

HEALTH RISK ASSESSMENT OF HEAVY METALS VIA CONSUMPTION OF CONTAMINATED VEGETABLES CULTIVATED AT DAR ES SALAAM AND MOROGORO REGIONS IN TANZANIA

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Abstract— The main objective of the present work is to focus on determination of heavy metals concentration in vegetables (*Amaranth*) and soil in different locations of two regions Dar es Salaam and Morogoro, so as to establish the risk associated in consumption of the vegetables contaminated with heavy metals. Atomic absorption spectrophotometry was used to determine the levels of these metals in vegetables and soil samples. The highest concentrations in vegetables were found in Arsenic with 1.533 mg/kg followed by chromium 0.211 mg/kg, lead 0.056 mg/kg cadmium 0.005 mg/kg and lastly mercury 0.771 µg/kg. The highest content of chromium were detected in the soil samples from Ngerengere areas (0.328 mg/Kg). The concentration of total chromium in plant samples 0.169 mg/kg is under the maximal recommended limit (2.30 mg/kg). The hazardous quotient (HQ) of the studied heavy metals indicated that except for As contamination, all vegetables were safe with no risk to human health in *Amaranth* consumption. Though by using HQ it showed that the consumption of vegetables grown in Morogoro and Dar es Salaam was nearly free of risks (except for HQ values for As), it is therefore recommended this could not be the guarantee that there is a relative absence of health risks associated with the ingestion of contaminated vegetables. The situation could however change in future depending on the dietary pattern of the community

and the volume of contaminants added to the ecosystems.

Keywords— Heavy metals, vegetables, Morogoro, Toxicity, hazardous quotient

I. INTRODUCTION

Heavy metal contamination of vegetables cannot be underestimated as these foodstuffs are important components of human diet. Vegetables are rich sources of vitamins, minerals, and fibers and also have beneficial antioxidative effects. However, intake of heavy metal-contaminated vegetables may pose a risk to the human health. Globally, heavy metal contamination to widely consumed vegetables has become a major challenge. This is because heavy metals are regarded as serious pollutants of aquatic ecosystems due to their environmental persistence, toxicity, bioaccumulation and biomagnifications in the food chain [1]. Though they are present at low concentration in aquatic ecosystem, the deposits of anthropogenic origin have raised their concentration, creating environmental problems in lakes, rivers, streams as well as households. Some of these pollutants are directly discharged into environment or water bodies by industrial plants and municipal sewage. Other potential source of pollutants are human activities such as mining and agricultural activities [2].

Like many other African countries, Tanzania is blessed of having big fresh water sources like lakes, rivers, ocean and springs. The contamination of these water bodies is very common from the discharge of untreated or partially treated industrial waste, disposal of domestic or sewage drainage [2, 3]. The country also is experiencing impact of environmental degradation. Dar es Salaam city for example, has witnessed a significant increase in population and industrial activities. These have led to conversion of open spaces into industrial residential areas [2]. The

effects have resulted into uncontrolled disposal of domestic and industrial waste and put constraint on access clean water.

The presence of heavy metals in soil can affect the quality of food, groundwater, micro-organisms activity, plant growth [4]. Heavy metals and some trace elements are biologically toxic and can affect and threaten the health of human being owing to their accumulation and persistence in the compartments of the food chain. Heavy metals get into the environment in different ways: with industrial, agricultural and household wastewaters, atmospheric deposits or in the process of extraction of natural resources [5]. The major part of heavy metal accumulates on the surface of soil and in the upper layers of bottom sediments of water basins. Mixed with the substances existing in such upper layers and change their characteristics. Soil reaction (pH) conditions a mobile form of heavy metal amounts and organic substances in sediments acting like a buffer and storing these materials for a long time [6].

Most of the heavy metals are not regarded as essential to human life. Their toxicity depends on several factors including the dose, route of exposure, and chemical species, as well as the age, gender, genetics, and nutritional status of exposed individuals [7]. Because of their high degree of toxicity, metals like arsenic, cadmium, chromium, lead, and mercury rank among the priority metals that are of public health significance [7]. Concern over such incidents of toxicity, has prompted numerous investigations into the metabolism and toxic effects of these four elements [8]. Though it is known that vegetables are rich sources of fibers, minerals and vitamins and also have beneficial antioxidative effects, consumption of heavy metal-contaminated vegetables may pose a risk to the human health. The prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases [5].

