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PIXE characterisation of natural and industrial spices: A nutritional assessment study

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Abstract

Elemental contents of naturally grown and industrially processed spices were determined for the nutritional benefits and assessment. The low temperature oven-dried samples were pulverised and self-supporting pellets made in triplicates. Elemental analysis was carried out using 2.5 MeV proton beams from a Particle Induced X-ray Emission (PIXE) spectrometric setup. Twenty-three elements: Na, Mg, P, S, Cl, K, Ca, Mn, Fe, Co, Cu, Zn, Se, Mo, Sr, Bi, V, Al, As, Pb, Po, Th and U were quantified. A Kapton-90 filter was used to attenuate bremsstrahlung contributions from high counts of low-Z elements, yielding optimal *signal-to-background* ratio required for better quantification of low peaks from high-Z elements. The low-Z elements counts were then corrected for underestimation by evaluating the amount of absorption. The concentrations of Pb, Po and As were below the maximum allowable levels. The essential elements: Mn, Fe, Co, Cu, Zn and Sr had high concentrations in most of the samples while elements like Pb and Cr could add to the heavy metal burden in human body, since they are capable of posing serious health challenges. A robust quality control measures for the cultivation and processing of spices is suggested to ensure the safety of the consumers.

Keywords: PIXE, Spices, Trace elements, Toxic metals, Nutritional assessment.

Introduction

Human body, most especially in the infant stage, requires about 8.6 g of trace elements for maintaining human health, several metabolic activities, normal functioning of the body, growth and development (Prashanth *et al.*, 2015). Trace elements constitute parts of

enzymes, hormones and cells in the body (Bhagwan and Chandravanshi, 2016; Giorgia *et al*., 2019). Globally, the nutritional deficiency of trace elements is of great concern because it is associated with ill health (Salgueiro *et al.*, 2002). It can also lead to body system disorder, abnormal functioning of neural systems, skin diseases, bone defect, disruption of connective tissue and degeneration of skeletal muscle (Yamada, 2013; Vincent, 2015; Messaoudi and Begaa, 2018). These health conditions may arise from poor access to nutritious food coupled with a decreased bio-availability of the essential elements in food. In order to increase the quantities of the essential elements in human body, the emphasis is now on eating more healthy diets that are low in fat and salt but rich in spices (Gaya and Ikechukwu, 2016; Messaoudi and Begaa, 2018; Bertella *et al.,* 2018).

Spices are plant materials (dried leaves, seed, fruit, root, rhizomes, bark and stem) of indigenous or exotic origin used to improve colour, aroma, palatability and acceptability of food. Spices are sometimes added to food for preservative purposes. They also stimulate appetite and increase the flow of gastric juice during digestion (Darkoand Voegborlo, 2014; Bertella *et al.,* 2018). Apart from nutritional benefits, most common spices have been reported to possess outstanding anti-microbial action, anti-diabetic ability and anti-oxidant potential; hence they are used in the preparation of a number of herbal medicines (Darkoand Voegborlo, 2014). Spices are part of the group of sources of essential trace and major elements in human nutrition (Olabanji *et al.,* 2013; Begaa and Messaoudi, 2018). The trace elemental contents of the spices can be affected by the type and condition of soil for cultivation. Environmental pollution through various anthropogenic activities such as automobile and industrial emissions, pesticides and fertilizers application, and improper waste disposal around the farmland can also influence the trace elemental contents of the spices (Nabulo, 2004). For the industrial spices, trace element contamination could occur through the production lines.

The determination of trace elemental concentrations of the species is imperative in the assessment of their associated nutritional benefits. This work demonstrates the application of Particle Induced X-ray Emission (PIXE) spectrometry as a rapid and non-destructive tool in the qualitative and quantitative assessments of the elemental contents of the locally and industrially processed spices. This is with a view to providing information on the nutritional quality and safety of the widely consumed spices. The advantages of PIXE over other nuclear and atomic based analytical methods are the multi-elemental analysis and high sensitivity (Govil, 2001; Abdullahi, 2008; Naidua *et al.,* 1999; Ramadurai *et al.*, 2010; Gowrishankar *et al*., 2010; Olabanji *et al*., 2013, 2016; Sukum *et al*., 2019).

