

# Levels of Heavy Metals in some Antimalarial Herbal Products Marketed in Kano State, Nigeria

Yusuf K. M., \*Abubakar U. S., Sa'adatu A. U., Sagir H.,  
Fatima S. S., Mujahid N. M., Muhammad S., Jamila G. A.

Bioresources Development Centre,  
Kano.

National Biotechnology Development Agency (NABDA),  
Abuja, Nigeria

Email: Umarfarouk2003@yahoo.com

---

---

## Abstract

*The presence of heavy metals in medicinal plants and herbal products has continued to gain global attention because of the toxicological risks posed by higher levels of such metals to human health. Therefore, this study investigates the levels of some heavy metals in some antimalarial herbal products marketed in Kano State, Nigeria. A total number of nine antimalarial herbal products were randomly collected from Kurmi market, which serves as a source of marketed herbal medicines or herbal preparations in Kano State. The levels of heavy metals were analyzed using Micro Plasma Atomic Emission Spectrophotometer (Agilent MPAES, 4100). The result showed the level of metals detected as follows; cobalt:  $3.97 \pm 0.2 - 4.41 \pm 0.08$  mg/kg, lead:  $1.04 \pm 0.02 - 1.25 \pm 0.01$  mg/kg, chromium:  $3.00 \pm 0.06 - 3.66 \pm 0.27$  mg/kg, arsenic:  $0.13 \pm 0.00 - 0.62 \pm 0.01$  mg/kg and mercury:  $0.06 \pm 0.00 - 0.63 \pm 0.02$  mg/kg, while none of the antimalarial herbal products analyzed was found to contain cadmium. This study has shown that the levels of arsenic, lead and mercury detected in the products did not exceed the World Health Organization (WHO) permissible limits. It is recommended that the National Agency for Food and Drug Administration and Control (NAFDAC) and other regulatory agencies should regulate, monitor and evaluate the use, safety, quality, manufacturing, handling, distribution and importation of herbal products in Kano State and Nigeria at large.*

**Keywords:** Herbal products, Heavy metals, Human health, Kurmi market, Permissible limits

## INTRODUCTION

Malaria is the most prevalent among the insect borne diseases. Every year, it kills between one and two million people, with as many as 300-500 million people being infected. It is estimated that nearly half the world population is at risk, with fatal rates being extremely high among young children below 5 years of age (WHO, 2018). Malaria is a classic example of a disease that affects the productivity of individuals, families and the whole society. It is more common in the poorer and less developed countries of the world (Heyneman, 2016; WHO, 2018).

---

\*Author for Correspondence

The use of plants in traditional medicine plays an important role in the healthcare of two-third of the world inhabitants. The knowledge of traditional medicine is scattered among communities, families, tribes and traditional practitioners (Samali *et al.*, 2017).

The World Bank estimates that trade in medicinal plants, botanical drug products and raw materials are growing at an annual rate of 5-15% (Benedum, 1998), however, the main hindrance is lack of established standard quality control measures. Some of these herbal medicines may contain elements of vital importance for human metabolism, diseases prevention and management. On the other hand, they may contain non beneficial elements which could pose health hazards to the body system due to unascertained safety and efficacy (Samali *et al.*, 2017).

Heavy metals are group of elements with high atomic mass and density of atleast  $5\text{gcm}^{-3}$ . The most common heavy metals include lead, chromium, arsenic, mercury, copper, cadmium, zinc, nickel and cobalt. Some heavy metals are essential in a trace amount but at higher concentrations can causes effect on central nervous system, respiratory system, cardiovascular system, vital organs and some have no any vital means (Díaz García and Arceo, 2018; Kumar *et al.*, 2018).

Cobalt (Co) is a key constituent of cobalam in which is known as vitamin B-12, the primary biological reservoir of cobalt as an ultra-trace element (Oladeji and Saeed, 2015) and plays an important role in biochemical reactions (Chivers, 2014). High and frequent cobalt exposures can affect nervous system and cause an axonopathy (Flora, 2014). Exposure to high level of cobalt results in lung and heart diseases and dermatitis. Chronic inhalational intake of cobalt dust can lead to diffuse-inflammatory reactions of the bronchial mucosa and chronic respiratory tract disorders (Cheyns *et al.*, 2014; Banza *et al.*, 2019). In large doses, some Co forms are carcinogenic. Some plant species called metallophytes have adapted to natural and contaminated cobalt-rich soils (Faucon *et al.*, 2007).

