ASSESSMENT OF SELECTED HEAVY METALS AND ESTIMATION OF HUMAN HEALTH RISK IN SOME COMMONLY CONSUMED FISH IN ABEOKUTA, OGUN - STATE, NIGERIA

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ABSTRACT
Fish is a major source of food in many parts of the world because they contribute to solving the global food problem and provide the well-known proteins, minerals, vitamin and trace elements content. Concentrations of iron (Fe), zinc (Zn), copper (Cu), nickel (Ni), lead (Pb) and cadmium (Cd) were determined in various parts of the fish (liver, gill, bone and muscle) of Mackerel (Scomber scombrus), Sardine (Sardinella longiceps), Hake (Merluccius merluccius) and Express (Platax scalaris) which were purchased from two major markets in Abeokuta, Ogun State, Nigeria. Heavy metal levels in the fish samples were analyzed by Atomic Absorption Spectrophotometer (AAS Buck 211). Health risks related to Fe, Zn, Cu, Ni, Pb and Cd were assessed based on target hazard quotient (THQ). Results obtained showed variations in the levels of heavy metals across the various tissues in the fish samples. The concentration of Fe was highest in all the tissues in the fish samples with the gill recording the highest level. There were higher concentrations of heavy metals in the gills than in other tissues of the fish samples. The trend of heavy metal levels in the tissues was found to be Fe > Zn > Cu > Ni > Pb > Cd. Highest levels of Pb and Cd in the muscle were 1.92 ± 0.04 and 0.64 ± 0.04 mg/kg in Merlucciusmerluccius and Scomberscombrus respectively. The concentrations of Pb and Cd were higher in the muscles of all the samples than European Community and Food and Agriculture Organization maximum permissible levels. The health risks from Ni and Pb were found highest among all the heavy metals in the fish samples. THQ values of Cu and Cd in Sardinelongiceps and Cu in Merlucciusmerluccius were higher than 1. Health risk assessment of Ni and Pb in all the fish studied and Cd in Sardinelongiceps indicates that their consumption may be unsafe.
Keywords: Heavy metals; Target Hazard Quotient; Maximum permissible level; Health risk assessment.

Introduction
Global food problem is potentially solved by the existence of fish and fish products which are important for human consumption in many parts of the world. Fish products provide proteins, minerals, vitamins and trace elements contents in human diet. Heavy metals from anthropogenic sources are continually released into the aquatic and terrestrial ecosystems. Contamination with heavy metals is a serious threat due to their toxicity, bioaccumulation and biomagnification in the food chain [1] and also its non-biodegradable properties in food [2]. Industrial wastes and mining can create a potential source of heavy metal pollution in aquatic environment [3, 4]. Heavy metal pollution in rivers is a threat to public water supplies and also to consumer of fishery sources [5]. Heavy metals enter into fish bodies by three possible ways: gills, digestive tract and body surface. The gills are considered as the significant site for direct uptake of metals from the water [6, 7], though the body surface is normally estimated to take minor part in uptake of heavy metals in fish [8]. Human health may be affected by organic and inorganic pollutants associated with aquatic systems by consumption of contaminated fish and other aquatic foods from the environment [9]. Heavy metals might accumulate up to toxic concentration under certain environmental conditions and cause ecological damage [10]. Metals such as iron, copper, zinc and manganese are essential metals since they play important role in biological systems, whereas mercury, lead and cadmium are toxic even in trace amounts. The essential metals can also produce toxic effects when present in high concentrations [11]. Fish, among other animals are used to determine the health condition of aquatic ecosystem [12, 13]. Heavy metals in fish come mainly from their diet, and
levels of bioaccumulation of contaminants are higher in fish which are higher in the food chain \[14\]. The pattern of heavy metal uptake in fish varies in different species depending on factors such as lipid content in the tissue, mode of feeding and age of the fish \[15\]. Different factors such as physical and chemical properties of water as well as seasonal changes are the reasons for significant augmentation of metals in different fish tissues \[16, 17\]. Metal residual problems in the fish epithelium are stern, because of the presence of higher metal concentrations in water and sediments \[18\]. The awareness of the Nigerian populace of the benefits of fish consumption appears to have increased in recent times. Therefore, it is pertinent to investigate the concentrations of heavy metals in some regularly consumed fish from the aquatic environment to establish the level of contamination and risks to human health.