Materials and Methods

Sample collection and preparation

Ten samples (*Murraya koenigii, Zingiber offcinale, Cinnamomum tamala, Allium sativum, Myristica fragrans, Laurus nobilis, Monodora myristica, Microdesmis puberula, Tetrapleura tetraptera,* and *Justicia adhatoda*) were purchased in the raw form at a local market in Ile-Ife, Osun State, Nigeria. Seven fresh samples (*Gongronema latifolium, Ocimum gratissimum, Aframomum melegueta, Syzygium aromaticum, Monodora excelsa, Xylopia aethiopica, Piper guineense*) were harvested from a farmland in Ile-Ife, Nigeria (7.25^0 N and 4.52^0 E). Three industrially processed samples (*Thymus vulgaris, Murraya koenigii,* and *Myristica fragrans*) were also purchased and included in the study. The harvested and purchased natural samples were washed with distilled water and air-dried at ambient temperature (35^0C) in the laboratory, and thereafter oven-dried (60^0C). The natural and industrial samples were then pulverized and self-supporting pellets of each pulverised sample (150 - 300 mg and 13 mm diameter) made in triplicates using Spec-caps (a shallow thin-wall Aluminium cap for reinforcing pellets) by applying 1333 Pa from hydraulic machine.

Analytical technique

The PIXE spectrometric analysis was performed using the 1.7 MV pelletron accelerator for Ion Beam Analysis (IBA) at the Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile-Ife, Nigeria. The facility has radio frequency (RF) charge exchange ion source which provides proton and helium ions. The pelletized samples were bombarded with 2.5 MeV H^+ beam from the PIXE channel of the IBA facility. The measurement was carried out with a beam spot of 4 mm in diameter and a low beam current of 3 - 6 nA. A CANBERRA Si(Li)detector (model ESLX 30-150), beryllium thickness of 25 µm, with full width at half maximum (FWHM) of 150.0eV at 5.9 keV with the associated pulse processing electronics, and a CANBERRA Genie 2000 (3.1) multichannel analyser (MCA) card interfaced to a personal computer were used for the spectra acquisition for about 1000 s. With respect to the beam direction, the sample's normal was located at 0^0 and the Si(Li) detector at 45^0. The computer code GUPIXWIN was used for the analysis of the PIXE X-ray spectra, with the thick target option. The details of the analytical setup and its calibration had been reported (Olise *et al.*, 2010).

The determination of trace amounts of medium or high-Z elements together with low-Z matrix elements at high concentrations require optimal signal-to-background ratio. A

Kapton-90 filter (absorber) was used in attenuating the bremsstrahlung contributions leading to background improvement and subsequent reduction in the counting rate or intensity from the major matrix elements. The reduction of the counting rate made it possible to use a relatively high current in order to increase the counting statistics at lower measuring times. The filter option with other experimental conditions in the spectrum analysis software gave good results for medium- and high-Z analytes with reference to the certified reference material and the investigated samples, respectively. The underestimation recorded at the low-Z region of the spectrum, as a result of having accurate results in the high-Z region, was accounted for by evaluating the amount of absorption from the first principle (Olise *et al.*, 2010).The spectrum of a typical sample (*Laurus nobilis*) with some of the identified peaks is shown in Fig. 1. For example, the high-Z element, uranium was detected in six spice samples with relatively high concentration (81-µg/g) in *Laurus nobilis* samples showing some level of abundance of the element. The concentration of Sr is significant but Rb is not detected. So only Sr could have overlapping peaks with U, hence the M_α lines of U were additionally used to identify it to avoid enhanced peak interference of its L_α lines with the K_α lines of Sr. The concentrations of eleven elements: Mg, Al, P, S, Cl, K, Ca, Mn, Fe, Zn and Sr in the Apple leaves (standard reference material SRM 1515 - NIST) were measured along with the samples for quality control. The recovery for the measured elements in the NIST materials, which is the percentage measured values compared to the certified values, ranged from 97 to 108 %, confirming the reliability of the PIXE spectrometric procedure used in this study.