Cadmium (Cd) may accumulate in the kidney on chronic exposure due to its long biological half-life, and may lead to kidney damage and hypertension (Maduabuchi *et al.*, 2006; Akan *et al.*, 2010), while high level exposure of lead may result in acute or chronic toxic effects on the nervous system, also associated with impairment of brain functions in children and reproductive system in adults (Horowitz, *et al.*, 2002; Okewole and Omin, 2013).

On the other hand, mercury (Hg) causes abnormalities in foetus during development, as well as reproductive, immune and respiratory toxicity (Adepoju-Bello *et al.*, 2002; Tchounwou *et al.*, 2003). Also, prolonged exposure to mercury damages the brain and causes timidity, tremors, memory problems, irritability, changes in hearing and vision, lung damage, vomiting, diarrhea, nausea, skin rashes, increased cardiac output and elevated blood pressure. Mercury stimulates the formation of reactive oxygen species, causing DNA damage and carcinogenesis (Flora and Pachauri, 2010).

Arsenic (As) induces apoptosis and genotoxicity in humans by inhibiting DNA repair in renal tissue, which causes chromosomal aberrations, micronuclei formation and epigenetic modifications (Dangleben *et al.*, 2013). Chromium (Cr) is a carcinogenic element and can contaminate many environmental systems due to its widespread use in various industries. It can also cause kidney damage, allergies, asthma and respiratory tract cancer in human, stomach ulcer, sperm damage and disorders of male reproductive system (Goulart *et al.*, 2005).

The use of herbal drugs prepared and dispensed by the traditional healers without formal training in the drug formulation and preparation is still very common, thus, there is the

need to assess the safety of the raw materials and finished herbal products (Abubakar *et al.*, 2019). Therefore, the present study was aimed to determine the levels of heavy metals in some marketed antimalarial herbal products sold in Kano State, Nigeria.

## MATERIALS AND METHODS

### Study Area

The study was conducted in Kano State, popularly referred to as the centre of commerce. It is located on 11° 30' N and 8°30' E coordinates, with a mean height of about 472.45 metre above sea level. It borders Katsina State to the Northwest, Jigawa State to the Northeast, Bauchi State to the Southeast and Kaduna State to the Southwest (Figure 1). Kano State has a total number of forty four (44) Local Government Areas, and according to 2006 census, it is the most populous State in Nigeria with a population of 9,383,682 (Federal States, 2014; Abubakar *et al.*, 2017).



Figure 1: Map of Kano State

### Collection of Samples

A total number of nine marketed antimalarial herbal products were randomly collected from Kurmi Market, which serves as a source of marketed herbal medicines or herbal preparations in Kano and some Northern States of Nigeria.

### Heavy Metal Analysis

Approximately 0.40 g of each sample was weighed and placed into a medium-pressure digestion rotor vessel, followed by 8 ml of HNO<sub>3</sub> and 2ml of H<sub>2</sub>O<sub>2</sub>, the mixture was then digested with an advanced microwave digestion system. The solutions were cooled at room temperature and each was diluted to 10 ml with distilled water. The solutions were then analyzed for the presence of cadmium (Cd), copper (Cu), nickel (Ni), arsenic (As), mercury (Hg), cobalt (Co), lead (Pb), manganese (Mn) and chromium (Cr) using Micro Plasma Atomic Emission Spectrophotometer (Agilent MPAES, 4100) available at the Centre for Dryland Agriculture, Bayero University, Kano, Nigeria.

### Statistical Analysis

The levels of heavy metals were determined in triplicates, and data was expressed as mean ± standard deviation (SD).

## RESULTS

The result of heavy metal analysis of marketed antimalarial herbal products was presented in Table 1; which showed that cobalt, lead, chromium, arsenic and mercury were detected in all the samples, while cadmium was not detected in all the samples analyzed. Cobalt showed the highest concentration, ranging from 3.97-4.41 mg/kg, while mercury had the lowest concentration ranging from of 0.06-0.63mg/kg.

