

## Levels of Potentially Toxic Metals In Selected Herbal Medicines In Lagos, Nigeria

Chionyedua T.Onwordi<sup>1\*</sup>, Ngozi Agbo<sup>1</sup>, Isiaka A.Ogunwande<sup>1</sup>,

<sup>1</sup> Department of Chemistry, Lagos State University, P.O Box 0001, LASU Post Office, Ojo, Lagos, Nigeria

\* Corresponding author: [teresachiedu@yahoo.com](mailto:teresachiedu@yahoo.com); [chionyedua.onwordi@lasu.edu.ng](mailto:chionyedua.onwordi@lasu.edu.ng)

### Abstract

The increasing popularity and widespread use of herbs as alternative medicine has sparked an interest in understanding their safety. Potential toxic metals (PTM) have been identified as a risk to human health through the usage of herbal medicines. This study aimed to assess the level and pH of PTM (Fe, Cu, Pb, Cd, Ni and Zn) in selected herbal medicines sold in Lagos, Nigeria. Eight commonly consumed herbal medicines: E-5000, MT-CAPS, B-CAPS, DD-TEA, YC-BITTER, JHD-POWD, KA-POWD, and H-CAPS were analysed for PTM using Atomic Absorption Spectrophotometer after wet digestion of the medicines. The highest concentration (mg/kg) of Fe ( $257 \pm 0.1$ ), Pb ( $33.8 \pm 0.01$ ) and Zn ( $38.9 \pm 0.01$ ) were found in KA-POWD. The result also showed that JHD-POWD had the highest concentration amount of Ni ( $54.0 \pm 0.01$  mg/kg), while B-CAPS had the highest concentration of Cu ( $4.14 \pm 0.02$  mg/Kg). All the herbal medicines analysed are high in Fe content with range between 6.2 to 257 mg/kg. The pH values ranged between slightly acidic of 5.6 - 5.7 to slightly alkaline of 8.0 - 8.65. Twenty five percent of herbal medicines analysed had concentrations of Pb above the the WHO limits of 10 mg/kg, while all the samples had Cd above the permissible limits of 0.3 mg/kg. KA-POWD had the highest concentrations of Pb, Zn, Fe, and Cd determined. All other PTMs in the other samples analysed are below the recommended permissible limits.

**Keyword:** Capsules; Herbal medicine; Lagos; Potentially toxic metals content; Powdered.

### 1 Introduction

Man exists in an environment which is no longer friendly to him as a result of high anthropogenic pressure, which releases high levels of toxic metals to the environment. These metals affect man and plants. Herbal products are obtained from plant and a growing number of Nigerians are using herbal medicine for preventive and therapeutic purposes. The practice and use of herbal medicine have been prevalent among Nigerians for many years. The World Health Organization (WHO) has estimated that up to 80% of the world's populations rely on plants for their primary health care (Pirzada *et al.* 2009). In most African countries including Nigeria, herbal medicine is recognized as an important component of health care system, especially among rural dwellers that constitute about 70% of the population (Esimone *et al.* 2002). A survey carried out in 1985 by WHO in Nigeria estimated that up to 75% of the population patronizes traditional medicine (Omoseyindemi 2003) as results of the ever increasing cost of orthodox health care services. Coupled with the side effects of certain synthetic drug therapies, has further caused a large proportion of patients in the developing countries to resort to alternative herbal health care which they feel is natural, safer, more accessible, and more economical and takes into consideration the people's socio-cultural values (Carter 2001). The use of herbal products is not regulated in Nigeria and in many low-income countries and is freely available to everyone. The safety of these herbal medicines is poorly understood (Obi *et al.* 2006). Herbal medicines are not tested with scientific rigour required for conventional drugs and most manufacturers of these products do not submit proof of safety and efficacy to regulatory bodies before marketing. This calls for proper monitoring by regulatory agencies (Akintola *et al.* 2008, Eisenbery *et al.* 1993)