Data analysis

All the measurements, for each sample, were taken with the same instrument and calibration, resulting in the same constant systematic error, "ε_{syst}". The statistical errors, "ε_{stat}" in these same data fluctuated from measurement to measurement. The statistically estimated errors were obtained as standard deviation (SD), which indicates the variation or dispersion of a set of data values around the mean of repeated measurements. The SD values close to 0 imply closeness of the data to the mean while high SD values indicate spreading of data around the mean. Using the same 95% confidence level for the random and the statistical errors, the total probable error, "ε_{tot}" was then obtained by combining the systematic and statistical errors as shown in equation 1:

$$\varepsilon_{tot} = \left((\varepsilon_{syst})^2 + (\varepsilon_{stat})^2\right)^{1/2} \qquad (1)$$

In this work, the systematic error is small, since one significant digit is sufficient in the expected error.

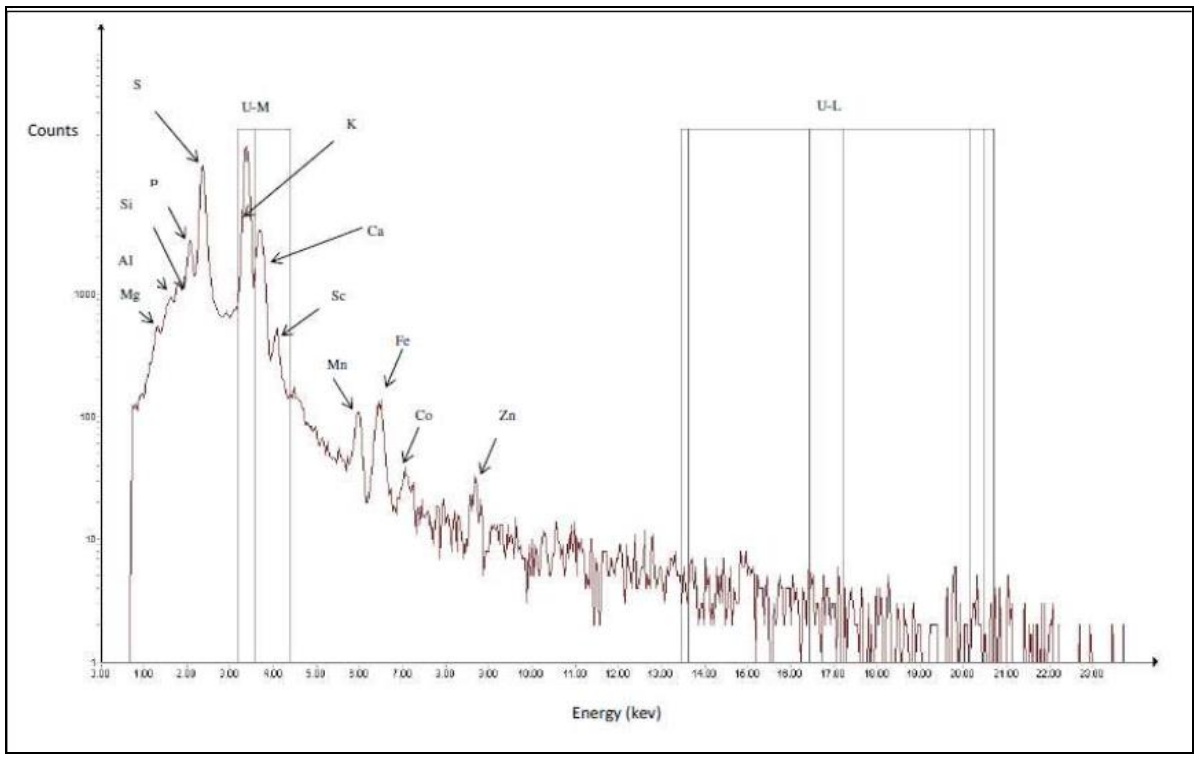

Figure 1: A typical Spectrum (*Laurus nobilis*) from the PIXE analysis of samples.

