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**CHEMICAL ANALYSIS OF BIOACCUMULATED HEAVY METALS IN FORMULATED MEDICINAL PRODUCED FROM NATURAL PRODUCTS FOR INDUSTRIALIZATION.**

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**ABSTRACT**

Bioaccumulation is essentially the build-up of contaminants such as heavy metals or pesticides in living organisms. That is, bioaccumulated heavy metals are those metals essentially build up in living organisms which are very pronounced among the aquatic organisms, because they absorbed contaminants from water around them faster than their bodies are able to excrete, which was the basic justification for this investigation on some selected fruits, domestically consumed. Whereas *heavy metals are any metallic elements that have a relatively high density and toxic or poisonous even at low concentration, which is an environmental issue that has become a global problem. The chemical analysis of bioaccumulated heavy metals in formulated natural medicinal from ginger, onion, garlic, alligator pepper and bitter kola, was evaluated using instrumental techniques. The formulated samples contain (5g ginger, 4g onion, 3g garlic,2g bitter kola and 1g alligator pepper) each, but subjected to different drying techniques to ascertain the most probable method. Where samples A = sun-dried, B= air-dried, and C= oven-dried. The results shown that, lead ranges between (A=0.16 - B=0.5ppm), while it ranges between (A=0.07 - B=0.13ppm) for cadmium. But differs with copper (C=0.05 to B=0.09ppm), whereas for zinc (A&C=0.10 - B=0.92ppm), and nickel (A&C=0.06 – B=0.07ppm) the same order was observed. Therefore, the bioaccumulated heavy metals observed was on the safer side for human consumption when compare to the WHO (0.001-3mg/L) human consumption thresholds, with sun-dried techniques preferably.*

*Keywords: Bioaccumulated, Density, Environmental, Heavy Metals, Poisonous, Toxic.*

**1.0 INTRODUCTION**

 Heavy metal pollution is an environmental issue that has become a global problem. The production and emission of heavy metals has increased along with increased industrial development. This has led to increasing concern over food safety due to soil polluted with anthropogenic heavy metals released from industry or agriculture, such as smelting industries, residues from metalliferous mines, pesticides, fertilizers, and municipal composts (Guala et al.,2010). Heavy metals are chemical elements that have a relatively high density, strong toxic effects and pose an environmental threat. Heavy metals are of considerable environmental concern due to their toxicity, many sources, non-biodegradable properties, and accumulative behaviours (Nica et al.,2012). The presence of heavy metals in foods poses serious health hazards, depending on their relative levels. The ability of plants to accumulate metals and possibly other contaminants varies with both the nature of the plant species and the nature of the metal contaminant. Cereals, in this case of Zea mays L (maize), are known to be good accumulators of contaminants (Orisakwe et al.,2012).

Agricultural soils in many parts of the world are slightly to moderately contaminated by heavy metal toxicity, such as cadmium (Cd), copper (Cu), zinc (Zn), nickel (Ni), cobalt (Co), chromium (Cr), lead (Pb), and arsenic (As). This could be due to long term use of phosphate fertilizers, sewage sludge application, dust horn smelters, industrial waste and poor watering practices in agricultural lands (Wang et al.,2017). The primary response of plants is the generation of reactive oxygen species upon exposure to high levels of heavy metals. Various metals either generate reactive oxygen species directly through Haber-Weiss reactions or overproduce reactive oxygen species and the occurrence of oxidative stress in plants could be the indirect consequence of heavy metal toxicity (Miedico et al.,2016). The indirect mechanisms include their interactions with the antioxidant system, disrupting the electron transport chain or disturbing the metabolism of essential elements (Afrin et al.,2015). One of the most deleterious effects induced by heavy metals exposure in plants is lipid peroxidation, which can directly cause biomembrane deterioration. Malondialdehyde, one of the decomposition products of polyunsaturated fatty acids of a membrane, is regarded as a reliable indicator of oxidative stress. Such toxic elements are considered soil pollutants due to their widespread occurrence, and their acute and chronic toxic effect on plants grown. Several studies on the effects of bioaccumulation in plants through uptake of heavy metals from soils at high concentrations have been carried out and indicate great health risks, taking into consideration food chain implications. Utilization of food crops contaminated with heavy metals is a major food chain route for human exposure, especially those under continuous cultivation. The cultivation of such plants in contaminated soil represents a potential risk since the vegetal tissues can accumulate heavy metals (Eqani., 2016). Heavy metals become toxic when they are not metabolized by the body and accumulate in soft tissues Chronic ingestion of toxic metals has undesirable impacts on humans and the associated harmful impacts become perceptible only after several years of exposure (Mahmood et al.,2010).