For example, lead toxicity has been one of the most noteworthy avoidable causes of neurologic morbidity from an environmental toxin [9 - 11]. The toxicity is a particularly insidious hazard with the potential of causing irreversible health effects [11]. It interferes with a number of body functions primarily affecting the central nervous, hematopoietic, hepatic and renal system producing serious disorders [11].

Much of the cadmium which enters the body by ingestion comes from terrestrial foods. Thus, directly or indirectly, it is the cadmium present in the soil and the transfer of this cadmium to food plants together with the cadmium deposited out of the atmosphere on edible plant parts which establishes the vast majority of human cadmium intake [5, 10]. The kidney is the critical target organ for the general population as well as for occupationally exposed populations. Cadmium is known to accumulate in the human kidney for a relatively long time, from 20 to 30 years, and, at high

doses, is also known to produce health effects on the respiratory system and has been associated with bone disease [7, 12].

The toxicity of mercury depends on its chemical form, and thus symptoms and signs are rather different in exposure to elemental mercury, inorganic mercury compounds, or organic mercury compounds (notably alkylmercury compounds such as methylmercury and ethylmercury salts, and dimethylmercury). Methylmercury is a well-documented neurotoxicant, which may in particular cause adverse effects on the developing brain [10]. Moreover, this compound readily passes both the placental barrier and the blood-brain barrier, therefore, exposure during pregnancy are of highest concern [13,14]. Also, some studies suggest that even small increases in methylmercury exposures may cause adverse effects on the cardiovascular system, thereby leading to increased mortality [10]. Given the importance of cardiovascular diseases worldwide, these findings, although yet to be confirmed, suggest that methylmercury exposures need close attention and additional follow-up. Moreover, methylmercury compounds are considered possibly carcinogenic to humans [10].

For Arsenic, the first symptoms of long-term exposure to high levels of inorganic arsenic (e.g. through drinking-water and food) are usually observed in the skin, and include pigmentation changes, skin lesions and hard patches on the palms and soles of the feet (hyperkeratosis) [5,10]. These occur after a minimum exposure of approximately five years and may be a precursor to skin cancer. In addition to skin cancer, long-term exposure to arsenic may also cause cancers of the bladder and lungs [10].

Chromium enters into various environmental matrices (air, water, and soil) from a wide variety of natural and anthropogenic sources with the largest release coming from industrial establishments. Industries with the largest contribution to chromium release include metal processing, tannery facilities, stainless steel welding and ferrochrome and chrome pigment production. The increase in the environmental concentrations of chromium has been linked to air and wastewater release of chromium, mainly from metallurgical, refractory and chemical industries [7, 15]. The main health problems seen in animals following ingestion of chromium (VI) compounds are irritation and ulcers in the stomach and small intestine, anemia, sperm damage and male reproductive system damage. Chromium (III) compounds are much less toxic and do not appear to cause these problems. Some individuals are extremely sensitive to chromium (VI) or chromium (III), allergic reactions consisting of severe redness and swelling of the skin have been noted [7]. An increase in stomach tumors was observed in humans and animals exposed to chromium (VI) in drinking water. Accidental or intentional ingestion of extremely high doses of chromium (VI) compounds by humans has resulted in

severe respiratory, cardiovascular, gastrointestinal, hematological, hepatic, renal, and neurological effects as part of the sequel leading to death or in patients who survived because of medical treatment [16].

The main objectives of the present work are to focus on determination of heavy metals concentration in vegetables and soil where two neighboring regions Dar es Salaam and Morogoro were used as sampling sites. We intend to assess the risk associated to consumption of the contaminated commonly used vegetables (*Amaranthus*). At the end recommendations on human diet foodstuff to assure significant improvement in food safety and advice policy makers and consumers was made appropriately.

II. MATERIAL AND METHODS

A. Study Area and Sampling Locations

This study was conducted at Dar es Salaam and Morogoro regions (Fig 1). The selection of the study areas was done based on the geographical location of the regions in relation to industrial settings in the city/municipality.