Experiment

Materials
All the chemicals used were of analar grade. Concentrated Hydrochloric acid, Nitric acid and Sodium hydroxide were obtained commercially from BDH, Fish samples, Mackerel (Scomber scombrus), Sardine (Sardinella longiceps), Express (Platix scalaris) and Hake (Merluccius merluccius) were purchased from two major markets (Lafenwa and Obantoko) in Abeokuta, Ogun State, Nigeria.

Sample treatment
The gills, bones, scales, livers and muscles were removed with stainless steel knife, washed with distilled water, mopped dry and weighed. The different parts were dried to constant weights in the oven at temperature of 70 – 80°C. The dried samples were crushed into a fine powdered form by using pestle and mortar and then sieved.

Digestion of fish samples
Each ground portion (gill, bone, scale, liver and muscle) from the various fish samples was treated by acid digestion. Into 100 mL conical flask was weighed 0.5 g of each sample and then digested with 10 mL concentrated HNO\(_3\) on a hotplate at a temperature range between 60 – 80 °C for a period of 8 hours. Each digested sample was filtered and made up to 20 mL with 5% nitric acid solution. The solutions obtained were transferred into the pre-labelled sample bottles for metal analysis using Atomic Absorption Spectrophotometer BUCK 211.

Blank Digestion
The blank digestion was carried out with 10 mL of concentrated nitric acid on a hotplate at a temperature of 60 – 80 °C for a period of 8 hours. The digested blank was made up 20 mL with 5% nitric acid solution and transferred into sample bottles.

Human health risk assessment
Health risk assessment is a very important tool to evaluate the consequences of human action and measures the adverse effect to public health. Results acquired from the muscle of fish samples were used in the determination of the health risk from consumption of the fish samples.

Target Hazard Quotient
Concentrations of heavy metals in the muscle of the various fish samples was employed to estimate the daily intake of heavy metals using Equation 1:

\[
THQ = \left(\frac{EFr \times ED \times FiR \times C}{RfD \times WaB \times TA}\right)^{10^{-3}}
\]

where, THQ is the Target Hazard Quotient; EFr is the Exposure Frequency (350 days/year); ED is Exposure Duration (54.4 years), equivalent to the average lifetime (life expectancy for Nigerian adult); FiR is the Fish ingestion Rate (20 g/day/Nigerian person); C is the concentration of metal in the muscle of fish (µg/g); RfDs is the oral Reference Doses: Fe = 0.7, Zn = 0.3, Cu = 0.04, Ni = 0.02, Pb = 0.0005 and Cd = 0.001 mg/kg respectively \[19\]; WaB is the average body weight (60.7 kg) and TA is the average exposure time for non-carcinogens (365 days/year) \[19\].

Estimation of the allowable daily consumption rates (CR\(_{\text{lim}}\))
In order to determine the allowable daily consumption rate of the fish samples, equation 2 was applied. Unit was expressed in kg/day.

\[
CR_{\text{lim}} = \frac{RfD \times Bw}{Cm}
\]

Where, CR\(_{\text{lim}}\) is the maximum safe daily consumption rate of fish (kg/d); RfD is the reference doses of metal (mg/kg/d); Bw is the average consumer body weight (60.7 kg); \(C_m\) is the measured concentration of chemical in the
fish (mg/g). RfD values were found on the basis of daily consumption of metals over a life span that would not be supposed to cause any adverse effects on health of human [19].

**Estimation of allowable weekly and monthly consumption rates (CR<sub>w</sub> and CR<sub>m</sub>)**

The allowable weekly (CR<sub>w</sub>) and monthly consumption rates (CR<sub>m</sub>) were estimated using Equations 3 and 4 respectively:

\[
CR_w = CR_{lim} \times 7
\]

Where, CR<sub>w</sub> is the maximum weekly consumption rate of fish (kg/d) over 7 days per week.

\[
CR_m = (CR_w \times Tap) \times MS^{-1}
\]

Where, CR<sub>m</sub> is the maximum allowable fish consumption rate (meals month<sup>−1</sup>), CR<sub>lim</sub> is the maximum weekly consumption rate of fish (kg/week), Tap is the average weeks in a month (taken as 4.3 week/month<sup>−1</sup>), and MS is the meal size, taken as 227 g (8 oz) for an adult [19].