Potential toxic metals, such as lead, chromium, nickel, arsenic, cadmium, zinc, iron and others are environmental pollutants particularly in areas with high anthropogenic pressure. Their presence in the atmosphere, soil and water even in traces can cause serious problems to all organisms especially man. Environment, pollution, atmosphere, soil, harvesting and handling are some of the factors which play an important role in contamination of herbal medicine by metal (Lynch & Braithwaite 2005, Schumacher *et al.* 1991). It is of major interest to establish the levels of some metallic elements in some herbal medicine because, at elevated levels these metals are dangerous and toxic (Sojo *et al.* 1997, Sheridan & Meola 1999). Heavy metals bioaccumulation in herbs can be especially highly dangerous to human health and may cause serious health hazards such as renal failure, symptoms of chronic toxicity and liver damage (Alabdulaaly & Khan 2009). Potential toxic metals intake by human population through herbs has been reported in many westernized countries with this problem receiving increasing attention from the public as well as government agencies (Chen *et al.* 2001). However, herbs contain both essential and toxic elements over a wide range of concentration (Schumacher *et al.* 1991).

Unlike the conventional drugs, herbal medicines are not regulated for purity and potency. For this reason, the adverse effects and drug interactions associated with the concentration of Potential toxic metals in herbal remedies are of great danger to human lives (Somers 1983).

Safety issues in herbal medicine are a subject of scientific interest and the balance between toxicity, risk and benefit must always be considered. The quality, efficacy and safety of herbal medicine containing potential toxic metals have to be critically assessed before the product can be put in clinical trials or placed in the market (Wang *et al.* 1996)

Several works have been done on the metal contents of herbal, medicinal, and aromatic plants from other parts of the world (Chizzola & Franz 1996, Abou-Arab *et al.* 1999, Lavilla, Filgueiras *et al.* 1999, Kaneez *et al.* 2000, Vartika *et al.* 2001, Kaneez *et al.* 2001) but little is known on level of metals in herbal drugs in Nigeria. Due to the high consumption of herbal medicine in Nigeria and the effect of potential toxic metals in human system, this study set to analyse heavy metals such as lead, cadmium, iron, nickel, zinc and copper in some selected herbal drugs to assure their quality: Hero Capsule (H-CAPS), Bodigad Capsule (B-CAPS), M & T Capsule (MT-CAPS), Kings Action Power (KA-POWD), Jim Herbal Dental Powder (JHD-POWD), Dara Dara Herbal Tea (DD-TEA), Eroxil 5000 (E-5000) and Yoyo Cleanser Bitter (YC-BITTER)

## 2 Materials and methods

All samples of the studied herbal medicine were purchased from different sources of herbal manufacturers in various stores within Lagos, Nigeria. The names of each herbal medicine were identified by the label attached to the container and information provided by the manufacturers. The herbal drugs were kept in the refrigerator prior to analysis.

Table 1.0 below gives further information on the studied herbal samples.

Table 1.0: Basic information of the herbal medicine.

Herbal medicine	Legend	Manufacturer	NAFDAC REG.NO <sup>a</sup>
EROXIL 5000	E-5000	De-Fayus International and Oko- Oloyun	A7-0263L
YOYO CLEANSER BITTER	YC-BITTER	De-Fayus International and Oko- Oloyun	04-5347L
JIM DENTAL POWDER	JHD-POWD	Yem-Yem International	04-9905L
KING ACTION POWDER	KA-POWD	Yem-Yem International	NA
DARA DARA TEA	DD-TEA	King Natural Therapy	04-9852L
BODIGAD CAPSULE	B-CAPS	Jim herbal Laboratories	04-8062L
M & T CAPSULE	MT-CAPS	Eurobridge Industries Limited	04-3841L
HERO CAPSULE	H-CAPS	Ablat herbal Remedy	04-8210L

NA – not available; <sup>a</sup>National Agency for Food Drugs Administration and Control, Nigeria

