0163-4984/97/5803-0223 $11.50

PIGE–PIXE Analysis of Medicinal Plants and Vegetables of Pharmacological Importance

S. O. OLABANJI,*[1,2] O. V. MAKANJU,[3] D. CECCATO,[2,4] M. C. BUOSO,[2] A. M. I. HAQUE,[2] R. CHERUBINI,[2] AND G. MOSCHINI[2,4]

[1]International Centre for Theoretical Physics, Trieste, Italy; [2]Istituto Nazionale di Fisica Nucleare (INFN), Laboratori Nazionali di Legnaro (LNL), I-35020 Legnaro, Padova, Italy; [3]Drug Research and Production Unit, Faculty of Pharmacy, Obafemi Awolowo University, Ile-Ife, Nigeria; and [4]Dipartimento di Fisica, Università di Padova, I-35131 Padova, Italy

Received September 8, 1995; Revised March 22, 1996;
Accepted June 1, 1996

ABSTRACT

PIGE and PIXE techniques were employed to the study of elemental constituents of some traditional medicinal plants generally used in curing many diseases and ailments in southwestern Nigeria. Analyses were also carried out on commonly edible vegetables of medicinal and pharmacological importance. PIGE measurements were carried out using 3.5-MeV collimated protons from the 7 mV CN Van-de-Graaff accelerator of INFN, LNL, Legnaro (Padova), Italy, whereas the PIXE measurements were carried out using 1.8 MeV from the 2.5 MV AN 2000 Van-de-Graaff accelerator of the same laboratory. The results show that many of the medicinal plants contain elements of cardinal importance in human metabolism. The results from the vegetables also show the presence of vital elements that are needed for growth and development. In addition, some of the toxic elements, which include As, Cd, Hg, and so forth, were not detected. However, some of the recipes contain trace amounts of Pb at very low concentrations. This calls for proper control of dose rates in some samples to prevent the attendant negative cumulative effects.

*Author to whom all correspondence and reprint requests should be addressed. ICTP Associate on leave of absence from: CERD, Obafemi Awolomo University, Ile-Ife, Nigeria.

Index Entries: PIGE; PIXE; INFN; LNL.

INTRODUCTION

Plants have been used by humans as medicine since time immemorial. Close observation of animal by people in the early days, noting what they use when they fall ill, led to the eventual discovery of medicinal uses of most of these plants *(1)*. It is now a common thing in African society, especially in the rural areas, to treat diseases and ailments using suspected bioactive medicinal plants. This is partly because orthodox medicines are difficult to come by in such areas and partly because of people's insatiable quest for solutions to problems that stare them in the face. Health is of paramount importance in any society. Prior to colonization and before the introduction of orthodox medicine, African people developed a series of medical procedures used in the treatment of various ailments. They relied basically on herbal products, which seem to be the convenient natural gift available to combat such diseases. Various diseases and ailments are identified and concomitant therapies are procured.

Most of these medicaments are made up of combination of different types and parts (conglomeration) of medicinal plants (leaves, stems, roots, and so on) sometimes numbering up to six or more with the addition of commonly used food ingredients. They are usually subjected to different types of local processes, such as grinding, ashing, roasting, and so forth, before making them available for administration or before being displayed for sale. Testimonies abound about the efficacy of various therapies for different diseases to the extent that some even despise orthodox medicine, and prefer the herbal mixtures to modern-day drugs.

It is difficult to jettison the special useful roles these medicaments play in African society. However, lack of knowledge of elemental constituents of these medicinal plants often poses dangers, since some may be poisonous. In addition, the dose rates of many of these locally produced drugs are not well defined, and are usually left to the whims and discretions of patients (the users). This sometimes create problems for users. To hasten cure, some tend to take an overdose, oblivious of the dangers in doing so. This cannot be acceptable if the medicinal plants will continue to serve the people as potent and reliable drugs for different ailments. There is therefore a need to analyze quantitatively the materials used for these drugs in order to suggest the best quantity required for the safe use of the good ones and identify the dangerous and bad ones which are to be avoided.

Most of the plants used in making these concoctions are well known to chemists, and efforts have so far been geared toward effective research into the chemistry of these natural products. However, most scientific efforts, particularly by chemists, have been toward the determination of the active constituents of these plants with a view to identifying, synthesizing, and probably modifying the structure(s) for a wider spectrum of activities.

Little attention have so far been focused on the toxicological studies of these herbs (*2*). The importance of a detailed elemental analysis of traditional medicaments cannot be overemphasized. The imbalance in human health has been linked with the excess or deficiency of trace elements in soils, waters, plants, and animals (*3*). The continued intake of diets that are excessively high in a particular trace element can influence changes in the functioning forms, activities, or concentrations of such element in the body tissue and fluids, so that they rise above the permissible limits (*4*).