**Hazard Index (HI)**

For the risk assessment of multiple heavy metals contained in fish, a total hazard index (HI) was estimated using Equation 5:

\[
HI = THQ (Fe) + THQ (Zn) + THQ (Cu) + THQ (Ni)
\]

where, THQ is the target hazard quotient of an individual element of heavy metals and HI is the total hazard index [19].

### Results and Discussion

| Fish species                  | Tissues   | Fe       | Zn       | Cu       | Ni       | Pb       | Cd       |
|------------------------------|-----------|----------|----------|----------|----------|----------|----------|
| **Scomber scombrus** (mg/kg) | Bone      | 5.96±0.08<sup>a</sup> | 2.64±0.12<sup>a</sup> | 1.44±0.80<sup>a</sup> | 1.04±0.04<sup>a</sup> | 0.72±0.08<sup>a</sup> | ND       |
|                              | Gill      | 6.04±0.04<sup>c</sup> | 3.00±2.80<sup>c</sup> | 2.24±0.80<sup>c</sup> | 1.80±0.12<sup>c</sup> | 1.24±0.04<sup>c</sup> | ND       |
|                              | Liver     | 5.92±0.04<sup>d</sup> | 3.12±0.08<sup>d</sup> | 2.04±0.08<sup>d</sup> | 1.44±0.04<sup>d</sup> | 1.28±0.08<sup>d</sup> | 0.64±0.04<sup>d</sup> |
|                              | Muscle    | 5.48±0.24<sup>c</sup> | 2.76±0.20<sup>d</sup> | 1.72±0.00<sup>d</sup> | 1.28±0.20<sup>d</sup> | 0.36±0.08<sup>d</sup> | ND       |
| **Sardinella longiceps** (mg/kg) | Bone      | 6.12±0.08<sup>c</sup> | 3.36±0.00<sup>c</sup> | 2.52±0.00<sup>c</sup> | 1.92±0.00<sup>c</sup> | 0.84±0.08<sup>c</sup> | ND       |
|                              | Gill      | 6.28±0.08<sup>c</sup> | 4.56±0.08<sup>c</sup> | 3.44±0.12<sup>d</sup> | 2.96±0.04<sup>c</sup> | 1.68±0.04<sup>c</sup> | ND       |
|                              | Liver     | 4.84±0.04<sup>f</sup> | 3.98±0.08<sup>f</sup> | 3.32±0.04<sup>d</sup> | 2.92±0.12<sup>f</sup> | 1.76±0.08<sup>d</sup> | 0.60±0.40<sup>g</sup> |
|                              | Muscle    | 5.88±6.00<sup>c</sup> | 3.16±6.00<sup>d</sup> | 2.64±0.48<sup>d</sup> | 2.56±0.04<sup>d</sup> | 1.28±0.04<sup>d</sup> | 0.60±4.00<sup>h</sup> |
| **Platx scalaris** (mg/kg)  | Bone      | 4.00±4.00<sup>f</sup> | 2.84±0.08<sup>d</sup> | 2.28±0.00<sup>c</sup> | 1.52±0.00<sup>c</sup> | 0.88±0.04<sup>c</sup> | ND       |
|                              | Gill      | 4.92±4.40<sup>d</sup> | 3.28±0.04<sup>d</sup> | 2.44±0.08<sup>d</sup> | 2.28±0.00<sup>d</sup> | 1.76±0.04<sup>d</sup> | ND       |
|                              | Liver     | 4.00±12.0<sup>c</sup> | 3.36±0.04<sup>d</sup> | 2.28±0.16<sup>c</sup> | 1.88±0.08<sup>c</sup> | 1.44±0.08<sup>d</sup> | ND       |
|                              | Muscle    | 4.00±4.00<sup>f</sup> | 2.84±0.00<sup>c</sup> | 2.24±0.16<sup>c</sup> | 1.88±0.08<sup>g</sup> | 1.36±0.04<sup>g</sup> | ND       |
| **Merluccius merluccius** (mg/kg) | Bone      | 5.48±0.40<sup>d</sup> | 4.32±0.28<sup>d</sup> | 3.68±0.00<sup>d</sup> | 1.92±0.32<sup>d</sup> | 1.24±0.04<sup>d</sup> | ND       |
|                              | Gill      | 6.08±0.28<sup>c</sup> | 4.92±0.04<sup>d</sup> | 4.08±0.04<sup>c</sup> | 3.28±0.08<sup>c</sup> | 2.32±0.00<sup>c</sup> | ND       |
|                              | Liver     | 5.96±0.04<sup>d</sup> | 4.72±0.08<sup>b</sup> | 3.68±0.08<sup>b</sup> | 2.40±0.08<sup>b</sup> | 2.08±0.04<sup>b</sup> | ND       |
|                              | Muscle    | 5.48±0.8<sup>d</sup> | 4.32±0.28<sup>b</sup> | 3.68±0.04<sup>b</sup> | 2.32±0.08<sup>b</sup> | 1.92±0.04<sup>b</sup> | ND       |