Results and Discussion

Table 1 presents the average concentrations of minor essential elements (Mn, Fe, Co, Cu, Zn, Se, Zr, Mo, Ni and Sr). The highest concentrations of Mn (455.4±0.8 µg/g) and Fe (318.0±2.6 µg/g) were in *Justicia adhatoda* and *Gongronema latifolium*, respectively. Zinc (26.2±0.2 µg/g) and Copper (7.3±0.2µg/g) had the highest concentrations in *Zingiber offcinale* and *Monodora excels*, respectively. These concentrations were below the Maximum Permissible Level (MPL) of 500, 425, 300 and 73 µg/day for Mn, Fe, Zn and Cu, respectively (FAO-WHO, 2012). This implies a safe consumption of the studied spices for several nutritional benefits and biological functions in human body. The concentrations of Co were below the recommended dietary allowance (RDA) of 8 µg/day stipulated by FAO-WHO, (2012) except in *Gongronema latifolium*. This implies that these samples are not good sources of this element. The average concentration of Fe (269.1±2.6 µµg/g) in *Piper guineense*, one of the widely and daily consumed spices, was higher than Fe (60 µg/g) reported by Asomugha *et al.*, (2016) for the *Piper guineense*. Similarly, Fe, Zn and Mn had 185.4±2.7, 13.1±0.2 and 8.1±0.2 µg/g in *Allium sativum* (locally called onions). These values were higher than the 10, 3 and 6 µg/g, respectively reported for Fe, Zn and Mn in *Allium sativum (*Gaya and Ikechukwu, 2016). However, Mn and Fe had several orders of magnitude higher than the RDA limits of 2.3 and 18 µg/day, respectively (FAO-WHO, 2012). Among the essential elements, Fe plays vital roles in transporting oxygen from the lungs to different parts of the body and in the respiratory pigments, haemoglobin, and insulin and mycoglobin. Copper is important in the treatment of body wounds, prevention of inflammation in arthritis and in major part of several enzymes including tyrosinase, which helps in the formation of melanin pigment (Asomugha*et al*., 2016). Strontium co-functions with Ca in the prevention and treatment of osteoporosis and other bone-related issues (Djama *et al*., 2011). The dietary intake, DI (i.e product of elemental concentration and food intake (0.345 kg/person/day) divided by the average body weight (60 kg)) of Mn, Fe, and Zn were 2.6, 1.8 and 0.15 µg/day for *Justicia adhatoda*, *Gongrone malatifolium* and *Zingiber offcinale*, respectively. When spices are mixed, they could contribute higher levels of the trace elements required in the body for metabolic activities. Incidentally, some of these spices are usually combined, most especially, in ingredients for making a delicacy called pepper soup.

Table 2 shows the average concentrations (µg/g) of major essential elements consisting of Na, Mg, K, Ca, P, S and Cl with Na and S not detected in some of the natural and industrial samples. Potassium and Chlorine had the highest concentrations in *Justicia adhatoda* while P and Mg concentrations were high in *Ocimum gratissimum*. These imply that addition of *Justicia adhatoda* and *Ocimum gratissimum* could enrich K, Cl, P and Mg contents in diet. Although Na is a macro element which regulates blood volume and pressure and osmotic equilibrium (SPQHF, 2010), the low concentrations (0.5 - 0.8 µg/g) of Na in *Allium sativum*, *Myristica fragrans*, *Microdesmis puberula*, *Aframomum melegueta* and *Piper*