 Food crops are one of the important parts of our diet, and they may contain a number of essential and toxic metals (Wodaje et al., 2014). Depending on growing media characteristics. Vegetables are the major source of human exposure to heavy metal and contribute about 90 % of the total metal intake, while the rest 10 % intake occurs through dermal contacts and inhalation of contaminated dust Food safety is a burning issue regarding human health in the recent decades because of the high demand for food. This scenario leads to stimulate researchers and scientists to work on health risk associated with consumption of heavy metals, pesticides, and toxin-contaminated food (Bhattacharyya et al.,2016).

 Essential and nonessential elements are regularly added to our food chain through excessive use of agrochemicals, municipal wastewater, industrial effluents, and raw sewage for irrigation (Elzwayie et al.,2017). The Agency for Toxic Substances and Diseases Registry (ATSDR 2011) has classified heavy metals and metalloids such as As, Pb, and Cd found in the environment as 1, 2, and 7 on the basis of toxicity. Some elements like Cu, Cr, Fe, Mn, and Zn are essential for animals and human beings because they play an important role in different metabolic functions, enzymatic activities, sites for receptors, hormonal function, and protein transport at specific concentrations (Hubbs et al., 2005). Other elements like As, Cd, and Pb are nonessential and have no beneficial role in plants, animals, and humans and have no nutritional function, as they are highly toxic. It is necessary to characterize the sources and contents of heavy metals in soil in order to establish quality standards and to determine the threats to human health and food safety (Hubbs et al., 2005). The associated health hazards of toxic metal depend on concentrations of these metals in specific media and exposure time. Long time and chronic exposure can cause health hazards even at low concentrations of toxic metal (Khan et al., 2011). The main objective of this study was to determine the bioaccumulated heavy metals in some selected fruits samples.

1. **MATERIALS AND METHODOLOGY**

**2.1 MATERIALS**

**2.1.1 Apparatus**

The following Apparatus were used; Crucibles, Beakers, Oven, Muffle furnace, Heating mantle, Volumetric flasks, Pair of forceps Analytical balance, Desiccators

**2.1.2 Reagents**

**The reagent used were**; Distilled water, HNO3, H2SO4, HCl

**2.1.3 Sample Collection**

The samples were obtained by market survey from Oje market in Ede, Osun State.

**2.1.4 Sample Treatment**

Each of the ingredients were sliced into smaller sizes. Each sliced sample was divided into three equal parts; The first part was sun-dried, the second part was Oven- dried, while the third part was air-dried. The dried samples were then grounded into smaller particles with the aid of a mortar and pestle and sieved to obtain homogeneous fine particles which was subjected to further analyses.

**2.1.5 Sample Formulation**

The powdered ingredients were measured as follows; 5g of ginger, 4g of onion, 3g of garlic, 2g of bitter kola and 1g of alligator pepper in three folds, the first one was sun dried(A), the second one oven dried(B), while the third one was air dried(C). Each of this samples were thoroughly mixed for homogeneous reaction.

**2.2 METHODOLOGY**

**2.2.1 Bioaccumulated Heavy Metal Content Determination**

**Procedure**: **5m**l of **10%** per chloric acid (**HCI03**), Trioxonitrate (V) (**HN03**) and **15ml** of Hydrofluoric acid **(HF**) were all added to the ash sample after weighed. The solution was stirred properly with glass rod/police rod and allowed to evaporate as it was heated at a temperature of 600C for 12hrs. 4ml of Hydrochloric acid (HCl) was then added to the cooled solution and warmed to dissolve the samples or any salt formed. Then filtered with glass wool (care was taken with glass wool, hand glove as well as test tube holder was used because it is carcinogenic). On cooling, the solution was then diluted to 50ml with deionized water. The solution was then introduced into AAS for the reading. The final results were obtained.