Values expressed as Mean ± Standard deviation with different superscripts in column are significantly different (p<0.05)

The mean concentrations (mg/kg) of heavy metals in various fish species studied are represented in Table 1. In *Scomber scombrus*, Fe concentration was the highest in bone, gill, liver and muscle with the values ranging from 5.48 ± 0.24 to 6.04 ± 0.04 mg/kg. Cd (0.64 ± 0.04 mg/kg) was only detected in the liver. The liver had the highest levels of Zn, Pb and Cd. The gill had the highest concentration of Cu. Zinc accumulates in the gills of fish and this designates a depressing effect on tissue respiration leading to hypoxia which results in death [20]. The level of Fe in the bone was significantly different from that in the gill and muscle. There was a significant decrease in the Zn and Ni concentrations of the bone from that...
of the gill and liver. The level of Pb in the liver was significantly higher than the bone and gill. In *Sardinella longiceps*, Fe concentration (6.28 ± 0.08 mg/kg) was highest in the gill. Cd was detected in liver and muscle. The concentrations of Fe, Zn, Cu and Ni were highest in the gill. The liver had the highest levels of Pb and Cd. In addition, Cd was not detected in the bone and gill. There was a significant decrease in the Fe concentrations in the bone from that in the other tissues. The level of Cu in the bone was also significantly different from that in the gill and liver. The level of Ni in the liver was significantly different from that in the bone and gill. There was significant decrease in Pb concentration in the liver from those in the bone and muscle (Table 1).

In *Platax scalaris*, Fe concentration was highest in all the tissues while Cd was not detected. The gill had the highest levels of Fe, Cu, Ni and Pb. The concentration of Zn was highest in the liver. There was significant increase in Fe concentration in the muscle from the bone and liver. The level of Zn in the bone was significantly reduced in the muscle. There was significant decrease in Cu and Pb concentrations of the gill from the bone and muscle. Ni concentration of the bone was significantly increased than that in the muscle, liver and gill (Table 1).

In *Merluccius merluccius*, Fe concentration was highest in bone, gill, liver and muscle while Cd was not detected. The highest concentrations of Fe, Zn, Cu, Ni and Pb were found in the gill. The lowest concentrations of the metals studied were found in the bone. There was significant decrease in Ni concentration of the gill from the bone, muscle and liver. The level of Pb in the liver was significantly different from the bone and gill (Table 1).

### Table 2: Standards of maximum permissible level of heavy metals in the fish samples and values from this study (mg/kg).

| References     | Fe  | Zn  | Cu  | Ni   | Pb  | Cd  |
|----------------|-----|-----|-----|------|-----|-----|
| WHO (2006)     | 14.80 | 12.00 | 7.80 | 5.48  | 3.44 | -   |
| EC (2005)      | -   | -   | -   | 0.2   | 0.05 | -   |
| FAO (2003)     | -   | -   | -   | 0.5-0.6 | 0.2 | 0.01 |
| EU (2001) µg/g | -   | 50  | 20  | 0.5-10 | 0.2 | 0.2 |
| FEPA (2003)    | -   | -   | -   | 0.1   | -   | -   |
| This study     | 6.12 | 4.92 | 4.08 | 3.28  | 2.32 | 0.64 |

The levels of Fe, Zn and Cu determined in all the tissues of the fish samples studied were lower than the maximum in the guidelines listed in Table 2. All the tissues had elevated levels of Ni beyond the maximum permissible level (0.5 - 0.6 mg/kg) recommended by FAO [23]. All the tissues in the fish studied displayed levels of Pb (0.72 ± 0.08 – 2.32 ± 0.00 mg/kg) higher than the permissible level of 0.2 mg/kg [25]. Cd was detected in the liver and muscle of *Sardinella longiceps* while it was found only in the liver of *Scomber scombrus*. The highest concentration of Cd (0.6 ± 0.4 mg/kg) found in the muscle was higher than the value proposed in the literature (Table 2).