In south-western Nigeria, some vegetables have dual roles. They are taken as food (*5*) and medicine (*6*). These vegetables can generally be classified as leafy, fruit, and seed vegetables. Other forms of vegetables include pulses and spices (*5*). However, the present study is focusing on the leafy vegetables that satisfy the dual roles of food and medicine. The analyzed vegetables, apart from being useful as food, also serve medicinally as local remedies for a variety of ailments. Daliel (*6*) reported the local uses of the analyzed vegetables as remedies for inflammation, yaws, skin diseases, rheumatism and swollen joints, syphilis, open wound, flatulence, colic, headache, and neuralgia. They also serve as diuretics, antihelmintics, antiscorbutics, and appetizers, whereas some are sometimes taken superstitiously to prevent punishment, forgetfulness, and to ensure favor.

The reported studies on Nigerian vegetables (*7–9*) have not focused on the elemental composition of the plants. Thus, only limited information is available on both their macro, minor, and trace elements compositions. Many essential elements in diets are toxic at higher concentrations, and the presence of toxic trace elements above permissible levels can lead to impaired health of consumers (*4*). The elemental concentration of these vegetables is required in order to guard against the possible hazards that overconcentration of these elements could have on innocent users.

It is important to epitomize the medicinal uses of some of the analyzed vegetables. "Odu" is used as a digestive tonic and appetizer for sick persons, especially convalescent patients. It is also used as diuretic and depurgative, and for the treatment of yaws. "Ewuro" is used for gastrointestinal troubles, as an antiscorbutic, and as a digestive tonic. It is also used for itching conditions, parasitic skin diseases, ring worms in infants, and so on. Breast-feeding mothers find this particularly useful in stimulating the breast milk and acting as a vicarious preventive of worms in infants. "Ewuro-odo" is used for the treatment of rheumatic pains. It is also used superstitiously to prevent forgetfulness, escape punishment, and to ensure favor.

"Tete" is a fresh plant used as a diuretic. It is also used in enema applications for internal troubles and for pile treatment. "Yarin" is used to induce walking in backward children. It is also used in the treatment of yaws. "Eeku" is used to prevent swelling. "Worowo" is used for wound dressing. "Elegede" is used in vermifuge against tapeworm. It is also used as a coolant for headache and neuralgia. "Sokoyokoto" is used as antiscorbutic and antihelmintic. It is also used as remedy for diarrhea. "Igbagba" is used for the treatment of rheumatic pains and swollen joints. It is also used in the treatment of syphilis.

The vegetable samples were properly identified by a plant taxonomist. The report of Gbile *(10)* supported this identification. A list of local, scientific, and family names of the vegetable samples is given *(10)* below.

Code	Local name (Yoruba)	Scientific name	Family name
EEK	Eeku	*Sesamum radiatum (2)*	Pedaliaceae
ELE	Elegede	*Cucurbita moschata*	Cucurbitacea
EWE-Y	Ewe-yaya	Not available	Not available
EWU	Ewuro	*Vernonia amygdalina*	Compositae
EWU-O	Ewuro-odo	*Struchium sparganonphora*	Compositae
GBU	Gbure funfun	*Talinum triangulare*	Portulacaceae
IGB	Igbagba	*Solanum gilo raddi (2)*	Solanaceae
ODU	Odu	*Solanum nodiflorum (3)*	Solanaceae
OSU	Osun	*Solanum aethiopium*	Solanaceae
SAF	Safara	Not available	Not available
SOK	Sokoyokoto	*Celosia argentea*	Amaranthaceae
TET-F	Tete funfun	*Amaranthus hybridus*	Amaranthaceae
TET-P	Tete pupa	*A. hybridus*	Amaranthaceae
WOR	Worowo	*Senecio biafrae*	Compositae
YAR	Yanrin	*Launea taraxacifolia*	Compositae

This article intends to make known the concentration of these elements in each of randomly sampled traditional medicaments that could cause the use of these products to be encouraged, modified, or discouraged.

EXPERIMENTAL

Some commonly administered traditional herbal medicament samples were collected from Abeokuta, Ado-Ekiti, Akure, Ibadan, Ile-Ife, and Osogbo, representing the four major states in southwestern part of Nigeria. They were coded according to medicinal use and origin as shown in the listing below. The samples were procured from hawkers in a ready-to-be-administered powdered forms. The local producers never use machines for making powdered materials. Instead, they normally use mortars and pestles to crush, grind, and blend the substances together. Others use hard and strong stones as grinders. Mortars, pestles, and stones are normally washed after each use before another preparation to prevent contamination from other sources. The samples were oven-dried at 60°C for 24 h and thereafter stored in air-tight containers. They were then pelletized using a 15- Perkin Elmer pellet machine and its accessory, the 13-mm die.

