221,645 people, mostly farmers who are also livestock keepers (National Bureau of Statistics, 2012). Major food and cash crops cultivated in Bahi District include finger millet, sorghum, maize, paddy, cassava, potatoes, groundnuts, sunflower, simsim, grapes and pigeon peas (URT, 2003). Cattle and goats are the most dominant livestock found in the district. Other economic activities include fishing of *African Clarias* (*Clarias gariepinus*) and *Nile Tilapia* (*Oreochromis niloticus*) in swamps and rivers during the rainy season and production of salt and soda ash during dry seasons for local consumption and selling outside the country.

Sample collection

Fully matured maize, finger millet and rice grains as well as cassava leaves were randomly collected in farmers' fields (plots) from three wards, namely, Ilindi, Mpamantwa and Bahi in Bahi District. In Ilindi and Mpamantwa Wards, 18 farms were randomly selected for sampling maize grains, finger millet grains and cassava leaves. Each food crop was sampled from 6 farms in triplicate for laboratory analysis. Samples in a plot were taken at an interval of about 50 meters (50m) from one another depending on the plot size. Sampling of rice grains was carried out in Bahi Ward, which is famous for growing and selling rice for local consumption and for export. A total of 15 samples of rice grains were collected from five randomly selected paddy fields.

Two types of water samples were collected, one from streams and another from underground well water. The water sources were randomly selected from each of the three wards. Surface water from streams was taken at three points; upstream, middle and downstream at approximately 100m apart. Triplicate water samples were then taken from each selected site giving a total of 27 water samples. In swamps or Playa Lake, water samples were taken at the edges and at the centre using a locally-made boat. For well water, samples were collected after running the tap from the well for 2–3 minutes in order to avoid contamination from previous users (Orloff *et al*., 2004). All water samples were collected in clean plastic bottles, which were washed thoroughly with ionized water and dried before going to the field for sample collection. The bottles were sealed after sampling and stored in a cool box before transferring them to the laboratory where they were put in a refrigerator at -10 ^{o}C.

Salt and soda ash samples were only collected from Ilindi Ward, where they are produced in larger quantities. Salt samples were collected from two sources in triplicate, making a total of six samples. At each source, salt samples were collected from the production site of three individual small-scale local producers. For soda ash, triplicate samples were collected from one source, which was the only one at Ilindi Ward. Three flamingos and three catfish from Ilindi Playa Lake were caught for laboratory analysis of U content.

Sample preparation and laboratory analysis

Fully matured cassava leaves as well as grains for maize, finger millet and rice were oven-dried at 70 °C to a constant weight, and later ground separately to pass through a 0.5 mm sieve (Okalebo *et al.*, 2002). Rice grains were husked before grinding. Flamingos were dissected and livers, gizzard, skin and flesh were separated according to Faucher *et al.* (2012). Similarly, for catfish, their skin and fillet were separated, after which, both fish and flamingo samples were sun-dried for two days and then oven-dried at 100^{o}C to a constant weight. Dried samples were then ground using domestic blender (HE HOUSE, Model number HE-3380-BL made in China). Salt and soda ash samples were in powder form so they were just sun-dried and sieved through a 0.5mm sieve as recommended by Okalebo *et al.* (2002). Water samples were preserved below freezing temperature at -10 ^{o}C for a month prior to laboratory analysis.

Laboratory analyses were conducted at the Government Chief Chemist Laboratories Agency (GCLA) in Dar es Salaam, Tanzania using Inductively Coupled Plasma-Optical Emission Spectrometry [(ICP-OES) model iCAP 6000 series from Thermo-fishers in England] as per ISO 2171: 2007 method. For the solid samples, 0.5g of maize, finger millet, cassava leaves, rice, salt, soda ash, flamingo and fish samples were weighed separately in an empty test tube by using precision analytical balance. Five ml of hydrogen peroxide (H_2O_2) was added followed by 10 ml of nitric acid. Later, 5ml of hydrogen peroxide was added, making the ratio of nitric acid and hydrogen peroxide to be 1:1.