**Table 1: Level of Heavy Metals in some Marketed Antimalarial Herbal Products (mg/kg)**

Samples	Cd	Co	Pb	Cr	As	Hg
1	ND	4.20 ± 0.18	1.04 ± 0.02	3.05 ± 0.03	0.62 ± 0.01	0.63 ± 0.02
2	ND	4.41 ± 0.08	1.11 ± 0.06	3.04 ± 0.04	0.85 ± 0.02	0.21 ± 0.01
3	ND	4.20 ± 0.18	1.06 ± 0.03	3.00 ± 0.06	0.79 ± 0.01	0.20 ± 0.01
4	ND	4.21 ± 0.18	1.14 ± 0.07	3.48 ± 0.18	0.81 ± 0.02	0.20 ± 0.01
5	ND	4.31 ± 0.13	1.15 ± 0.08	3.31 ± 0.10	0.83 ± 0.02	0.15 ± 0.01
6	ND	3.97 ± 0.2	1.08 ± 0.04	3.36 ± 0.12	0.80 ± 0.02	0.06 ± 0.00
7	ND	4.15 ± 0.21	1.25 ± 0.01	3.66 ± 0.27	0.13 ± 0.00	0.10 ± 0.00

Key: ND= Not Detected

## DISCUSSION

The levels of lead detected ranges from 1.04±0.02-1.25±0.01mg/kg, but these values didn't exceed the World Health Organization standard of 10mg/kg (WHO, 2007). Also, the levels of lead reported in this study were lower than those reported by Samali *et al* (2017) and Odoh and Ajiboye (2019), but higher than the values reported in some antimalarial herbal decoctions collected from Lagos State, Nigeria; this could be attributed to the differences in the chemical constituents and geographical areas where the herbal products were collected (Alaribe *et al.*, 2018). Lead was reported to bioaccumalate in human tissues causing spontaneous abortion, low birth weight and impaired neuro-development, so patients who use herbal products with higher lead content over a long period of time might be at risk of chronic lead toxicity and should be monitored for any sign of lead poisoning (Okewole and Omin, 2013).

Cadmium was not detected in all the samples tested. The absence of cadmium in this study didn't agree with the work of Samali *et al* (2017), who reported the presence of this metal in some antimalarial herbal preparations collected from the same study area. This could be due to the differences in the chemical constituents and soil compositions of the areas where the raw materials and/or the finished products were collected. Accumulation of cadmium may lead to liver and kidney damage. Also, chronic exposure to cadmium may lead to renal injury, obstructive pulmonary disease, alteration of reproductive hormones and cardiovascular diseases (Akinloye *et al.*, 2006).

Mercury was detected in all the antimalarial herbal products analyzed; however, the values didn't exceed the World Health Organization permissible limit of 1mg/kg in medicinal plants and herbal products (WHO, 2007). On the other hand, the values obtained in this study were lower than the values (1.612-9.147mg/kg) reported in some antimalarial herbal decoctions collected from Lagos State, Nigeria; this may be due to the differences in the chemical constituents and geographical areas where those herbal products were collected (Odoh and Ajiboye, 2019). Prolong exposure to mercury damages the brain and causes timidity, tremors, memory problems, irritability and changes in hearing and vision. It also causes vomiting, diarrhea, nausea and skin rashes (Tchounwou *et al.*, 2003).

Arsenic was detected in all the samples; however, the permissible limit of As is 10mg/kg (WHO, 2007). The presence of Arsenic in herbal products might have been through natural geological sources leaching into aquifers, anthropogenic inputs such as the use of arsenical insecticides, pesticides, fungicides, combustion of fossils fuels, contaminated water and industrial activities (Dangleben *et al.*, 2013).

Chromium was detected in all the samples analyzed, the values range from 3.00± 0.06-3.66± 0.27 mg/kg. The contamination of these products with chromium could be from the tannery industries in Kano State. Many studies indicates that this heavy metal is released to the environment through industries such as tannery, stainless steel production and welding (Oke *et al.*, 2006). However, the levels of chromium detected in this study were lower than those reported by Umar *et al* (2016) who conducted a similar work in Kano State. Although, the permissible limit of chromium is not yet standardized by the WHO and other regulatory bodies because it is considered as micronutrient, however the permissible limit of chromium by Canadian Health Authority for raw medicinal plant is 2mg/kg and 0.02 mg/kg in finished products. High intake of chromium can causes skin cancer, skin rashes, nose irritation, kidney and liver damages (Goulart *et al.*, 2005).