Diseases for which sample were used			Sources from where samples were obtained	
English name	Yoruba name	Name code	Town	Town code
Gonorrhea	Atosi	AT	Abeokuta	AB
Convulsion	Giri	GR	Ado-Ekiti	AD
Malaria	Iba	IB	Akure	AK
Onchocerciasis	Inarun	IN	Ibadan	IB
Piles/Hemorrhoids	Jedijedi	JD	Ile-Ife	IF
Dizziness	Oyiju	OY	Osogbo	OS

The vegetable samples were procured from the hawkers at Ife central market, Ile Ife, Osun State, Nigeria. They were properly identified by H. C. Illoh, a plant taxonomist at the Department of Botany, Obafemi Awolowo University, Ile-Ife, Nigeria; sample vouchers with samples were kept in the herbarium of the faculty of pharmacy of the same university for further reference. The samples represent the commonly used vegetables in southwestern Nigeria. The leaves and the edible parts of the stem were plucked, air-dried, and cut into pieces. They were further dried in oven at 60°C for 24 h to constant weight. The dried samples were blended to a fine powder using a Moulinex electric blender. The powdered samples were thereafter stored in air-tight sachets and later pelletized using the pellet machine mentioned above. About 250 mg of medicinal plants and 200 mg of vegetable samples were used for making the pellets. The National Bureau of Standards (NBS), Standard Reference Materials (SRM) 1547 (peach leaves) and NBS, SRM 1572 (citrus leaves) also contain the elements of interest and hence the same quantities of standards were used in each case.

For the PIGE measurements, the same quantities of the samples and standards were pelletized and bombarded with 3.5-MeV protons from the 7-MeV CN Van de Graaff accelerator at the Istituto Nazionale di Fisica Nucleare (INFN), LNL at Legnaro, Padova, Italy, while maintaining the same experimental conditions. Thus, the signals came from the same area (diameter of the proton beam) and depth (a few microns) from the surface of both samples and standards, providing calibration data for the determination of concentrations of these elements. The mathematical expression in ref. *(12)* was used for the evaluation of the concentrations. This bombarding energy of 3.5 MeV is adequate for the detection of all light elements of interest using PIGE technique mainly from (p, p′γ), (p, γ) and (p, αγ) reactions.

The PIGE setup used for this work is the same as the one reported in ref. *(11)*. The emitted γ-rays were detected using a Ge(Li) detector with an energy resolution of 2.1 keV (FWHM) at $E_\gamma = 1.33$ MeV and with an efficiency of 22% relative to a 7.6 × 7.6 cm NaI(Tl) detector for 1.33 MeV. The diameter of the entrance collimator was 2 mm. The detector was placed at a distance of 14 cm to the target, which was inclined at 45° to the incoming proton beam. The dead time was below 10% using beam currents of 0.5–30 nA. Energy and efficiency calibrations of the spectra were made using a ^{152}Eu radioactive source. The γ-ray spectra were stored in a Micro-Vax computer and analyzed using the conventional LEONE software. The errors of each photopeak of the light elements were determined by optimizing the best fit to such peaks after careful subtraction of background and then normalized to the same charge.

For the PIXE measurements, the same quantities of samples and standards were pelletized and coated with 20 µg/cm^2 carbon to ensure good electrical contact and bombarded with 1.8 MeV collimated protons from the 2.5 MeVAN-2000 Van de Graaff accelerator at the INFN, LNL at Legnaro, Padova, Italy. All experimental conditions for both samples and standards remained the same. Consequently, the depth and area of information from both samples and standards were the same. The same standards NBS SRM 1547 and NBS SRM 1572 were also used, respectively. The PIXE setup used in this work is the same as reported in ref. *(12)*. The emitted X-rays were detected using a 30-mm^2 Si(Li) detector and the associated electronic setup for routine analysis. The detector resolution was 180 eV (FWHM) at 5.9 keV. The count rates were reduced using a 30-µm thick Al absorber. The dead time of measurements was generally $< 10\%$. A 200 µg/cm^2 gold diffuser foil was used to reduce the beam damage to the target samples. The diameter of the proton beam was 6 mm. The beam current was in the range of 1–18 nA. X-ray data were stored and analyzed using a conventional computer program that incorporated data from standards in its calibration for producing the concentration of all the elements reported.

RESULTS AND DISCUSSION

Medicinal Plants

The results of PIXE measurements for the determination of the concentration of major, minor, and trace elements in medicinal plants are presented in Table 1, but the PIGE results for the concentration of light elements are presented in Table 2. Figure 1 shows a typical PIGE γ-ray spectum of the medicinal plants. The medicinal plant samples were grouped and coded according to their uses to facilitate the comparison of data. Their various uses are indicated in the last column of Table 2.