guineense should favour their incorporation into the diet of obese and hypertensive patients. This is because of the well-known effect of low sodium intake in the prevention of cardiac failure related conditions (Okonkwo and Ogu, 2014). The Na/K ratios of *Murraya koenigii** and *Murraya koenigi*** are 0.0004 and 0.0009. This indicates that *Murraya koenigii* in the diet would probably reduce high blood pressure since *Murraya koenigii* had been reported to be of great importance in the prevention of high blood pressure (Okonkwo and Ogu, 2014; Messaoudi and Begaa, 2018). Moreover, *Murraya koenigii* and *Justicia adhatoda* are parts of credible and appealing spices for infants, being highly enriched in Ca and K; and may be used therapeutically in the area of medicine and to meet the RDA of these two elements for infants. The average concentrations of K, Ca, Cl, S, P, Mg and Na exceeded 100 µg/day stipulated by FAO-WHO, (2012) in human body. The consumption of spices with high levels of K, Ca, Cl, S, P and Mg will be good for optimum human dietary intake. Chlorine aids the proper digestion of food and prevention of goitre in human body. Potassium is responsible for hormone secretion action and gastrointestinal mobility and regulation of the water balance of the body (Olabanji *et al*., 2006). Calcium had been reported to help in development and maintenance of strong teeth and bone and it supports synthesis of blood cells (Linus and Wingo, 2014).The highest dietary intake (DI) values of K, Ca, and P were 53, 25 and 1.8µg/day, for *Justicia adhatoda*, *Murraya koenigii** and *Ocimum gratissimum*, respectively. All the essential elements are required in equilibrium in the body. However, excessively high concentrations may lead to several health issues. For instance, excess Ca in human body results in calcification, hypercalcemia and creation of kidney stones while high concentration of Cl may cause electrolyte imbalance (hyperchloremia) and damage to body cells.

Table 3 shows the average concentrations of Bi, V, Al, Pb, U, As, Po and Th. Lead was detected only in *Tetrapleura tetraptera*. Its concentration was higher than 0.2 µg/g permissible limit, which shows that caution should be taken when utilising it for food to prevent body Pb burden. Although the elemental toxicity depends on the concentration, ionic and molecular species of the element, the Agency for Toxic Substances and Disease Registry (ATSDR) had listed Pb as one of the priority toxic elements without any biological significance in human body (ATSDR, 2015). Even at this allowable level, a prolonged intake can be hazardous to human beings. Some of known Pb health effects are nervous system disorder, renal and gastro-intestinal tract damage, immune system reduction and low intellectual development in young children and mental retardation in adults (Uhegbu *et al.,* 2011). The possible sources of contamination of Pb could be from soil and other sources like poor handling/industrial production process (Nabulo, 2004). Uranium was detected in *Laurus nobilis, Murraya koenigii**, *Thymus vulgaris*, *Myristica fragrans*, *Monodora excelsa* with average concentrations of 81.0±0.5, 35.7±0.6, 26.5±0.6, 19.3±0.6 and 15.1±0.8 µg/g. The RDA of U is not available and it has no specific function in the human body. Uranium has been reported to be a reproductive toxicant, which is responsible

Table 1: Average concentrations of minor essential elements (µg/g)