Cobalt had the highest concentration among the heavy metals analyzed. There is no permissible limit set by the WHO and other regulatory agencies for cobalt content in medicinal plants and herbal product. The values of cobalt detected in this work were higher than those reported by Samali *et al* (2017), but lower than those reported by Moses *et al* (2012) who conducted a similar work in Southwestern Kenya. This could be due to the differences in the phytochemical constituents and geographical areas of the herbal products investigated. Cobalt play important role in the formation of cyanocobalmin-vitamin B-12, blood cells production, repair myelin, formation of haemoglobin, regulate and stimulate the production of some co-enzymes (Chivers, 2014).

## **CONCLUSION**

This study has shown that the levels of arsenic, lead and mercury detected didn't exceed the World Health Organization permissible limits for these metals in herbal products and medicinal plants. It is recommended that the National Agency for Food and Drug

Administration and Control (NAFDAC) and other regulatory agencies should regulate, monitor and evaluate the use, safety, quality, manufacturing, handling, distribution and importation of herbal products in Kano State and Nigeria at large.

#### ACKNOWLEDGEMENT

The authors are grateful to Bioresources Development Centre, Kano for funding this project. We also acknowledge the technical assistance of the Technologists in the Central Laboratory, Centre for Dryland Agriculture, Bayero University Kano, Nigeria.

#### REFERENCES

- Abubakar, U.S., Yusuf, K. M., Abdu, G. T., Saidu, S. R. Jamila, G. A. et al. (2017). Ethnopharmacological Survey of Medicinal Plants Used for the Management of Pediatric Ailments in Kano State, Nigeria. *Research Journal of Pharmacognosy*, 4(3): 29-39
- Abubakar, U. S., Abdullahi M. S., Jamila G. A., Mujahid M. N., Sagir, H. et al. (2019). Heavy Metals Evaluation and Acute Toxicity Study of *Artemisia annua* L. (Asteraceae). *Specialty Journal of Biological Sciences*, 5(2): 7-11.
- Adepoju-Bello, A. A., Issa, O. A., Oguntibeju, O. O., Ayoola, G. A. and Adejumo, O. O. (2012). Analysis of some Selected Toxic Metals in Registered Herbal Products Manufactured in Nigeria. *African Journal Biotechnology*, 11(26):6918-6922.
- Akan, J. C., Abdulrahman, F. I., Sodipo, O. A., Ochanya, A. E. and Askira, Y. K. (2010). Heavy Metals in Sediments from River Ngada, Maiduguri Metropolis, Borno State, Nigeria. *Journal of Environmental Chemistry Ecotoxicology*, 2(9):131-140.
- Akinloye, O., Arowojulu, A. O., Shittu, O. B., and Anetor, J. I. (2006), Cadmium Toxicity: a Possible Cause of Male Infertility in Nigeria. *Reproductive Biology*, 6:7-30.
- Alaribe, C. S., Oladipupo, A. R., Musah, A. A. and Coker, H. A. B. (2018). Investigation of Selected Metallic Impurities in Antimalarial Herbal Decoctions Agbo Iba Collected from Four Locations in Lagos State, Nigeria. *UNILAG Journal of Medicine, Science and Technology*, 6(1): 45-56.
- Banza, C. L. N., Nawrot, T. S., Haufroid, V., Decrée, S., De Putter, T. et al. (2009). High Human Exposure to Cobalt and other Metals in Katanga, a Mining Area of the Democratic Republic of Congo. *Environmental Research*, 109, 745-752.
- Benedum, J. (1998). In *Phytopharmaka IV. Forschung und Klinische Anwendung*, D. Loew, N. E. Rietbrock (Eds.).
- Cheyne, K., Banza Lubaba Nkulu, C., Ngombe, L. K., Asosa, J. N., Haufroid, V. et al (2014). Pathways of Human Exposure to Cobalt in Katanga, a Mining Area of the D.R. Congo. *The Science of the Total Environment*, 490: 313-321.
- Chivers, P. T. (2014). Cobalt and Nickel. In Maret, W., and Wedd, A. (eds.). *Binding, Transport and Storage of Metal Ions in Biological Cells*. Cambridge, UK: The Royal Society of Chemistry, pp. 381-428.
- Dangleben, N. L., Skibola, C. F. and Smith, M. T. (2013) Arsenic Immunotoxicity: a Review. *Environmental Health*, 12: 73-86
- Díaz García, J. D. and Arceo, E. (2018). Renal Damage Associated with Heavy Metals: Revision Work. *Nephrology*, 1: 43-53.
- Faucon, M. P., Shutcha, M. N., and Meerts, P. (2007). Revisiting Copper and Cobalt Concentrations in Supposed Hyperaccumulators from SC Africa: Influence of Washing and Metal Concentrations in Soil. *Plant and Soil*, 301: 29-36.
- Federal States and Major Cities Statistics and Maps on City Population. (2014). Retrieved from [www.citypopulation.de/Nigeria-Cities.html](http://www.citypopulation.de/Nigeria-Cities.html)