Fourteen different elements, namely, K, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Rb, Sr, and Pb, were determined using PIXE technique. Many of these elements are of cardinal importance in human metabolism. They are considered essential for the growth of living organisms. The concentrations of potassium and calcium in medicinal plants are relatively high at percentage levels. The concentration of Fe is also relatively high in all the various local drugs for all the listed diseases. Fe together with hemoglobin and ferrodoxin play important roles in human metabolism. Per-

Table 1
PIXE Determination of Elemental Concentration in Medicinal plants (in ppm, Unless Indicated)

Sample	K (%)	Ca (%)	Ti	Cr	Mn	Fe	Co	Ni	Cu	Zn	Br	Rb	Sr	Pb
ATAD	5.1 ± 0.1	2.5 ± 0.2	279 ± 6.6	13.5 ± 1.5	103 ± 2	1207 ± 13	-	6.8 ± 0.7	23.8 ± 0.6	55 ± 1	7.5 ± 0.8	15.9 ± 1.3	86.3 ± 2.5	6.8 ± 1.5
ATIB	2.6 ± 0.3	12.7 ± 0.4	52.1 ± 8.3	-	23.6 ± 1.6	667 ± 2.5	15.4 ± 2.5	5.8 ± 0.7	9.9 ± 0.5	12.2 ± 0.6	16.1 ± 0.9	7.3 ± 1.5	502.6 ± 5.6	10 ± 1.8
ATOS	5.2 ± 0.2	3.4 ± 0.3	247± 12	10 ± 2.6	161 ± 3.3	141 ± .16	-	6 ± 1.3	26.4 ± 1.1	71.2 ± 1.6	24.9 ± 1.8	25.2 ± 2.9	87.8 ± 4.5	-
GRAD	46.3 ± 0.1	-	14.9 ± 2.4	-	2.3 ± 0.7	165 ± 1	4.9 ± 0.8	2.2 ± 0.4	17.2 ± 0.4	12.2 ± 0.4	78.3 ± 0.9	194 ± 2	3.7 ± 0.6	3.4 ± 1.1
IBAD	1.9 ± 0.1	6.4 ± 0.2	108± 7	9.2 ± 1.4	32 ± 1.2	720 ± 2	8.2 ± 2.2	4.6 ± 0.5	15.2 ± 0.3	12.6 ± 0.4	3.5 ± 0.4	4.1 ± 0.8	175.7 ± 2.5	5.6 ± 1
IBAK	1.1 ± 0.1	0.5 ± 0.1	15.3 ± 2.6	2.4 ± 0.6	17.2 ± 0.6	381± 2	-	1.4 ± 0.3	2.9 ± 0.2	6 ± 0.2	-	2.5 ± 0.6	15.7 ± 1	2.7 ± 0.7
IBIB	3.2 ± 0.1	5.8 ± 0.1	34.2 ± 3.8	3 ± 1	38.1 ± 0.9	603± 2	-	2.8 ± 0.4	5.8 ± 0.3	24.5 ± 0.4	11 ± 0.5	10.7 ± 0.8	127.9 ± 1.9	13± 1.
IBIF	1.7 ± 0.1	5.2 ± 0.1	74.5 ± 4.4	6.3 ± 0.9	27.0 ± 0.8	448 ± 1.3	4.6 ± 1.4	1.8 ± 0.4	2.9 ± 0.2	5.8 ± 0.2	-	6.6 ± 0.6	81.6 ± 1.8	-
IBOS	2.0 ± 0.1	0.9 ± 0.1	649 ± 6	25.5 ± 1.2	89.8 ± 1.5	982± 2.4	-	5.2 ± 0.6	4.9 ± 0.4	27.6 ± 0.5	14.7 ± 0.8	18 ± 1.1	35.4 ± 1.4	4.9 ± 1.3
INAB	13 ± 0.1	1.7 ± 0.2	276± 7	25.5 ± 1.6	43.8 ± 1.8	1022 ± 3.2	-	15.9 ± 0.9	17.1 ± 0.7	134 ± 1.3	10.6 ± 0.9	76.3 ± 2.3	32.2 ± 1.8	-
INAD	2 ± 0.3	12.4 ± 0.3	80.6 ± 7.7	9 ± 1.6	22.6 ± 1.5	647 ± 2	14.1 ± 2.3	5.9 ± 0.7	54.5 ± 0.7	20.6 ± 0.6	14.3 ± 0.7	7.3 ± 1.2	262 ± 3.5	7.2 ± 1.6
INAK	6.0 ± 0.1	1.9 ± 0.1	83 ± 3	-	80.1 ± 0.8	334± 2	3.6 ± 1.1	3 ± 0.3	9 ± 0.2	19.1 ± 0.3	6.6 ± 0.4	25 ± 0.9	57 ± 1.3	4.2 ± 0.7
INIB	3.1 ± 0.2	7.8 ± 0.3	44 ± 6	8.4 ± 1.4	19 ± 1.2	520 ± 2.2	8 ± 2	4.3 ± 0.5	8.5 ± 0.4	20.6 ± 0.6	3.3 ± 0.6	13.5 ± 1.3	190 ± 4	4.1 ± 1.5
INIF	2.1 ± 0.2	13.2 ± 0.3	101 ± 6.4	8.9 ± 1.4	48.5 ± 1.2	683 ± 2.2	9.1 ± 2.2	4.3 ± 0.6	4.7 ± 0.4	11.9 ± 0.4	2.8 ± 0.5	3.5 ± 1	196.3 ± 3.5	-
INOS	8 ± 0.1	1.8 ± 0.1	32.7 ± 4.1	2 ± .1	22 ± 1	279± 2	4.9 ± 1.2	2.2 ± 0.4	3.8 ± 0.3	14.2 ± 0.4	15 ± 0.6	4.9 ± 0.8	62.3 ± 1.7	3.4 ± 1
JDAB	10.5 ± 0.2	6.2 ± 0.3	51.4 ± 6.1	-	53 ± 1.3	494.4 ± 2	12.9 ± 1.8	20.1 ± 3.6	86 ± 1.8	80.5 ± 2	10.8 ± 3	15.8 ± 4.3	116.7 ± 5.8	-
JDAK	17.5 ± 0.4	5.1 ± 0.6	105± 15	-	107± 6	8395 ± 14	-	7.1 ± 0.6	15.3 ± 0.5	40.6 ± 0.7	27 ± 0.9	29.5 ± 1.5	239.7 ± 3.6	-
JDIB	3.3 ± 0.3	16.7 ± 0.4	123± 9	12.5 ± 2	22.3 ± 1.6	930± 23	22 ± 2.8	6.4 ± 0.7	22.5 ± 0.6	19.5 ± 0.6	19.5 ± 0.9	7.2 ± 1.4	665.7 ± 5.6	9 ± 1.7
JDOS	6.8 ± 0.1	3.9 ± 0.2	167± 7	-	176± 2	1600± 4	-	6.7 ± 1	10.9 ± 0.6	74.7 ± 0.9	32.2 ± 1.2	33.8 ± 1.9	102.7 ± 2.6	6.3 ± 1.9
OYAD	2.6 ± 0.2	10.3 ± 0.3	739± 14.	19.3 ± 3	163 ± 2.7	1856± 4	-	6.3 ± 1.2	9.1 ± 0.6	41.5 ± 0.8	10.5 ± 1	20.1 ± 1.6	369.6 ± 4.7	-