**3.0 RESULTS AND DISCUSSION**

**3.1 Results**

**3.1.1 Bioaccumulated Heavy Metal Content**

The result presented in Table 1, showed the bioaccumulated heavy metal composition with respected to dried techniques.

**Table 1: Bioaccumulated heavy metal content in samples**

|  |
| --- |
| **Samples Pb Cd Cu Zn Ni** **ppm ppm ppm ppm ppm** |
| A 0.16 0.07 0.07 0.10 0.06 |
| B 0.50 0.13 0.09 0.92 0.07 |
| C 0.35 0.08 0.05 0.10 0.06  |

**Key: A=Sun-dried sample, B=Oven-dried sample, C=Air-dried sample**

**Key: A = Sun-dried sample B = Oven-dried sample C = Air-dried sample**

**Figure 1:** Levels of bioaccumulated heavy metals in the samples

**3.2 Discussion**

The levels of Pb, Cd, Cu, Zn, and Ni in the samples and their descriptive statistical parameters were shown in Table 1. Lead ranged from 0.16 – 0.50 ppm, where the least content was observed for sun-dried (A=0.16ppm) and highest with oven-dried (B=0.50ppm) sample. Although, the lowest limit of the above range was sparingly higher than the value set for foods or food products by the most regulatory bodies. Because, a maximum permissible limit of 1mg/kg was prescribed by the FAO/WHO (2002) for food additives, while the threshold values (mg/kg) for cereal grains, leafy vegetables and fruiting vegetables are 0.1, 0.2 and 0.05 respectively, (FAO/WHO, 2012). Differently, a value of 0.03 mg/kg was prescribed by (WHO, 1999) for medicinal plants in their final dosage form. This slight different may not be unconnected with the agricultural practice, the plant and the soil properties (such as cancerous nature of soil) that mighty affected the uptake of the metal which was in accordance with the work of Kitata et al (2012).

 While the Cd (ppm) ranged from 0.07- 0.13, which fall within the required limits. With the highest value observed for oven-dried and least with sun-dried.

Copper was observed to have the range (0.37-0.90ppm), which was also slightly above the threshold for woman consumption, as recommended by WHO. This disparity hold for the same reason advanced for lead, which was in accordance with the work of Diriigen, (2010).

 However, there has been dramatic increase in the use of Zinc based fertilizers and addition of sludge to the soil under plants, so it is important to monitor the zinc levels in spices. In this study, zinc levels ranges (0.10-0.90ppm). The most interesting part of it was that, both sun and air-dried recorded the lowest value (010ppm) but highest with the oven-dried (0.90ppm).

The same pattern was observed for nickel, which ranges thus (0.06-0.07ppm), where both the sun and air-dried sample recorded same value bearably minima, but highest with oven-dried. That is, the determined values are within tolerable limits, which was in accordance with the work of Kitata (2012). The above observation can be inferred from figure 1 above, which shown clearly at glance the level and distribution of those metals with respect to different dried techniques.

**4.0 CONCLUSION AND RECOMMENDATION**

**4.1 Conclusion**

 The levels of Pb, Cd, Cu, Zn, and Ni in the spectrum of the samples was successfully determined and the health risk associated with intake of these bioaccumulated heavy metals has been indirectly assessed, with the reference to the available standards in accordance with global ethical practices. The accumulation of heavy metals by the active ingrident followed the pattern sun-dried, air-dried and oven-dried, in decreasing order. Inferred that, sun-dried is most suitable techniques for bioaccumulated heavy metals investigation in most edible samples.

**4.2 Recommendations**

The following recommendations were made after the study:

* Further study should be carried out on effect and toxicity level of bioaccumulated heavy metals on food.
* Further research should be carried out on the pattern of distribution of these bioaccumulated heavy metals.

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