- Flora, S. J. S., (2014). *Metals*. In Aronson, J. K., (ed) Side Effects of Drugs Annual. Amsterdam, pp. 297-322.
- Flora, S. J. and Pachauri, V. (2010). Chelation in Metal Intoxication. *International Journal of Environmental Research and Public Health*, 7(7):2745-88.
- Goulart, M., Batoreu, M. C., Rodrigues, A. S., Laires, A. and Rueff, J. (2005). Lipoperoxidation Products and Thioli antioxidants in Chromium Exposed Workers. *Mutagenesis*, 20(5):311-315.
- Heyneman, D. (2016). *Malaria*. Health and Medical Sciences Program, University of California, Berkeley, California.
- Horowitz, Y., Greenberg, D., Ling, G., Lifshitz, M. and Acrodynia, A. (2002). A Case Report of Two siblings. *Archives of Disease in Childhood*, 86:453-455.
- Kumar, A., Singh, N., Pandey, R., Gupta, V. K. and Sharma, B. (2018). Biochemical and Molecular Target of Heavy Metals and their Actions. In: Rai M., Ingle A., Medici S. (eds) Biomedical Applications of Metals. Springer.
- Maduabuchi, J. M. U., Nzegwu, C. N., Adigba, E. O., Alope, R. U., Ezomike, C. N. et al. (2006). Lead and Cadmium Exposures from Canned and Non-canned Beverages in Nigeria: A public health concern. *The Science of Total Environment*, 366:621-626.
- Moses, A. G. Maobe, E. G., Leonard, G. and Henry, R. (2012). Profile of Heavy Metals in Selected Medicinal Plants Used for the Treatment of Diabetes, Malaria and Pneumonia in Kisii Region, Southwest Kenya. *Global Journal of Pharmacology*, 6 (3): 245-251.
- Oke, I. A., Otun, J. A., Okuofu, C. A. and Olarinoye, N. O (2006). Characteristics of Tanning Industries in Nigeria for Aquatic Animals and Plants. *Research Journal of Agriculture and Biological Sciences*, 2(5): 209-217.
- Okewole, A. I. and Omin, B. E. (2013). Assessment of Heavy Metal Contents of Some Paints Produced in Lagos, Nigeria. *Journal of Science of Technology*, 8:60-66.
- Oladeji, S. O. and Saeed M. D. (2015). Assessment of Cobalt Levels in Wastewater, Soil and Vegetable Samples Grown Along Kubanni Stream Channels in Zaria, Kaduna State, Nigeria. *African Journal of Environmental Science and Technology*, 9(10): 765-772
- Odoh, R. and Ajiboye, O. (2019). Emmanuel Quality Assessment of Some Selected Herbal Medicinal Products Consumed in Wukari, Taraba. *Acta Scientific Microbiology*, 2(9): 28-36.
- Samali, A., Mohammed, M. I., and Ibrahim, M. B. (2017). Analysis of Heavy Metals Concentration in Kano Herbal Preparations for Major Disease Conditions. *ChemSearchJournal*, 8(2): 22-28.
- Tchounwou, P. B, Ayensu, W. K., Ninashvili, N. and Sutton, D. (2003). Environmental Exposure to Mercury and its Toxicopathologic Implications for Public Health. *Environmental Toxicology*, 18: 149-175.
- Umar, A., Mohammed, Y., Garba, S. and Faruruwa, M. D. (2016). Quantitative Determination of Heavy Metals in some Commonly Consumed Herbal Medicines in Kano State, Nigeria. *Journal of Scientific and Engineering Research*, 3(2):39-46
- WHO. (2007). Guidelines for Assessing Quality of Herbal Medicines with Reference to Contaminants and Residues. Geneva, Switzerland.
- WHO. (2018). World Malaria Report. World Health Organization, Geneva, Switzerland.