Table 2
Elemental Concentrations (in ppm) in Medicinal Plants Samples Obtained with PIGE Analysis

Sample	Na	Mg	Al	Cl	Activity
ATAD	34437 ± 2593	1268 ± 74	172 ± 6	1566 ± 124	Gonorrhoea
ATIB	2128 ± 160	18507 ± 107	586 ± 19	-	
ATOS	-	32846 ± 2340	702 ± 23	30273 ± 2417	
GRAD	2073 ± 163	2449 ± 149	915 ± 30	563 ± 67	Anticonvulsant
IBAD	2146 ± 172	-	1552 ± 50	-	Antimalarial
IBAK	716 ± 60	135 ± 18	221 ± 7	-	
IBIB	2374 ± 194	-	1773 ± 57	-	
IBIF	1851 ± 150	-	313 ± 10	-	
IBOS	1959 ± 166	-	416 ± 47	-	
INAB	378 ± 28	834 ± 48	60 ± 2	844 ± 67	Onchocerciasis
INAD	14813 ± 1111	4140 ± 248	429 ± 14	653 ± 52	
INAK	2645 ± 575	7371 ± 433	656 ± 21	604 ± 77	
INIB	-	49944 ± 2981	1417 ± 46	-	
INIF	1576 ± 118	5048 ± 293	500 ± 16	-	
INOS	3248 ± 244	-	297 ± 10	-	
JDAB	7106 ± 550	6992 ± 410	781 ± 25	-	Pile Haemorrhoids
JDAK	834 ± 69	2486 ± 145	240 ± 8	1147 ± 106	
JDIB	230 ± 80	2746 ± 164	307 ± 10	858 ± 84	
JDOS	10029 ± 776	-	1028 ± 33	783 ± 95	
OYAD	39643 ± 2989	1388 ± 93	667 ± 22	715 ± 63	Dizziness

haps the therapeutic propensity of various medicinal plant recipes hinges on the concentrations of these three elements. It is worthy of note, too, that Rb is present in all various therapies. K is known to have close physiochemical relationship with rubidium. Ca is important for the growth and development of bones and teeth.

Mn, Cu, Ni, and Zn are present at low concentrations (ppm levels) in all the samples. Grimanis et al. *(13)* reported that Cu is a very important and essential trace element for organisms partly because it acts as a catalyst for fat oxidation. In addition, Cu enzymes have been reported to be uniquely important in catalyzing the reduction of molecular oxygen to water.