Spice samples	*Mn*	*Fe*	*Co*	*Cu*	*Zn*	*Se*	*Mo*	*Sr*
*Murraya koenigii**	*20.8±0.2*	*166.8±2.2*	*ND*	*2.9±0.1*	*23.9±0.2*	*ND*	*ND*	*20.6±0.4*
Zingiber offcinale	*86.6±0.2*	*199.2±2.7*	*2.99±0.1*	*2.5±0.2*	*26.2±0.2*	*ND*	*13.3±0.6*	
Cinnamomum tamala	*8.1±0.2*	*110.4±2.7*	*ND*	*ND*	*20.5±0.2*	*ND*	*ND*	*12.1±0.4*
Allium sativum	*8.1±0.2*	*185.4±2.7*	*ND*	*BDL*	*13.1±0.2*	*3.2±0.2*	*ND*	*23.3±0.4*
Myristica fragrans	*4.3±0.1*	*13.9±2.6*	*ND*	*3.9±0.2*	*3.5±0.2*	*ND*	*ND*	*8.2±0.4*
Laurus nobilis	*17.3±0.1*	*26.3±2.7*	*3.1±0.1*	*ND*	*17.6±0.2*	*ND*	*46.0±0.6*	*ND*
Monodora myristica	*3.7±0.1*	*200.9±2.6*	*ND*	*4.9±0.2*	*8.8±0.1*	*ND*	*ND*	*6.2±0.4*
Microdesmis puberula	*64.9±0.1*	*35.1±2.6*	*ND*	*4.9±0.2*	*7.1±0.1*	*ND*	*ND*	*ND*
Tetrapleura tetraptera	*87.2±0.1*	*138.1±2.6*	*ND*	*7.0±0.2*	*14.2±0.2*	*ND*	*ND*	*8.9±0.4*
Justicia adhatoda	*455.4±0.8*	*188.7±2.6*	*ND*	*2.2±0.3*	*18.7±0.1*	*ND*	*38.3±1.0*	*32.1±0.3*
Gongronema latifolium	*28.1±0.9*	*318.0±2.6*	*9.5±0.1*	*2.8±0.3*	*20.7±0.2*	*2±0.6*	*34.3±0.6*	*60.5±0.3*
Ocimum gratissimum	*29.3±0.2*	*25.51±2.8*	*2.3±0.1*	*2.7±0.2*	*9.6±0.1*	*ND*	*ND*	*12.7±0.4*
Aframomum melegueta	*222±0.8*	*82.4±2.7*	*ND*	*2.9±0.2*	*7.4±0.1*	*5.5±0.8*	*ND*	*18.4±0.4*
Syzygium aromaticum	*1.9±0.2*	*39.5±2.6*	*2.5±0.1*	*5.9±0.2*	*6.1±0.1*	*ND*	*21.0±0.6*	*ND*
Monodora excels	*32.4±0.2*	*105.5±2.7*	*ND*	*7.3±0.2*	*4.4±0.1*	*ND*	*ND*	*16.8±0.6*
Xylopia aethiopica	*4.6±0.1*	*29.7±2.7*	*2.2±0.1*	*2.9±0.2*	*6.7±0.1*	*ND*	*14.8±0.7*	*18.9±0.4*
Piper guineense	*41.8±0.1*	*269.1±2.6*	*ND*	*ND*	*10.4±0.2*	*ND*	*14.8±0.7*	*18.9±0.4*
Thymus vulgaris	*3.5±0.1*	*36.2±2.4*	*ND*	*ND*	*5.3±0.1*	*ND*	*ND*	*ND*
*Murraya koenigii***	*12.9±0.1*	*63.5±2.7*	*ND*	*2.9±0.2*	*6.7±0.1*	*ND*	*ND*	*ND*
Myristica fragrans	*18.9±0.2*	*185.5±2.7*	*ND*	*5.9±0.2*	*4.1±0.2*	*ND*	*20.1±0.6*	*ND*
Detection limit	*0.8*	*1.1. 1.3*	*1.6*	*1.2*	*0.9*	*2.8*	*12.3*	*4.4*

Table 2: Average concentrations of major essential elements (µg/g)