The samples were block-digested using FOSS Tecator 2040 digester at 180^oC for one hour and then cooled to room temperature (25^oC). Digested samples were then filtered using Whatman filter paper (grade one) and diluted with de-ionized water to the mark of 100ml volumetric flask. The samples were mixed by shaking thoroughly and 10ml of the digested samples were transferred into the vials for analysis by ICP- OES. For water samples, 10ml was mixed with 2ml of 1% HNO_3 and analysed (ISO 2171: 2007). Blank and reference standard samples from SpexCertiprep, USA were used as a quality control measure. The samples, reference standard and blanks were all analyzed in one batch in a randomized order generated by using Microsoft Excel. All reagents were of analytical grade. The detection limit of the ICP- OES instrument was 10ppb.

Statistical analysis

Laboratory data were subjected to analysis of variance and means were compared by Duncan's multiple-range test using the MSTAT-C software (Freed *et al*., 1991).

Results and Discussion

Uranium in surface and underground waters

Table 1 shows concentration of U in selected surface and underground water sources in the studied wards. The highest concentration was found in water sample from Ilindi Playa Lake followed by well water at Ilindi Primary School. These values exceeded the WHO (2012) and USEP (2011) guidelines of 30 µg L^{-1} by a factor of 41 and 3, respectively. Higher values in the lake could be due to its low position relative to the surrounding area, which enable it to accumulate U through run-off. Water in the Ilindi Playa Lake is used primarily by livestock as well as for salt and soda ash production. Whereas, well water near Ilindi Primary School is the source of drinking water for pupils and other local residents in Ilindi Ward. High levels of U above the safety limit are likely to pose a risk to people and livestock of Ilindi Ward. Since natural U is classified as both a radiological and a chemo-toxicological agent (Hakonson-Hayes *et al.,* 2002); prolonged utilization of these waters in Ilindi Ward raise concerns of potential radiological and toxicological risks to human and livestock consumers. This water is also used for production of tea, coffee, and local beverages as well as for cooking foods by local food venders who serve local customers and travelers. Consumption of high U values in water for drinking and other consumptive domestic uses is likely to enhance U daily intake by the people and livestock, which is dangerous (Anke *et al.,* 2009) and should if possible be avoided.

Table 1: Uranium levels in surface and underground waters

Ward	Water source	Coordinates	Uranium level, μg L^{-1}
Ilindi	Shallow well near Ilindi Primary School	05° 56′ 45" S & 035° 32′ 26" E	95.4b
	Shallow well in the village	05° 57′ 08"S & 035° 32′ 41" E	<10e
	Ilindi Playa Lake	05° 57′ 50" S & 035° 31′ 34" E	1233a
Mpamantwa	Shallow well near village office	05° 57′ 19" S & 035° 23′ 11" E	<10e
	Shallow well near Dodoma-Manyoni road	05° 59′ 12" S & 035° 23′ 04" E	<10e
	Spring water seeping from the ground	05° 58′ 20" S & 035° 23′ 38" E	<10e
Bahi	Tributary of Bubu River	05° 58′ 18" S & 035°18′ 24" E	62.9c
	Private tap water	05°58′19.1" S & 035° 18′ 22" E	20.7d
	Private tap water	05° 58′ 20" S & 035° 18′ 21" E	17.6d
Standard error (s.e.)			12.3
Coefficient of variation (CV), %			10.0

The means of uranium level with similar letter are not significantly different at 5% level of probability based on New Duncan's Multiple Range Test (NDMRT).