Dar es Salaam is located at 6°48' south, 39°17' East on a natural harbour on the eastern coast of Africa, along the Indian Ocean coast and covers a total area of 139.3 km². Administratively, the city is divided into three municipalities; namely Kinondoni, Temeke and Ilala. Dar es Salaam is the commercial city of the country; it is one of the fastest growing cities in Africa. It has a population of 4,364,541 [17], with a population increase of 5.6 percent per year from 2002 to 2012, the city is the third fastest growing in Africa (ninth fastest in the world), after Bamako and Lagos [18]. The average income earner in Dar es Salaam is responsible for four people, this is a significant burden given a low level of earnings. Most workers are self-employed rather than wage earners. The majority of the poor is proprietors of small businesses and account for 20 to 40% depending on the area of the city. In another study [19], it was established that petty traders or street vendors selling items including vegetables ranges from 15 to 20% and consists mainly of male youth between 20 and 29 years of age. Morogoro is a city with a population of 315,866 [17] in the eastern part of Tanzania 196 km west of Dar es Salaam city, lies at the base of the Uluguru Mountain and is a centre of agriculture in the region.

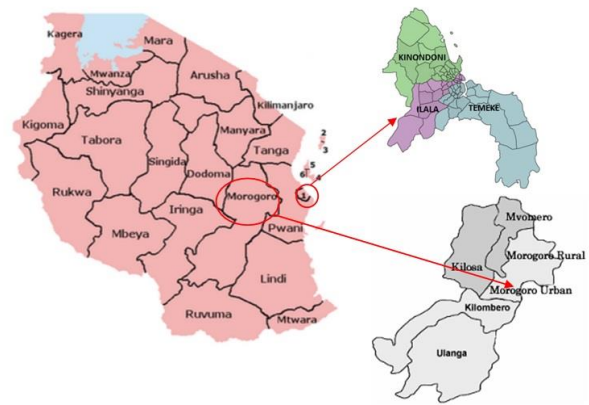


Figure 1: Map of Tanzania and Morogoro Region and Dar es Salaam City

B. Sample Collection

Samples of soil, and vegetables (*amaranth*) were appropriately were sampled at the same location. About 500 g vegetable was bought from local gardens and same time about 500 g of soil sample were taken from the same site.

C. Sampling of Vegetable

The vegetable samples were washed with distilled water to eliminate air-borne pollutants. The leafy stalks were removed from all samples and these were sliced and dried on a sheet of paper to eliminate excess moisture. Once dried, each sample was weighed and oven-dried at 60°C to constant weight. Each oven-dried sample was ground in a mortar until it could pass through a 60 mesh sieve. The samples were then stored in a clean, dry, stoppered glass container before analysis. The method adopted is according to literature [20] the dry ashing method is appropriately used followed by atomic absorption spectrophotometric analysis as stipulated in the Perkin-Elmer manual for atomic absorption spectrophotometry [21].

D. Vegetable Sample Analysis

Each determination was carried out by accurately measuring a sample of 1 g of a ground sample in a crucible. The crucible with its content was placed in a furnace and ashed at 450°C for 12 h. The ash was digested with 5 ml of 20% (v/v) AnalaR HCl solution. The residue was filtered into a 50 ml volumetric flask using Whatman filter paper No. 44 and the solution was made to the mark with demonized water. Atomic absorption spectrophotometry (AAS) was used for the determination of the heavy metals. Each sample solution was run in duplicate to ensure the repeatability of the obtained results. The same procedure was followed for each sample and the appropriate dilution factors were used in the calculation.

E. Sampling of Soil Samples

A soil sample were collected at depth of about 15cm using hand auger, stored in polyethylene bags and were oven dried at 60°C for 2days, followed by grinding with mortar and pestle and sieved using a 2 mm sieve. About 1.0g of the oven dried sample ground sample was weighed using a top loading balance and placed in a 250 ml beaker which has been previously washed with nitric acid and distilled water. The sample was reacted with sample was reacted with 5ml of HNO₃, 15ml of concentrated H₂SO₄ and 0.3ml of HClO₄ using dropping pipette. The mixture was digested in a fume cupboard, heating continued until a dense white fume appeared which was then ingested for 15 minutes, set aside to cool and diluted with distilled water. The mixture was filtered through acid washed Whattmann No.44 filter paper into a 50ml volumetric flask and diluted to mark volume [22]. The sample solution was then aspirated into the Atomic Absorption Spectroscopic machine at intervals.