Spice samples	*Na*	*Mg*	*K*	*Ca*	*P*	*S*	*Cl*
*Murraya koenigii**	*1.8±0.4*	*37.9±0.6*	*4780.4±1.2*	*4327.4±3.5*	*262.8±4.5*	*ND*	*1134.8±0.7*
Zingiber offcinale	*ND*	*27.6±0.7*	*4917.8±1.2*	*494.9±3.5*	*148.2±4.5*	*ND*	*350.4±0.7*
Cinnamomum tamala	*ND*	*7.1±0.5*	*1835.5±0.5*	*2444.9±3.5*	*96.9±0.6*	*24.3±1.1*	*84.1±0.7*
Allium sativum	*0.5±0.3*	*3.8±0.6*	*1831.5±5.5*	*2306.9±3.5*	*66.0±4.6*	*59.0±2.1*	*76.1±1.3*
Myristica fragrans	*0.8±0.4*	*11.6±0.7*	*1176.1±1.5*	*230.2±3.6*	*138.4±4.5*	*43.1±2.2*	*10.8±0.7*
Laurus nobilis	*ND*	*3.7±0.6*	*1096.5±1.6*	*162.6±3.6*	*100.6±4.6*	*23.3±2.2*	*3.7±0.7*
Monodora myristica	*ND*	*13.9±0.6*	*12963±.1.5*	*328.7±3.5*	*146.3±4.5*	*41.6±2.1*	*18.0±0.7*
Microdesmis puberula	*0.8±0.4*	*10.2±0.6*	*227.9±1.5*	*943.4±3.5*	*78.0±4.0*	*26.8±1.9*	*410.0±0.77*
Tetrapleura tetraptera	*ND*	*2.4±0.6*	*1802.4±1.5*	*150.4±3.5*	*42.7±4.6*	*10.3±1.9*	*7.0±0.7*
Justicia adhatoda	*ND*	*32.8±0.6*	*9217.3±1.5*	*1925.1±3.5*	*137.3±4.6*	*ND*	*1519.7±0.8*
Gongronema latifolium	*1.3±0.4*	*31.9±0.6*	*3851.2±1.5*	*2110.3±3.5*	*118.7±4.5*	*ND*	*272.5±0.7*
Ocimum gratissimum	*1.5±0.4*	*41.7±0.6*	*5638.6±1.5*	*4199.8±3.5*	*307.9±4.6*	*ND*	*924.6±0.7*
Aframomum melegueta	*0.7±0.4*	*12.7±0.6*	*2360.6±1.5*	*135.9±3.5*	*99.6±4.6*	*14.2±1.9*	*238.7±0.7*
Syzygium aromaticum	*2.4±0.4*	*11.3±0.6*	*560.0±1,5*	*1690.6±3.5*	*87.5±4.6*	*ND*	*427.7±0.7*
Monodora excels	*ND*	*10.0±0.6*	*2348.9±1.5*	*229.1±3.5*	*222.6±4.5*	*49.5±1.9*	*26.2±0.7*
Xylopia aethiopica	*ND*	*8.2±0.6*	*2535.1±1.5*	*892.4±3.5*	*83.9±4.5*	*9.1±1.9*	*327.9±0.7*
Piper guineense	*0.8±0.4*	*6.8±0.6*	*3269.2±1.5*	*308.3±3.6*	*37.2±4.5*	*4.6±1.9*	*905.9±0.7*
Thymus vulgaris	*ND*	*18.9±0.6*	*2650.8±1.5*	*2313±3.5*	*116.4±4.5*	*6,2±1.9*	*166.6±0.7*
*Murraya koenigii***	*25.7±0.7*	*23.3±0.6*	*27761.2±1.5*	*721.6±3.6*	*155.7±4.6*	*ND*	*3479.2±1.7*
Myristica fragrans	*1.3±0.4*	*18.4±0.6*	*1557.5±1.5*	*554.9±3.6*	*148.9±4.5*	*74.22.6±*	*30.4±0.7*
Detection limit	*0.5*	*1.2. 13*	*3.7*	*6.5*	*3.3*	*3.8*	*2.1*

ND, * and ** indicate not detected, natural and processed samples respectively

Table 3: Average concentrations of non essential elements (µg/g)