In Bahi Ward, the highest mean U concentration was found in one of the tributaries of Bubu River (Table 1). Uranium concentration in the river decreased as water flowed downstream, implying that the source of U in the area is from the upstream side that has isolated hills of granites. Concentration of U in the river decreased downstream probably part of it being trapped by soil and organic matter as water flows downstream. Water in the Bubu River, which had the highest concentration of U is used for drinking by domestic animals and for paddy farming. During data collection for this study, children were found playing and swimming within that stream and surrounding fields. This is dangerous as the contaminated water was in direct contact with their skin, which is one of the pathways of U into the body. Although dermal contact is considered a relatively unimportant type of exposure since little of the U will pass across the skin into the blood, U could enter the systemic circulation through open wounds or from embedded U fragments (Burkart *et al*., 2002).

The level of U in tap water from Bahi ward as well as surface and underground waters from Mpamantwa ward (at 17.6 and 20.7 µg L^{-1}respectively) were within the tolerable level (30 µg L^{-1}) recommended by WHO (2012) and USEPA (2011). It implies that water from these sources is safe for consumption by humans and animals.

Uranium in maize, finger millet, rice and cassava leaves

Laboratory analysis showed low levels of U of <10µg kg^{-1} in maize and rice grains (Table 2). This is a sign of insignificant transfer of U from soils to maize and rice tissues. This finding in particular for maize is in line with Vandenhove *et al.* (2007), who found insignificant amounts of U in maize shoots compared to levels found in root tips, indicating poor U translocation from the root tips to maize grains. Sheppard (1980) reported that once U is absorbed by plant roots it is stored as a yellow deposit in the cell nuclei of the meristem. This results in the destruction of the chromatin and cessation of cell nuclear activity, which consequently prevents U translocation to other parts. In most plants, a high concentration of U occurs in roots and the lowest is in fruits or grains (Laroche *et al*., 2005).

The low values of U in rice grains could be associated with the environment in which the crop is grown. In Bahi District, rice is grown by flooding the paddy fields with water during the rainy season and remains with water most of the time up to nearly the time of harvesting. Availability of U under that condition among other factors is governed by the water pH. At a pH lower than 6, U is more readily available for plant uptake and translocation as uranyl cation (UO_2^{2+}), whereas at high pH 6-8 it exists in solution form as hydroxyl and carbonate complexes, which are taken and transferred less readily to various plant tissues (Ebbs *et al*., 1998). Analysis of water in paddy fields showed a pH of 7.3-8.7, which likely contributed to the observed lower uptake of U by rice plants and corresponding translocation to grains.

Table 2: Uranium levels in maize, finger millet and cassava leaves

Ward	Material analysed	No. of samples	Concentration of U, µg kg^{-1}	
			Min-Max	Median
Ilindi & Mpamantwa	Maize grains	18	<10	<10
Ilindi & Mpamantwa	Finger millet grains	18	10.4- 32.0	23.5
Ilindi & Mpamantwa	Cassava leaves	18	<10 -13.83	12.54
Bahi	Rice grains	15	<10	<10

When comparing finger millet with other cereal grains which were all collected from the same Wards, finger millet was observed to contain relatively higher levels of U than maize and rice. The ability of finger millet to accumulate U in grains may be attributed to its root system. Finger millet has permanent adventitious roots developed from the second internodes and above. The roots of a finger millet plant branch laterally (about 1 m^2) interlacing the soil vertically in search of nutrients to supply the plant. In addition, the roots can grow more than 2 m in pursuit of water and soil nutrients (ICRISAT, 2007). Since U contents in the study area increases with depth, this provides finger millet plants an added advantage of taking up more U in their tissues compared to maize and rice.

The U level in finger millet at 10.4 - 32.0µg kg^{-1} may pose a health hazard to humans since the tolerable daily intake (TDI) for soluble U established by the World Health Organization (WHO, 2012) based on the lowest-observed-adverse-effect-level (LOAEL) is 0.6 µg/kg body weight per day. This means when U concentration above TID is consumed daily, a person is at risk of getting cancer or other diseases caused by consuming U. Studies of rats indicate that toxicity from ingested U compounds increases with the solubility of U compound consumed (EFSA, 2009). This implies that its toxic effects in the human body will depend on the solubility of ingested U compounds. This study did not establish the solubility of U that was found in finger millet and thus, hence, it is difficult to predict the potential hazard posed to humans.