F. The Atomic Spectrophotometer (AAS)

Determination

Prepared samples was aspirated into the instrument after all necessary set-up and standardization procedures. All metals were determined by Atomic Absorption Spectrophotometer (AAS), Pelkin-Elmer AA700. The instrument was calibrated with standard chemical solutions prepared from commercially available chemicals (Merck, Germany). For analytical quality assurance, the result of each metal was corrected by subtracting the value from the blank. Analytical blanks were run in the similar way as that of the samples and the concentrations were determined using the standard solutions prepared in the same matrix. After every five sample readings, standards were run to make sure that the obtained results were within range. Other AAS conditions employed in these determinations were set according to operation manual [21].

III. RESULTS AND DISCUSSION

The concentrations of heavy metals found in vegetables and soil sampled from cultivated sites are summarized in Table 1.

Table 1: Levels of Heavy Metals in Vegetables and Soil in Sampling Sites

Dar es Salaam Sampling Sites						
Met al	Type	Makum busho	Mbe zi Luis	Kigo go	Kiba sila	Taz ara
Cd (mg/kg)	Amar anth	0.005	0.005	0.003	0.004	bd
	Soil	bd	bd	0.009	0.011	0.00

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As (mg/kg)	Amar anth	0.795	0.750	0.001	0.002	0.384
	Soil	bd	0.002	bd	bd	bd
Pb (mg/kg)	Amar anth	0.008	bd	0.056	bd	0.023
	Soil	bd	bd	0.003	bd	0.161
Cr (mg/kg)	Amar anth	0.113	0.148	0.163	0.160	0.140
	Soil	0.115	0.273	0.071	0.058	0.110
Hg (µg/kg)	Amar anth	bd	bd	bd	0.771	bd
	Soil	0.211	0.739	1.875	0.508	5.225

Morogoro Sampling Sites					
		Ngerengere	Tung i	Mindu Dam	Mji Mpy a
Cd (mg/kg)	Amar anth	0.002	0.008	0.003	0.007
	Soil	bd	0.002	bd	0.006
As (mg/kg)	Amar anth	0.085	0.904	0.065	1.533
	Soil	bd	bd	bd	bd
Pb (mg/kg)	Amar anth	0.016	bd	0.023	0.014
	Soil	bd	bd	0.017	bd
Cr (mg/kg)	Amar anth	0.045	0.169	0.023	0.211
	Soil	0.328	0.225	0.158	0.183
Hg (µg/kg)	Amar anth	0.919	2.298	bd	bd
	Soil	bd	bd	bd	bd

The results show that the concentrations in vegetables the highest was found in Arsenic with 1.533 mg/kg followed by chromium 0.211 mg/kg, lead 0.056 mg/kg cadmium 0.005 mg/kg and lastly mercury 0.771 µg/kg. The As concentrations were many folds higher than the WHO recommended permissible limit for drinking water (0.01 mg l⁻¹) and FAO permissible limit for irrigation water (0.10 mg l⁻¹). The total arsenic concentrations in the studied samples ranged from 0.001 mg kg⁻¹ (Kigogo ward in Dar es Salaam city to 1.533 mg kg⁻¹ at Mji Mpya Morogoro region.

The concentration of arsenic in soil is considerably lower ranging from bd to the majority of sampling sites to 0.002 mg/kg found at Mbezi Luis in Dar es Salaam city. This is lower than levels detected in vegetables. This is contrary to what was observed earlier [23], which indicated that the concentrations of arsenic in soil may be 10 to 1000 times greater than their concentrations in plants growing on that soil. Because of this, failure to remove soil particles that adhere or

become trapped on the outside surfaces of garden crops can substantially increase dietary lead and arsenic obtained by eating garden plants.

It was not surprising to detect high level of arsenic in Mbezi Luis (Dar es Salaam) because it was found in literature that arsenic often is high around existing metal ore smelters and former smelter sites due to lead- and arsenic-rich ash emitted from smokestacks, or lead- and arsenic-rich dusts blown off ore and slag piles [23]. Metal ores frequently contain arsenic which is released during smelting as arsenic-rich gases. This is very common activity at Mbezi Luis sampling site.