Spicie sample	*Bi*	*V*	*Al*	*Pb*	*U*	*As*	*Po*	*Th*
*Murraya koenigii**	*ND*	*ND*	*29.9±0.1*	*ND*	*37±0.6*	*1.3. ND*	*1.4. 0.8±0.4*	*ND*
Zingiber offcinale	*ND*	*ND*	*24.1±0.1*	*ND*	*ND*	*0.5±0.2*	*1.5. ND*	*1.7±0.5*
Cinnamomum tamala	*ND*	*ND*	*8.7±0.1*	*ND*	*ND*	*ND*		*BDL*
Allium sativum	*13.2±0.7*	*ND*	*11.4±0.1*	*ND*	*ND*	*BDL*	*ND*	*ND*
Myristica fragrans	*ND*	*ND*	*1.5±0.2*	*ND*	*19.3±0.6*	*ND*	*ND*	*1.1±0.6*
Laurus nobilis	*ND*	*ND*	*3.3±0.1*	*ND*	*81.0±0.5*	*BDL*	*ND*	*ND*
Monodora myristica	*ND*	*1.8±0.6*	*6.9±0.1*	*ND*	*ND*	*2.1±0.6*	*ND*	*ND*
Microdesmis puberula	*ND*	*ND*	*7.4±0.1*	*ND*	*ND*	*ND*	*ND*	*ND*
Tetrapleura tetraptera	*ND*	*ND*	*7.7±0.1*	*3.8±0.5*	*ND*	*ND*	*ND*	*ND*
Justicia adhatoda	*ND*	*4.5±0.6*	*44.6±0.2*	*ND*	*ND*	*4,3±0.6*	*ND*	*ND*
Gongronema latifolium	*ND*	*ND*	*11.8±0.1*	*ND*	*ND*	*ND*	*ND*	*ND*
Ocimumgratissimum	*ND*	*ND*	*31.2±0.1*	*ND*	*ND*	*ND*	*3.3±0.3*	*ND*
Aframomummelegueta	*4.5±0.8*	*ND*	*3.8±0.1*	*ND*	*ND*	*3.1±0.3*	*2.7±0.2*	*ND*
Syzygium aromaticum	*10.9±0.7*	*ND*	*14.4±0.1*	*ND*	*ND*	*ND*	*ND*	*ND*
Monodora excels	*7.1±0.7*	*ND*	*7.9±0.1*	*ND*	*15.1±0.5*	*ND*	*ND*	*ND*
Xylopia aethiopica	*ND*	*ND*	*7.0±0.1*	*ND*	*ND*	*3.1±0.3*	*ND*	*1.3±0.5*
Piper guineense	*ND*	*ND*	*3.2±0.2*	*ND*	*ND*	*ND*	*ND*	*ND*
Thymus vulgaris	*ND*	*ND*	*52.2±0.2*	*ND*	*26.5±0.6*	*ND*	*ND*	*ND*
*Murraya koenigii***	*ND*	*ND*	*8.1±0.1*	*ND*	*ND*	*ND*	*ND*	*ND*
Myristicafragrans	*ND*	*0.9±0.7*	*9.9±0.1*	*ND*	*ND*	*ND*	*ND*	*ND*
Detection limit	*3.5*	*0.7*	*1.2*	*2.9*	*8.5*	*0.3*	*0.3*	*0.2*

BDL, ND, * and ** indicate below detection limit, not detected, natural and processed samples, respectively.

for increased risk of cancer (Hindin*et al.*, 2005). Vanadium was detected in *Monodora myristica, Justicia adhatoda* and *Myristica fragrans* with observed concentration range between 0.9 and 4.6 µg/g. Aluminium was detected in all the samples with highest concentration of 52.2 ±0.2 µg/g in *Thymus vulgaris* and lowest concentration of 1.5±0.2 µg/g in *Myristica fragrans* samples. Only *Justicia adhatoda* and *Thymus vulgaris* had their concentrations above the permissible limit of 40 µg/day (Dolara and Piero, 2014). Aluminium had been reported to reduce renal function (Ferreira *et al.*, 2008). The possible sources of Th and U could be from naturally occurring radioactive material in the soil where natural spices are cultivated. Although the concentrations of Pb and As as well as Th and U (radioactive) elements in both the natural and industrial spices were very low, prolonged consumption of such spices might lead to gradual accumulation, resulting in concentrations above the allowable levels. This could pose a great risk to human health. However, since there is a growing interest in the consumption of spices based on their dietary benefits throughout the world, a standard platform for the cultivation of spices towards making them available as dietary supplement should be encouraged by all stakeholders.

Conclusion

Some natural and industrial spices have been characterised to provide information on the levels of the minor/major essential and toxic elemental contents as well as the safety or otherwise of their consumption. An optimised PIXE spectrometric technique was employed in the determination of elemental concentration of both harvested and processed spices samples. Considering some essential elements, it was observed that the natural spices have more nutritional benefits when compared with the industrial ones. This implies that these spices are good sources of essential elements and also safe for consumption. The spices that have low level of toxic elements should be taken with caution to reduce their burdens in human body.

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