### Table 3: Mean concentrations of heavy metals in the muscle of selected fish (mg/kg)

| Sample                  | Fe     | Zn     | Cu     | Ni     | Pb     | Cd     |
|-------------------------|--------|--------|--------|--------|--------|--------|
| *Scomber scombrus*      | 5.48±0.24 | 2.76±0.20 | 1.72±4.00 | 1.28±0.20 | 0.36±0.08 | ND     |
| *Sardinella longiceps*  | 5.88±6.00 | 3.16±6.80 | 2.64±0.48 | 2.56±0.04 | 1.28±0.04 | 0.6±0.4 |
| *Platax scalaris*       | 4.00±4.00 | 2.84±0.80 | 2.24±0.16 | 1.88±0.08 | 1.36±0.04 | ND     |
| *Merluccius merluccius* | 5.48±0.80 | 4.32±0.28 | 3.68±0.04 | 2.32±0.08 | 1.92±0.04 | ND     |
Table 3 shows the concentrations of heavy metals in the muscle of the selected fish (mg/kg). The muscle is the edible part of the fish and the different fish species displayed varied levels of heavy metals in their muscles. This could be due to species dependence of metal bioaccumulation, feeding habits and habitats [29] in addition to size, and age [30]. The levels of Pb and Cd obtained were higher than the limit values proposed by European Community [25] and could be said to make the fish samples unfit for consumption, though large quantities of the fish species (100 – 880 g) have to be taken daily by a person (Table 4) for it to be harmful to human health [31]. Among all the fish studied, Sardinella longiceps has the highest concentrations of Fe, Ni and Cd while highest concentrations of Zn, Cu and Pb were found in Merluccius merluccius.

Table 4: Allowable consumption rates of heavy metals in the muscle of fish samples

| Fish specie             | Fe   | Zn   | Cu   | Ni   | Pb   | Cd   |
|-------------------------|------|------|------|------|------|------|
| Daily [CR<sub>lim</sub> (kg/d)] |
| Scomberscombrus         | 7.75 | 6.59 | 1.41 | 0.95 | 0.08 | ND   |
| Sardinellalongiceps     | 7.23 | 5.76 | 0.92 | 0.47 | 0.02 | 0.10 |
| Plataxscalaris          | 10.62| 6.41 | 1.08 | 0.65 | 0.02 | ND   |
| Merlucciusmerluccius    | 7.75 | 4.22 | 0.66 | 0.52 | 0.02 | ND   |
| Weekly [CR<sub>wm</sub> (kg/w)] |
| Scomberscombrus         | 54.27| 46.18| 9.88 | 6.64 | 0.59 | ND   |
| Sardinellalongiceps     | 50.58| 40.33| 6.44 | 3.32 | 0.17 | 0.71 |
| Plataxscalaris          | 74.38| 44.88| 7.59 | 4.52 | 0.16 | ND   |
| Merlucciusmerluccius    | 54.28| 29.51| 4.62 | 3.66 | 0.11 | ND   |
| Monthly [CR<sub>mm</sub> (mm<sup>-1</sup>)] |
| Scomberscombrus         | 52978.36| 45080.95| 9645.23| 6480.39| 576.03| ND   |
| Sardinellalongiceps     | 49374.39| 39374.51| 6284.01| 3240.19| 162.01| 691.24|
| Plataxscalaris          | 72580.36| 43811.08| 7406.16| 4412.18| 152.48| ND   |
| Merlucciusmerluccius    | 52978.36| 28801.73| 4508.09| 3575.39| 108.01| ND   |