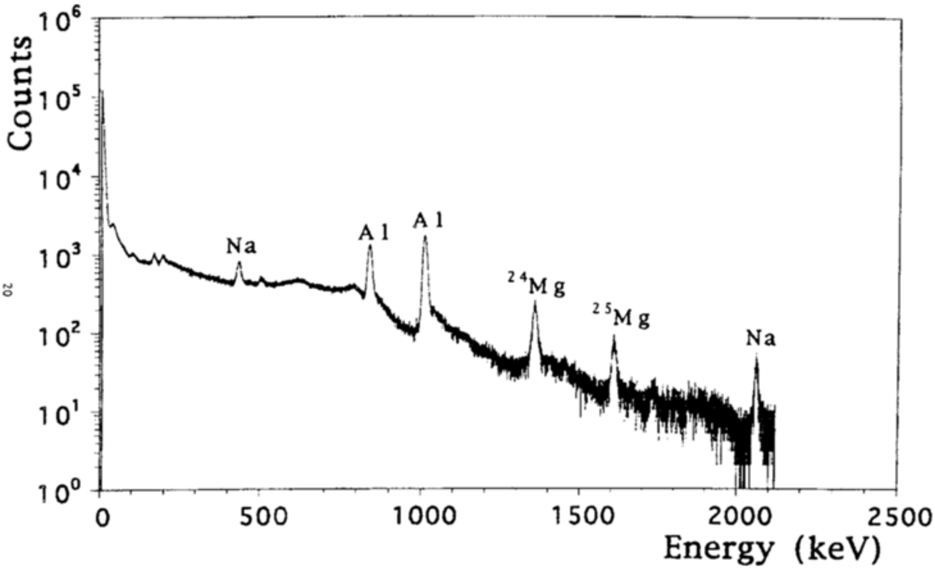

Fig. 1. A typical γ-ray spectrum of medicinal plant samples.

For the treatment of gonorrhea, malaria, onchocerciasis, pile, hemorrhoids, and especially dizziness, it is apparent that relatively high concentrations of Ti, Fe, and Sr are very prominent, as reflected in the results of the coded samples for their treatments. The efficacy of these locally produced drugs may depend on them. However, one is not sure whether some of these concentrations are approaching the toxic level, especially those of Sr. One should therefore take some of these recipes with some caution to prevent any possible attendant hazardous effects of their use. The significant difference in the Fe content of the samples used for piles/hemorrhoids, JDAK, JDIB, and JDAB having concentrations of 8395.5, 930.1, and 494.4 ppm, respectively, is quite interesting. The cause(s) of this could be owing to differences in the mode of their preparation or in the location of growth of the plants *(4)*. The concentrations of Cr, Co, and other trace elements are below the accepted toxic levels as presented by Bowen *(14)*.

The prominent light elements detected in all these locally produced drugs using PIGE techniques are Na, Mg, and Al. Other elements determined are P and Cl. Some of these elements are of immense importance in human metabolism. The concentrations of Na, Mg, and Al are generally relatively high, as evidenced in Table 2. In particular, the relatively high Na concentration may pose some dangers for patients who suffer from hypertension, because Na is known to favor and enhance high blood pressure in humans. Most therapies have high sodium contents, which are inimical to the health of such patients, and consequently, these patients can be advised to refrain from taking them except in the treatment of malaria fever. However, some recipes contain trace amounts of Pb at very low concentrations. This may be because of contamination from the atmosphere and soil. Although it is true that the detected concentration of Pb falls below the toxic level of 1 mg/d, there is a need to have proper control of dose rates of the medicinal plants to prevent the concomitant negative cumulative effects.

Vegetables

Tables 3 and 4 show the PIXE results for vegetables of medicinal and pharmacological importance, whereas Table 5 presents the PIGE data. As a test of the accuracy of our measurements, we also irradiated two NBS Standard Materials (SRM 1547 and 1572), namely, peach and citrus leaves, respectively. The results are displayed in Table 4. The agreement between the certified results and our measurements are very good as evidenced. The uncertified values are put in parentheses. The concentrations are in ppm unless otherwise indicated.

PIXE results show the presence of vital elements in the vegetables that are needed for human growth and development. It is interesting to see the significant differences between the elemental compositions of the two varieties of *A. hybridus* (TETE) examined.

K and Rb are found to be higher in the white *A. hybridus* (TET-F) than in the red species. Rb, which is about 300% higher in the white variety (215.9 ppm), is ironically the highest in all the vegetables analyzed. However, purified diets containing up to 200 ppm or Rb are nontoxic, but levels > 1000 ppm lead to a decrease in growth, reproductive performance, and survival time *(4)*. Other elemental compositions of the *A. hybridus* show higher values in the red specie with Fe content taking the lead. The differences in their elemental composition may be as a result of the effects of geochemical positions of their cultivation.