### 2.1 Reagents and glassware.

The concentrated acids, HNO<sub>3</sub> and HClO<sub>4</sub> used were of BDH analar grade. Demineralised water was used during the course of the analysis. Stock standard solutions of Pb, Cd, Fe, Ni, Cu, and Zn containing 1000ppm of each metal were used. Calibration standards of each metal were obtained by appropriate dilution of the stock solutions. All glassware were soaked in 10% Nitric acid for 24 hours, rinsed with distilled water then with demineralised water and oven dry at 100°C for 10 minutes before use to avoid any form of heavy metal contamination.

### 2.2 Atomic absorption spectrophotometer.

A VGP model 210 (Bulk scientific, USA) was employed for the determination of the heavy metals in the samples. Buck scientific hollow cathode lamps for Pb, Cu, Ni, Fe, Cd and Zn were used as recitation sources. Under optimum operating conditions, the metals were measured using an air-acetylene flame. The absorbance of each metal in the sample solution was obtained and the calibration curve prepared to obtain the concentration.

### 2.3 pH determination

An 11751 SensorE-5000 pH meter (Garden Grove scientific, USA) was used for pH determination. The pH of Bodigad, M & T, Hero Capsules, Jim Herbal Dental Powder and Dara Dara Tea was determined by dissolving some quantity in demineralised water in the ratio 1:1, while Eroxil 5000 and Yoyo Cleanser Bitter were determined directly without the addition of demineralised water.

### 2.4 Digestion of the samples

Samples digestions were achieved by using the Wet Digestion method. 5 g of the herbal medicines were weighed and dissolved in 30mL of mixed acid (HNO<sub>3</sub>/HClO<sub>4</sub> in the ratio 5:1) for 24 hr. The mixture was then heated on a hot plate until the brown fumes changed to white. It was allowed to cool and 10 mL of distilled water was added.

The mixture was again heated until complete white fumes were generated. After cooling, it was filtered into 50 mL volumetric flask. The final volume was made up to 50 mL exactly with demineralised water, which was used as a test solution for the determination of Pb, Cu, Cd, Fe, Zn and Ni (Chunjie *et al.* 2007). AAS A VGP model 210 (Bulk scientific, USA) was employed for the determination of the potential toxic metals in the digested samples. Reagent blank was also carried out in the same way. Two sub-samples of various herbal medicines were digested using the above method. All measurements were done in duplicate for the sample and standard solution. Recovery test was done on some of the Potential toxic metals analysed.

### 3 RESULTS AND DISCUSSION

Table 2 shows the result of mean concentration of heavy metals in capsulated herbal medicine; Table 3 shows the result of mean concentration of heavy metals in powdered herbal medicine while Tables 4 and 5 show the mean concentrations of the heavy metals in liquid herbal medicine and the pH values obtained from the studied herbal medicine respectively. Table 6 shows the permissible limits of some potential toxic metals in herbal medicines

The Pb concentrations ranged from 1.54  $\mu\text{g/g}$  to 33.8  $\mu\text{g/g}$  in all the herbal medicines (Table 2 and 3). However, no trace of Pb was detected in YC-BITTER, E-5000 and DD-TEA (Table 3 and 4). KA-POWD recorded the highest level of Pb with mean concentration of  $33.8 \pm 0.01 \mu\text{g/g}$  and B-CAPS Powder with the lowest mean level of  $1.54 \pm 0.0 \mu\text{g/g}$ . According to (FAO/WHO 2006), the provisional weekly intake of Pb is between 25 to 50  $\mu\text{g/g}$ , while ADI (Acceptable Daily intake) is  $10 \mu\text{g/g}$ . The analysed herbal medicines accumulated this metal at a level below the permissible weekly intake but KA-POWD and JHD-POWD has there mean values higher than the ADI. 75% of the analysed herbal medicines had the ADI lower than the (FAO/WHO 2006) limits.