The allowable consumption for one day would not essentially cause either chronic or acute health effects. However, consumption rate over a long period of time may be harmful to human health [34]. In Table 4, the highest allowable consumption rate for a day was found in Fe (10.62 kg/d) in Platax scalaris. The allowable consumption rates in Zn, Cu, Ni and Pb were 6.59, 1.41, 0.95 and 0.08 kg/d respectively in Scomber scombrus. In Sardinella longiceps, the allowable consumption rate for Cd was 0.10 kg/d. Consumer may ingest heavy metals which has the ability to cause adverse health effects through the consumption of the fish samples. The highest allowable consumption rate for a week was found in Fe (74.38 kg/w) in Platax scalaris. The highest allowable consumption rate for a week in Zn, Cu, Ni and Pb were 46.18, 9.88, 6.64 and 0.59 kg/w respectively in Scomber scombrus. In Sardinella longiceps, the allowable consumption rate for Cd was 0.71 kg/w (Table 4). The highest allowable monthly consumption rate was found in Fe (72580.36 mm<sup>-1</sup>) in Platax scalaris. The highest allowable consumption
rate for month in Zn, Cu, Ni and Pb were 45080.95, 9645.23, 6480.39 and 576.03 mm$^{-1}$ respectively in *Scomber scombrus*. The highest allowable consumption rate in Cd was 691.24 mm$^{-1}$ in *Sardinella longiceps* (Table 4).

### Table 5: Target Hazard Quotient (THQ) intake and Total Hazard Index (HI) of Fe, Zn, Cu, Ni, Pb and Cd in the muscle of the fish samples

| Fish Specie            | Total Hazard Quotient (THQ) | Total Hazard Index HI |
|------------------------|----------------------------|-----------------------|
|                        | Fe  | Zn  | Cu  | Ni  | Pb  | Cd  |                |
| *Scomber scombrus*     | 0.135 | 0.158 | 0.739 | 1.100 | 12.375 | ND | 14.507         |
| *Sardinella longiceps* | 0.144 | 0.181 | 1.134 | 2.200 | 44.000 | 10.312 | 57.973         |
| *Platax scalaris*      | 0.098 | 0.162 | 0.962 | 1.615 | 46.750 | ND | 49.589         |
| *Merluccius merluccius*| 0.135 | 0.247 | 1.581 | 1.993 | 31.625 | ND | 35.582         |

Table 5 depicts the Target Hazard Quotient intake and total hazard index of the heavy metals in the muscle of the various fish samples. Khan and co-workers [32] reported that THQ values greater than 1 indicate potential health risk from the metals and exposure through consumption of fish may likely lead to deleterious effects in human. The higher the THQ value, the higher the probability of hazard risk to the human body. The health risks related to THQ values of Ni and Pb were highest among all the heavy metals studied. THQ values for Cu in *Sardinella longiceps* and *Merluccius merluccius* were higher than 1. In *Sardinella longiceps*, THQ value for Cd was 10.312 indicating risk to health. The consumers of these fishes are at a risk due to elevated Ni and Pb THQ values with potential for adverse health effects. The highest THQ value was found in Pb (46.750) for *Platax scalaris*. THQ values in Fe and Zn were all below 1 in all the fish samples and those metals do not pose health risk to consumers of the fish [33].

The total hazard index (HI) was derived from the summation of THQs of all the heavy metals studied. The highest values of HI were found in *Sardinella Longiceps* while the lowest values were obtained in *Scomberscombrus*. Hazard quotient based risk assessment method does not promote a quantitative estimate for the probability of an exposed population experiencing a reverse health effect but rather provides an indication of the risk level due to exposure to pollutants.

### Conclusion

The levels of Fe, Zn and Cu determined in all the tissues of the fish samples were lower than the maximum levels and guidelines [23, 25] except for Ni and Pb. It was only in *Scomberscombrus* and *Sardinellalongiceps* that Cd was detected. The concentrations of all heavy metals in the fish samples were highest in the gills. Fe was the metal with the highest concentration in all the tissues. The muscle which is the edible part of the fish recorded low levels for some of these heavy metals compared to other tissues. The muscle of *Sardinellalongiceps* had elevated level of Cd compared to maximum levels and guidelines. The THQ values for Fe and Zn in the muscles of the fish indicated no risk from the fish consumption while that for Ni and Pb indicated high risk from the fish consumption for most of the health risks related to the heavy metals in all the fish samples. There could also be health risks from Cu in *Merlucciusmerluccius* as well as Cu and Cd in *Sardinellalongiceps*. Consuming fish species with metals of high THQ on a large scale for a long period could be unsafe.

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