The highest content of chromium were detected in the soil samples from Ngerengere areas (0.328 mg/Kg). The concentration of total chromium in plant samples 0.169 mg/kg is under the maximal recommended limit (2.30 mg/kg recommended by FAO/WHO [24]. The variation in chromium concentrations in vegetables and soil of the same site may be ascribed to the differences in their morphology and physiology for chromium uptake, exclusion, accumulation and retention as have been described in other studies [25, 26].

In another studies [27, 28] assess the ability to uptake Cr from the soil by different organs of *Populous alba*, *Morus alba* and *Amaranthus dubius* leaves accumulated higher levels of Cr than stems or roots. This confirms our results that *amaranth* vegetables can accumulate more chromium in leaves because most of the sampling sites levels of chromium in vegetables. The results obtained declared that, lead values have irregular fluctuation between different sampling sites (Table 1). The amount of Pb in the samples vegetables was found to be higher at Kigogo (0.056 mg/kg) followed by sample from Mindu Dam (0.023 mg/kg). Soil sample ranges from bd to 0.161 mg/kg which was detected at Tazara in Dar es Salaam city.

A study [29] on water sample around Mindu Dam have indicated that textile effluents surrounding the dam contain Cr metal in excess compared to other metals and have the lowest Pb concentrations. Chromium is mainly found in waste water from the dyeing process in textile industries and Chromium salt used in the tanning process in the tannery industries. This could be the source of Cr levels in sampled from Mindu dam both in soil as well as in vegetable.

Increase of Pb levels in Kigogo area may be attributed to heavily traffic in this area which lead to the accumulation of Pb emitted from cars exhaustions as well as garages around river Msimbazi where the vegetables are grown. However, the results are lower than values obtained by [30], who reported the Pb concentration (17.54–25.00 mg/kg) in vegetables grown in industrial areas. According to another study [31], it was reported that the level of Pb (6.77 mg/kg) in vegetables irrigated with mixtures of wastewater and sewage from Zimbabwe to be higher than WHO safe limit (2 mg/kg).

It is well documented that Pb first enters into plants like vegetables through contaminated soil and by lead

dust accumulating on the plants [26, 28]. The lead is taken up by plants through roots and leaves. Lead from the atmosphere that lands on soils has low mobility and tends to stay in the top inch of soil. Therefore, shallow-rooted plants, such as grasses and common vegetables, are particularly vulnerable to picking up lead contamination that originated in the atmosphere [28]. Therefore, though the level of lead in the soil may be below detection but this can be due to quick uptake of plants of that small amount exists.

Cadmium all the samples analyzed, their level was observed to be very low varying between below detection limit and 0.08 mg/kg (Tazara in Dar es Salaam city). Various values have been previously reported [32] for leafy vegetables which include 0.09 mg/kg in pumpkin plants and 0.06 mg/kg in cabbages. According to [33], FAO/WHO the safe limit for Cd consumption in vegetables is 0.2 mg/kg therefore, the values in this study is well below allowable values. The source of Cadmium in the samples could be due to used batteries, pigments, fertilizers, detergents and it is also present in refined petroleum products.

Mercury is more toxic than Cd and Pb and causes serious health problems such as loss of vision, hearing and mental retardation and finally death occurs. Mercury was not detected in most of sampling sites at Dar es Salaam city except Kibasila which recorded 0.771 µg/kg while the highest level was detected at Tungi (Morogoro) with 2.298 µg/kg. The recorded values exceeded the permissible levels (0.03 µg/g) reported by the WHO [8], though lower than the value detected recently (Pan, *et al.*, 2016) which detected 0.002 mg/kg in *Amaranthus tricolor* L. in China.

Surprisingly, all soil samples from Morogoro did not detect presence of mercury while all samples from Dar es Salaam range between 0.211 µg/kg (Makumbusho) to 1.875 µg/kg (at Kigogo). It is well documented [34] that the concentrations of Hg in soil are greatly influenced by parent material and soil properties, including organic matter, soil microbes, and soil pH as well as human activities, such as non-ferrous mining, petroleum refining and fossil fuel combustion, discharge of wastes from industry production, and applications of fertilizers. Some studies [35, 36], suggested that anthropogenic sources are leading to a general increase in Hg on local, regional and global scales [35].