The Cd mean concentrations ranged from 0.48 to 3.08  $\mu\text{g/g}$  (Table 2-4). E-5000 and YC-BITTER had the lowest amount of Cd with mean concentration of  $0.48 \pm 0.01 \mu\text{g/g}$  and  $0.52 \pm 0.01 \mu\text{g/g}$  respectively while the highest Cd concentration level was observed in B-CAPS (Table 2) with mean concentration of  $3.08 \pm 0.01 \mu\text{g/g}$ . The levels of Cd were higher in all the herbal medicines analysed compared with the ADI of 0.3  $\mu\text{g/g}$  by (FAO/WHO 2006) limits.

The concentrations of Ni ranged from 0.73 to 54.0  $\mu\text{g/g}$ . The highest amount of Ni was observed in JHD-POWD (Table 3) with a mean concentration of  $54.0 \pm 0.01 \mu\text{g/g}$  and DD-TEA had the lowest level of  $0.73 \pm 0.01 \mu\text{g/g}$ . According to (Edward 2008), Pb, Ni and Cd even at a very low concentration cause a number of disorders in human system. Ni and Cd have clearly been seen to produce cancer of the muscle cell (rhabdomyosarcoma), respiratory tract and kidney (Sybril 1980).

KA-POWD contained the highest amount of Fe and Zn having mean levels of  $257 \pm 0.1 \mu\text{g/g}$  and  $38.9 \pm 0.01 \mu\text{g/g}$  respectively as shown in Table 2. The lowest level of Fe and Zn were found in E-5000 and YC-BITTER with concentration mean of  $5.11 \pm 0.01 \mu\text{g/g}$  and  $6.11 \pm 0.01$  respectively for Fe while Zn contents were  $0.86 \pm 0.01$  and  $0.35 \pm 0.01 \mu\text{g/g}$  respectively (Table 4). B-CAPS possessed the highest mean Cu concentration of  $4.14 \pm 0.01 \mu\text{g/g}$  and DD-TEA Powder had the lowest Cu concentration of  $0.08 \pm 0.01 \mu\text{g/g}$ . The FAO/WHO (2006) on contaminants recorded that the maximum daily intake for Zn and Cu is  $50 \mu\text{g/g}$  and  $24.2 \mu\text{g/g}$  respectively. The result shows that these metals falls within the permissible limits. The results of the present investigation have shown that Cu and Zn concentrations are not a matter of concern from the toxicity point of view for the analysed samples. However, both metals are essential for humans, but can be dangerous beyond the permissible limits. The herbal medicines studied are rich in Fe, Zn and Cu which are needed for body metabolism (Hooker 1982). Fe is a component of the respiratory pigments (haemoglobin and myoglobin) and enzymes e.g cytochromes, catalases, peroxidases, aldehyde oxidase, and succinic dehydrogenase etc concerned in tissue oxidation. Iron is essential for oxygen and electron transport within the body (Fraga&Oteiza 2002). Copper is an essential component of numerous oxidation-reduction enzyme systems. For example, copper is a component of the enzyme caeruloplasmin (ferroxidase). It is involved with iron metabolism. Copper plays a function in the formation of the pigment melanin and consequently skin pigmentation; for the formation of bone and connective tissue, and for maintaining the integrity of the myelin sheath of nerve fibres (Kanumakala *et al.* 2002). Zinc is an essential component of more than 80 metalloenzymes, serves as a cofactor in many enzyme systems, acts as an active component or cofactor for many important enzyme systems. Zinc plays a vital role in lipid, protein, and carbohydrate metabolism; being particularly active in the synthesis and metabolism of nucleic acids (RNA) and proteins. Zinc contributes positively in wound healing (Hamilton *et al.* 2000).