The Fe content of the vegetables are relatively high compared with other elements determined. Apart from *V. amygdalina* having 172.2 ppm of Fe, all other vegetable samples showed an Fe content of between 303.6 and 2866 ppm. The importance of Fe has been stressed above. The observed concentrations may pose no health hazards, since Fe absorption is significantly lower and the dietary Fe requirement is expectedly higher in vegetal diets than foods from other sources *(15,16)*. K and Ca contents of the vegetables are expectedly high in reasonable percentage, whereas Cr and Pb are absent in some samples. The presence of Pb in some of the samples is probably owing to environmental pollution or contamination, rather than taken as a constituent in the growth of the vegetables. Interestingly, the trace elements determined are at permissible levels that pose no health hazards to the users. Further biogeochemical studies on some of the vegetables may be necessary to establish the environmental effect on their elemental compositions.

Table 3
PIXE Determination of Elemental Concentrations in Vegetables of Medicinal and Pharmacological Importance

Sample	K %	Ca %	Ti	Cr	Mn	Fe	Ni	Cu	Zn	Br	Rb	Sr	Pb
TET-P	16.4 ± 0.2	3.24 ± 0.03	236.8 ± 5.8	-	333.2 ± 2.2	1018 ± 3	11.6 ± 0.8	13.7 ± 0.5	219.9 ± 1.2	26.9 ± 1.0	79.4 ± 2.0	128. ± 3	-
ELE	15.8 ± 0.2	5.78 ± 0.03	268.2 ± 6.1	46.1 ± 1.8	135.1 ± 2.5	1697 ± 3.6	10.3 ± 1	23.2 ± 0.6	50.8 ± 0.8	8.9 ± 0.9	44.3 ± 1.8	139 ± 3	-
EWU	14.0 ± 0.1	0.74 ± 0.02	-	-	177.8 ± 1.3	172.2 ± 1.4	4 ± 0.4	26.7 ± 0.5	56.3 ± 0.7	8.1 ± 0.6	56.1 ± 1.5	10.7 ± 1.0	-
SOK	46.5 ± 0.3	0.81 ± 0.05	136.8 ± 4.9	8.4 ± 1.4	34.8 ± 1.7	968.1 ± 2.5	5.2 ± 0.9	12.6 ± 0.6	102.4 ± 1.0	5.6 ± 0.8	34.5 ± 1.5	27.1 ± 1.4	14.7 ± 1.8
EWUO	14.6 ± 0.2	4.98 ± 0.03	337.1 ± 6.7	-	88 ± 3.3	2603 ± 4	11.9 ± 1.6	34.4 ± 0.7	136.2 ± 1.1	7.4 ± 1.2	43.1 ± 1.9	86.6 ± 2.4	7.2 ± 2.4
GBU	34.3 ± 0.2	0.76 ± 0.04	88.7 ± 4.6	-	169.3 ± 1.7	303.6 ± 2	5.3 ± 0.6	11.8 ± 0.7	70.3 ± 1.0	9.7 ± 0.7	27.0 ± 1.5	23.8 ± 1.4	-
TET-F	18.4 ± 0.2	1.10 ± 0.02	75.0 ± 4.7	-	99.7 ± 1.2	391 ± 2	3.0 ± 0.6	6.1 ± 0.7	65 ± 1	23.8 ± 1.0	215.9 ± 3.1	65.1 ± 2.0	-
IGB	15.8 ± 0.2	3.84 ± 0.03	231.4 ± 6.3	10.1 ± 1.5	46.9 ± 1.7	1306 ± 3	7.3 ± 1.0	30.2 ± 0.7	74.2 ± 1.1	17.2 ± 1.0	24.4 ± 1.7	63.8 ± 2.4	5.7 ± 1.7
WOR9	19.1 ± 0.2	1.20 ± 0.03	115.7 ± 5.7	18.5 ± 1.6	104.3 ± 1.7	963.4 ± 3.1	10.1 ± 0.6	36.8 ± 0.7	52.2 ± 1.0	10.2 ± 0.8	26.4 ± 1.5	62.3 ± 2.2	4.8 ± 1.8
ODU	8.71 ± 0.13	1.22 ± 0.02	113.5 ± 5	18.8 ± 1.4	40.2 ± 1.5	1121 ± 3	11.9 ± 0.8	37.7 ± 0.7	66.3 ± 0.9	-	22.4 ± 1.4	28.4 ± 1.4	6.1 ± 1.9
EEK	9.35 ± 0.14	1.71 ± 0.02	68.5 ± 3.9	4.7 ± 1.5	29 ± 1	410.6 ± 1.8	5.8 ± 0.6	28.2 ± 0.7	43.3 ± 0.7	5.0 ± 0.6	11.8 ± 1.0	29.0 ± 1.4	-
OSU	16.5 ± 0.2	1.17 ± 0.02	84.6 ± 5.2	6.4 ± 1.5	367.5 ± 2.1	593.6 ± 2.9	9.9 ± 0.6	26.5 ± 0.7	86.3 ± 1.1	20.9 ± 1.0	32.2 ± 1.6	70.7 ± 2.1	2.0 ± 1.6
YAR	27.6 ± 0.3	5.28 ± 0.05	613 ± 11	15.4 ± 2.5	84.6 ± 3.0	2866 ± 6	14.4 ± 1.7	21.6 ± 0.9	51.6 ± 1.1	31.1 ± 1.6	64.9 ± 3.1	93.8 ± 3.5	8.4 ± 2.9
SAF	20.6 ± 0.2	1.16 ± 0.03	204.6 ± 5.2	20.0 ± 1.3	31.8 ± 1.4	1147 ± 3	6 ± 0.8	7.5 ± 0.5	34.9 ± 0.6	15.9 ± 1.0	58.7 ± 1.9	27.3 ± 1.5	2.6 ± 1.6
EWE-Y	20.6 ± 0.2	2.23 ± 0.03	90.5 ± 5.3	8.3 ± 1.4	68.3 ± 1.5	932.9 ± 2.9	8.7 ± 0.7	26.5 ± 0.7	101.7 ± 1.1	1.8 ± 0.7	77.4 ± 2.3	35.6 ± 1.8	-