For cassava leaves, U values ranged between <10 and 13.83 µg/kg for Ilindi and Mpamantwa Wards, respectively. These concentrations did not differ much from figures reported by WHO (2001), which range from less than 1 up to 11 µg/kg in a variety of tropical staple foods including cassava, banana, maize and sweet potato. This means cassava leaves in the study area are generally safe for human and animal consumption.

Uranium in fish and flamingo

Levels of U in flamingo and catfish were as shown in Table 3. The skin of catfish and liver of flamingo had higher values (17.9 µg kg^{-1} and 31.7 µg kg^{-1}, respectively) than other parts of the body mass, which were found to be below the detection limit. Uranium level in catfish was in line with that established by Faucher *et al.* (2012) in zebra fish (*Danioreri*). Correa *et al.* (2008) also found that within fish gills and skin accumulate the highest U levels. The high U accumulation levels detected in the catfish skin indicate that a significant portion of U could remain adsorbed on the skin or it could be absorbed into skin cells (Correa *et al.*, 2008). The skin is an important organ for U accumulation due to adsorption of the mineral on the skin from the surrounding water or because more U is transferred from the gills to the blood and finally accumulates in the skin (Correa *et al.*, 2008). Thus, prolonged consumption of catfish exposes the communities in the Bahi district to U toxicity.

Table 3: Uranium levels in flamingo and catfish tissues

Part analysed	Uranium levels (µg kg^{-1})
Flamingo liver	31.78a
Flamingo gizzard	<10c
Flamingo muscles	<10c
Flamingo skin	<10c
Cat-fish skin	17.98b
Cat-fish muscle	<10c
Standard error	5.09
Cofficient of variation, (%)	29.3

The means of uranium level with similar letter are not significantly different from each other at 5% level of probability based on New Duncan's Multiple Range Test (NDMRT).

For the flamingo, the highest value of U was found in the liver (31.7 µg kg^{-1}). Strumińska-Parulska *et al*. (2013) also found the highest U content in the liver when assessing polonium, U and plutonium bioaccumulation in marine birds. Higher values of U in the liver arise probably because this organ is responsible for detoxification (Eagles-Smith *et al.*, 2008).

In most waters, sediments acts as a sink for U hence, the concentration of U in sediments and suspended solids are several orders of magnitude higher than in surrounding water (Brunskill and Wilkinson, 1987). The high frequent contact and ingestion of sediment during feeding exposes flamingos to U intake much more than other species which are merely collecting plants and prey from the environment (Zweers *et al.*, 1995). Apart from feeding on sediments, flamingos also eat fish that have adsorbed U. Thus, eating flamingo and fish exposes the community to additional U toxicity. Flamingo is protected bird but in Bahi District its meat is eaten secretly, and is sold in black markets within the country.

Uranium in salt and soda ash

Table 4 shows the concentration of U in soda ash and table salt. The concentrations of U in boiled and sun-dried salt were below the detection limit of the instrument. These results were in line with those reported by Simion *et al*. (2006), who found the concentration of U in iodinated commercially available sodium chloride salt sample to be ≤ 10 ppb.

Table 4. Uranium levels in soda ash and table salt from Ilindi Ward

Products	Uranium levels (µg kg^{-1})
Soda ash	1910 ± 35
Boiled salt	<10
Sun-dried salt	<10

Mean value of soda ash with standard error.