The high concentration of PTM in the herbal medicine might be as a result of contamination during processing, for example, grinding in metal fabricated machine / other manufacturing utensil, leaching of metals from metallic containers during storage (Saper *et al.* 2004, Edward 2008). Deficiency or excess of Cu, Zn, Cd, Ni and Fe may cause a number of disorder as reported by (Ahmed *et al.* 1994). These elements take part in

neurochemical transmission and serve as constituent of biological molecules as a cofactor for various enzymes and in variety of different metabolic processes (Mayer & Vyklícky 1989). High metal concentration in the herbal medicine might also be due to improper extraction and clean up of the metals from the herbs when grown on contaminated soil from which the herbal medicines were produced. This contamination could be from the soil from which herbs were harvested prior to processing into various forms. Herbs and plants generally take up metals by absorbing them from a contaminated soil, as well as from metals deposits on parts of the herbs exposed to the air from polluted environments (Robin & James 2003, Peter 2005). It is pointed that the cultivation of herbal plants used for the manufacture of herbal medicine, around industrial areas and waste dumps should be minimized and discouraged as much as possible, as these are means of metals contamination of herbal medicines (Blumenthal 1998).

The levels of PTM observed in the herbal medicines studied is similar to the earlier work done by (Akintola *et al.* 2008). PTM are known to bio-accumulate in the tissue of organisms at higher tropic levels as stated by (Edward 2008), therefore continuous administering of the herbal medicines may lead to increase in the levels of these metals in humans.

A cursory look at the concentration of metals in Table 3 shows that the metal content of the DD-TEA Tea bag was higher than the DD-TEA Powder. It was also evident that a higher metal content was found in capsulated medicines samples represented in Table 2 than in samples in Table 3 that shows the metal content in their respective powder. These differences in concentration of the herbal medicine showed that the tea bag of DD-TEA and the capsules of B-CAPS, MT-CAPS and H-CAPS are also sources of heavy metal to the herbal medicine. This also suggests the different level of contamination during handling and processing.

The pH result (Table 5) shows that E-5000 and YC-BITTER are acidic with values of  $3.35 \pm 0.07$  and  $3.45 \pm 0.07$  respectively, while B-CAPS and H-CAPS were slightly acidic having pH value of  $5.6 \pm 0.14$  and  $5.56 \pm 0.07$  respectively. JHD-POWD and KA-POWD were slightly alkaline at pH of  $8.65 \pm 0.07$  and  $8.00 \pm 0.14$  respectively. However, DD-TEA and MT-CAPS had pH values of  $6.65 \pm 0.07$  and  $6.45 \pm 0.07$  respectively. According to (Ahalya *et al.* 2006), the bioaccumulation of metals in herbs increases at high pH resulting in high concentration of metals in herbal medicine.

From the principal component biplot (Fig 1.0), the points to the left (E-5000, YC-BITTER, H-CAPS, DD-TEA, MT-CAPS, B-CAPS) of the origin had lower concentrations of the metals than points on the right (KA-POWD, JHD-POWD) of the origin. KA-POWD has the highest levels of Pb, Zn, Fe, and Cd compared to the other herbal medicine studied while Cu and Ni are higher in B-CAPS and JHD-POWD respectively. The herbal studied are grouped into 4 clusters. Cluster 1 comprises of KA-POWD; cluster 2 comprises of DD-TEA, YC-BITTER, E-5000; cluster 3 comprises of B-CAPS, MT-CAPS, H-CAPS; while cluster 4 is JHD-POWD (Fig 2.0)

#### 4 CONCLUSION

The present study revealed the potential toxic metals profile in selected herbal medicines in Lagos, Nigeria and established the possible sources of PTM. This study is useful to estimate the level of metal contamination in herbal medicines to assure their safety and quality.

Consumers at every point need to be informed that “natural” does not necessarily mean ‘free from risk’ and the adverse effects as a result of the administering of herbal medicine are undeniable reality due to PTMs contamination (Kubelitz 1998). As a means of periodical assessment of these metals, further research work is needed to determine level of metals in some other herbal medicines being consumed locally in Nigeria. In addition, individuals should be more enlightened on the risk of heavy metals accumulation in the human body, hence the need for the National Agency for Food Drug Administration and Control (NAFDAC) to promulgate and enforce strictly the law guiding the use and safety of herbal products in Nigeria.