Health Risk from Consuming Vegetable

The health risks associated with metals ingested through vegetable consumption were assessed using hazard quotient (HQ) [37, 38]. This is a ratio of determined dose to the reference dose (RfD) (Eq. 1).

$$HQ = (Div) \times (C_{metal}) / R_{fD} \times B_o \dots\dots\dots(1)$$

Where:

Div is the daily intake of vegetables ((kg/person)/day),

C_{metal} is the concentration of metal in the vegetable (mg/kg),
 R_{FD} is the oral reference dose for the metal (mg/kg per day)
 B_0 is the human body mass (kg).

The RfD is an estimation of the daily exposure to which the human population is likely to be without any appreciable risk of deleterious effects during a lifetime. The values of R_{FD} for heavy metals were taken from Integrated Risk Information System (2003) and Department of Environment, Food and Rural Affairs [38, 39] The HQ is a highly conservative and relative index. When HQ is < 1 , there is no obvious risk from the substance over a lifetime of exposure, while HQ is > 1 , the toxicant may produce an adverse effect. The Rfd of Cd, Pb, As, Cr and Hg used are 1×10^{-2} , 3.3×10^{-3} , 3×10^{-4} , 1.5 and 3×10^{-4} (mg/kg)/day respectively [38 – 40] The Body weight (B_0) is an average body weight of African male which is taken as 57 kg [41]. According to another study [42] an average man of about 50 - 57 Kg needs a 55 g of vegetable per day. Taking the highest concentration of each metal, the estimation of HQ of each region is summarized on Table 2.

Table 2: Highest Concentration and Respective HQ of Sampling Regions (mg/kg)

Region	Cd	Pb	As	Cr	Hg
Dar es Salaam (Conc)	0.005	0.056	0.795	0.163	7.771×10^{-4}
HQ (Dar es Salaam)	0.0025	0.08	1.33	0.005	0.001
Morogoro (Conc)	0.008	0.023	1.533	0.211	2.298×10^{-3}
HQ (Morogoro)	0.004	0.03	2.56	0.005	0.003

From the result, the HQ values of all heavy metals in all vegetables were all below the one (1) (except for As for both Morogoro and Dar es Salaam). In different study [40] the HQ of As metal found in Spinach as high as 5.30. This high HQ for As observed in vegetable consumed had greatest potential to pose health risk to the consumer. The results indicated that those living around the Makumbusho in Dar es Salaam, Mji mpya and Tungi both in Morogoro were probably exposed to some potential health risk through the intake of As via consuming locally grown *Amaranth*. Even though there was no apparent risk when each metal was analyzed individually, the potential risk could be multiplied when considering all heavy metals.

Although HQ was higher for Pb in sample neither population suffered from ingestion of vegetables contaminated with this heavy metals. Higher HQ for Pb were also reported elsewhere [42].

IV. CONCLUSION

Although the concentrations of the metals established for the vegetables are lower than those permitted by FAO/WHO, what matters in the long run is the quantities consumed and the frequency of intake. There is a cumulative effect on sustained intake of heavy metals, as they are not easily removed from the body. The low presence of heavy metals in the vegetables sampled at the time of this study cannot guarantee the safety of the vegetables always for consumption, regular monitoring should be conducted to detect increasing levels of toxic heavy metals in vegetables. Many rural and urban low-income families in Tanzania consume large quantities of vegetables on a daily basis and this exposes them to the health risks associated with heavy metals ingestion. The high HQ for As observed in vegetable consumed had greatest potential to pose health risk to the consumer. The results indicated that those living around the Makumbusho in Dar es Salaam, Mji mpya and Tungi both in Morogoro were probably exposed to some potential health risk through the intake of As via consuming locally grown *Amaranth*. However, this HQ method considers only exposure to heavy metals via consumption of vegetables, without taking into account other routes like dermal contact, soil ingestion and other factors such as the presence of agrochemicals and herbicide molecules. In order to assess the overall potential for non-carcinogenic effects posed by more than one HM, a Hazard Index (HI) approach can be developed based on the EPA's Guidelines for Health Risk Assessment of Chemical Mixtures.

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