However, the concentration of U in soda ash (Na_2CO_3) was 1, 910 µg kg^{-1} as shown in Table 4. This value was the highest of all samples tested in this study. The reason for this higher value could be due to the affinity of U to carbonate. It has been reported that carbonate in soils increases the mobility of U through the formation of anionic U and CO_3 complexes (Sheppard and Evenden, 1987). In the presence of carbonate, U is solubilized as tetra sodium uranyl tricarbonate ($Na_4UO_2\ (CO_3)_3$). Higher carbonate concentrations could cause some U to precipitate as sodium diuranate (Dry, 2010). This could be the reason for higher levels of U in soda ash. In Bahi District, soda ash is mainly used in food and cooking as a reactive agent. It is also used in animal feeds. It was reported that, some soda ash from Bahi District was sold in Rwanda and Burundi and some is locally consumed in Tanzania. Thus, the problem of U contamination and toxicity is spreading beyond Tanzania. Screening of soda ash for U is recommended before it is sold to consumers.

Mitigation measures against U exposure

This study has revealed the presence of high levels of U in drinking water and in some foodstuffs in Bahi District. People in Bahi District have been exposed to U contaminated food and drinking water for many years unknowingly. This information needs to be disseminated to the public as awareness creation. As a way of disseminating the information to the public, brochures were made and copies were provided to the District Executive Director. Similarly, Ward Executive Officers (WEO) were involved during data collection and feedback was given to them. The government should look for possible mitigation measures in order to ensure the people's safety. This study however, did not establish the status of health of the people in the study area in relation to the presence of U within their environment. The paper also did not address the effects U to the health or people and livestock in Bahi district. This should be a subject of further studies.

Studies indicate that the effects of U in humans and livestock are highly variable, depending not only on the content of ingested U but also on its solubility (Anke *et al*., 2009; EFSA, 2009). Once U is ingested part is deposited in the bones while others are deposited in the kidney, liver, and other soft tissues, which may cause cancer (Schnug *et al.*, 2005; Orloff *et al.*, 2004). Quick assessment of medical records available at Mirembe and Dodoma hospitals, the government hospitals that are about 50 km from the study area, showed some few cases of people from Bahi District with brain, heart, lung, kidney and liver problems. Medical experts who were consulted on this issue indicated their worry of associating those diseases with U exposure. This is because not only U could cause those diseases and thus, a thorough study involving medical experts needs to be done before drawing a conclusion.

Problem of high levels of U in drinking water and food products is not uncommon. It has been reported in a number of countries (Orloff *et al*., 2004, EFSA, 2009) and different techniques are used as mitigation measures. For instance, in the USA in South Carolina, high levels of U content was found in some underground drinking water, exceeding the required standard. People were advised to stop using U contaminated water and alternative water source was supplied by the government (Orloff *et al*., 2004). This technique could also be used to people in Bahi district. Alternatively bioremediation techniques could be used. A study by Finneran *et al.* (2002) showed that soluble U (VI) in groundwater and sediments could be reduced to insoluble U(IV) by microbial reduction. The reduction process precipitates insoluble U(IV) thus becoming immobile. Such reduction is possible by adding acetate to groundwater to stimulate anaerobic condition that enable microbial organisms to reduce soluble U(VI) to insoluble U(IV). This is another possibility could be used to reduce the level of U in drinking water within Bahi district.

Conclusions and Recommendations

Findings from this study show that Soda ash, surface and well waters in Bahi District are highly contaminated with U up to levels exceeding the recommended limit set by WHO (2012) and USEPA (2011) by a factor of more than 41. This implies that such items should not be used for human and animal consumption. Finger millet grains, catfish and flamingo meat from the district also contain relatively high levels of U, above the recommended values. Continuous ingestion of foodstuffs containing such high levels of U exposes the local community and livestock to U risk. Thus, it is recommended that the public should be informed so that safety measures can be taken to minimize such risks.

Not all food commodities that are commonly consumed in the study area were covered by this study. Moreover, this study did not investigate the specific health impacts or such high U levels on the local population within the study area. More studies are therefore necessary to be conducted to analyze U levels in other food groups especially root crops due to their direct contact with U in soils. Further research is also recommended on domestic animals at Ilindi Ward since they use Ilindi playa lake water as the main source of drinking water, which has a high level of U. Another study is recommended to assess hospital records in order to establish incidences of cancer among patients from Bahi District.

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