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Table 2: Result of mean concentrations (mg/kg) of heavy metals in Capsulated Herbal Medicine

Herbal medicine	Cd	Fe	Pb	Ni	Zn	Cu
H-CAPS	2.28 ± 0.01	36.4 ± 0.2	3.93 ± 0.01	4.52 ± 0.01	21.3 ± 0.01	1.18 ± 0.01
MT-CAPS	2.68 ± 0.01	40.7 ± 0.1	3.47 ± 0.01	3.66 ± 0.01	19.0 ± 0.01	3.18 ± 0.01
B-CAPS	3.08 ± 0.01	47.2 ± 0.01	2.22 ± 0.01	3.73 ± 0.01	19.3 ± 0.01	4.14 ± 0.01

Mean concentration ± standard deviation

Table 3: Result of mean concentrations (mg/kg) of heavy metals in Powdered Herbal Medicine

Herbal medicine	Cd	Fe	Pb	Ni	Zn	Cu
H-CAPS POWDER	2.15 ± 0.01	27.6 ± 0.01	2.57 ± 0.01	3.76 ± 0.01	21.1 ± 0.01	0.89 ± 0.01
MT-CAPS POWDER	1.61 ± 0.01	8.01 ± 0.01	2.85 ± 0.01	2.48 ± 0.01	10.4 ± 0.01	2.35 ± 0.01
B-CAPS POWDER	1.88 ± 0.01	18.7 ± 0.01	1.54 ± 0.01	1.76 ± 0.01	17.3 ± 0.01	3.15 ± 0.01
JHD POWDER	2.35 ± 0.01	20.2 ± 0.01	24.1 ± 0.01	54.0 ± 0.01	13.3 ± 0.01	1.04 ± 0.01
DD-TEA POWDER	1.64 ± 0.01	6.19 ± 0.01	ND	0.73 ± 0.01	8.50 ± 0.01	0.08 ± 0.01
DD-TEA + BAG	2.11 ± 0.01	13.1 ± 0.03	4.75 ± 0.01	3.42 ± 0.01	23.0 ± 0.01	2.30 ± 0.01
KA POWDER	2.41 ± 0.01	257 ± 0.1	33.8 ± 0.01	1.10 ± 0.01	38.9 ± 0.01	1.88 ± 0.01

ND- not detected; Mean concentration ± standard deviation

Table 4: Mean Concentrations (mg/kg) of heavy metals in Liquid Herbal Medicine

Herbal medicine	Cd	Fe	Pb	Ni	Zn	Cu
YC-BITTER	0.52 ± 0.01	6.10 ± 0.01	ND	0.51 ± 0.01	0.35 ± 0.01	2.65 ± 0.01
E-5000	0.48 ± 0.01	5.11 ± 0.01	ND	0.29 ± 0.01	0.86 ± 0.01	0.20 ± 0.01

ND- not detected

**Table 5: pH of Herbal Medicines**

Herbal medicine	pH
E-5000	3.55±0.07
YC-BITTER	3.45±0.07
JHD-POWD	8.65±0.07
KA-POWD	8.00±0.14
DD-TEA	6.65±0.07
B-CAPS	5.60±0.14
MT-CAPS	6.45±0.07
H-CAPS	5.65±0.07
E-5000	3.55±0.07

**Table 6.0: Permissible limits of some potential toxic metals in herbal medicines**

Potential Toxic metals	Type of sample	WHO	US Food and Drugs Administration	Health Science Authority Singapore	Pharmacopoeia of China	Canada	Thailand	Malaysia
Cd	Raw herbal material	NS	NS	NS	0.3 mg/Kg	0.3	NS	NS
	Finished herbal products	0.2 mg/Kg	0.3 mg/Kg	0.05 mg/Kg	NS	0.006mg/day	0.3 mg/Kg	NS
Pb	Raw herbal material	NS	NS	NS	5.0 mg/Kg	10.0	NS	NS
	Finished herbal products	10.0 mg/Kg	10.0 mg/Kg	20.0 mg/Kg	NS	0.02mg/day	10.0 mg/Kg	10.0 mg/Kg
Cu	Raw herbal material	NS	NS	NS	20.0 mg/Kg	NS	NS	NS
	Finished herbal products	20.0 mg/Kg	20.0 mg/Kg	150 mg/Kg	NS	NS	NS	NS
Zn	Raw herbal material	NS	NS	NS	NS	NS	NS	NS
	Finished herbal products	50.0 mg/Kg	50.0 mg/Kg	NS	NS	NS	NS	NS

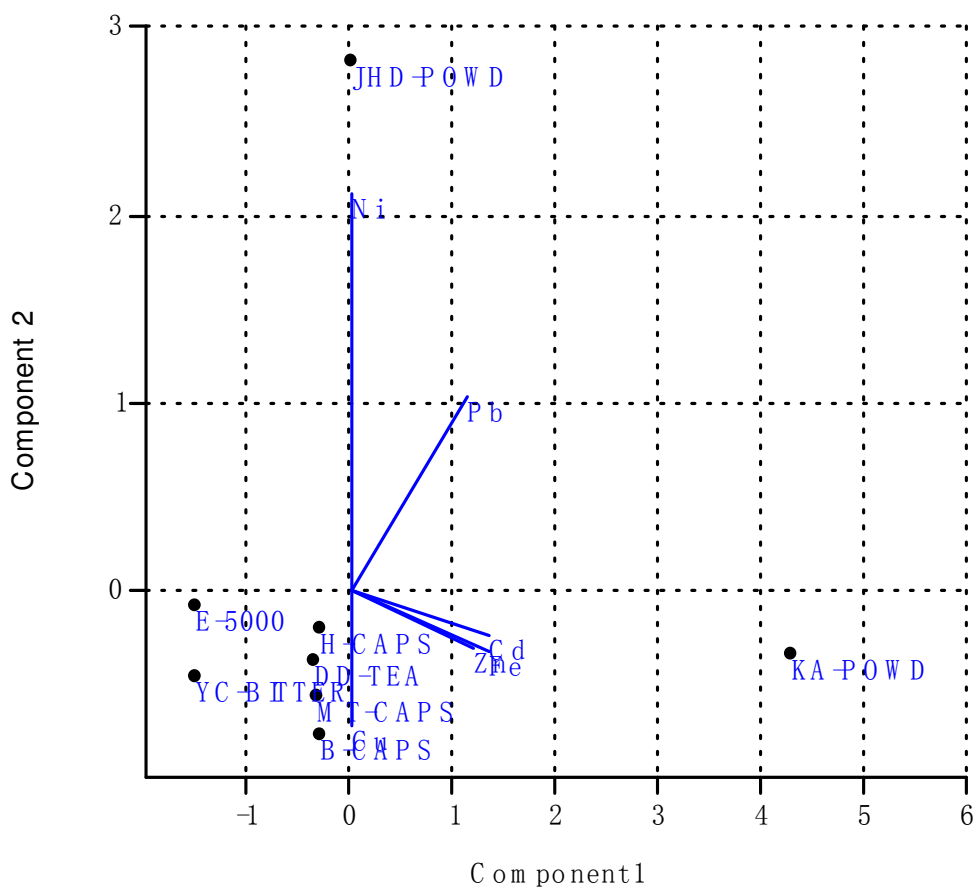


Fig 1.0: Principal component biplot of herbal medicine metal data



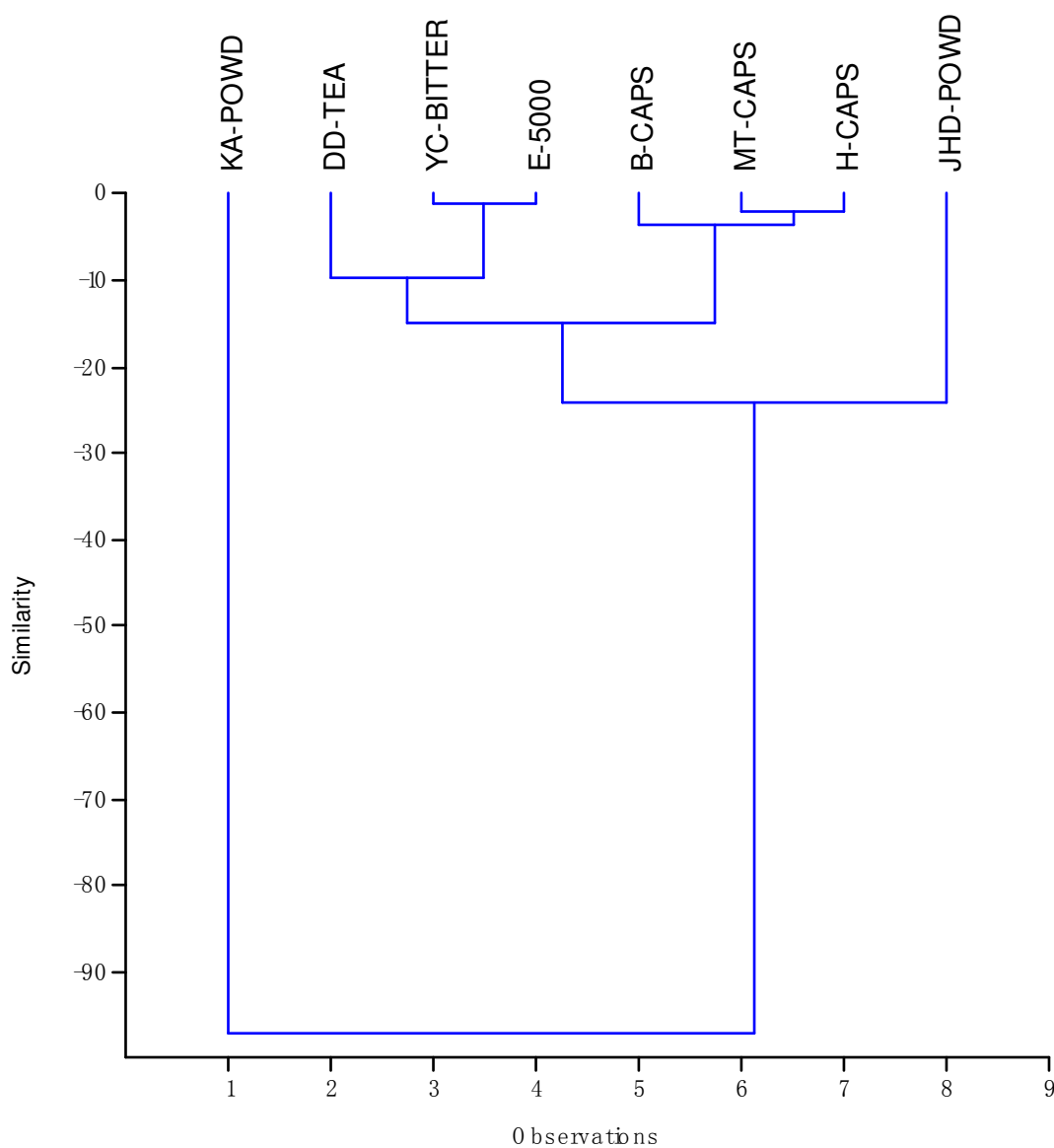


Fig 2.0: Cluster analysis dendrogram for herbal medicine using the six metal concentration.

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