Table 4
Comparison of Our PIXE Measurements with Certified Values from NBS SRM 1547 and 1572, Peach and Citrus Leaves, Respectively

Elements	Peach L. Measured	Peach L. NBS SRM	Citrus L. Measured	Citrus L. NBS SRM
K	(2.36 ± 0.05) %	(2.43 ± 0.03) %	(2.03 ± 0.07)%	(1.82 ± 0.06) %
Ca	(1.41 ± 0.01) %	(1.56 ± 0.02) %	(3.16 ± 0.01) %	(3.15 ± 0.10) %
V	7.4 ± 1.5	0.37 ± 0.03	4.8 ± 1.3	0.26 ± 0.03
Cr	---------	---------	2.3 ± 0.9	0.8 ± 0.2
Mn	100.2 ± 1.2	98 ± 3	25.4 ± 0.9	23 ± 2
Fe	274.2 ± 1.6	(220)	156.7 ± 1.2	90 ± 10
Co	4.2 ± 1.3	(0.07)	6.1 ± 1.0	(0.02)
Ni	2.1 ± 0.5	0.69 ± 0.09	2.7 ± 0.5	0.6 ± 0.3
Cu	6.9 ± 0.4	3.7 ± 0.4	23.9 ± 0.6	16.5 ± 1.0
Zn	23.9 ± 0.6	17.9 ± 0.4	44.3 ± 0.8	29 ± 2
Ga	2.1 ± 0.5	(1)	1.5 ± 0.6	------
As	1.1 ± 0.5	0.06 ± 0.02	5.9 ± 0.8	3.1 ± 0.3
Se	-------	0.12 ± 0.01	0.6 ± 0.7	(0.025)
Br	13.1 ± 0.8	(11)	10.1 ± 0.8	(8.2)
Rb	21.1 ± 1.4	(19)	6.8 ± 1.2	4.84 ± 0.06
Sr	65.4 ± 2.2	53 ± 4	123.0 ± 3.1	100 ± 2
Y	---------	---------	3.7 ± 1.7	-------
Ba	---------	124 ± 4	---------	---------
Pb	--------	0.87 ± 0.03	19.6 ± 2.3	13.3 ± 2.4

CONCLUSIONS

PIGE and PIXE complementary techniques were employed for the determination of elemental constituents of some commonly administered traditional herbal medicaments in southwestern Nigeria. The results are of immense importance in monitoring, controlling, and modifying the therapeutic effects of these very important and commonly used traditional African medicinal products. The results were geared toward solving unique African problems and are of immense scientific importance. The novel results show that many of these locally produced drugs contain elements of immense importance in human metabolism. Toxic elements, which include As, Cd, Hg, and so on, were not detected. However, some of the medicaments contain trace amounts of Pb, the concentrations of which are below the accepted toxic level. The results show no serious danger for the use of most of the medicinal plants, but suggest a caution for some of their uses by hypertensive patients because of

Table 5
Elemental Concentrations (ppm) in Vegetables of Medicinal and Pharmacological Importance Using PIGE Technique

Samples	Na	Mg	Al	P	Cl
TET-P	-	1271 ± 74	425 ± 14	-	-
ELE	-	716 ± 42	566 ± 18	-	-
EWU	-	729 ± 24	423 ± 18	-	-
SOK	-	2435 ± 147	107 ± 3	-	-
EWUO	-	1272 ± 74	438 ± 14	-	-
GBU	-	1321 ± 77	616 ± 20	-	-
TET-F	-	1230 ± 71	687 ± 22	-	-
IGB	-	1179 ± 125	528 ± 17	-	-
WOR	-	2646 ± 154	211 ± 7	-	-
ODU	-	2415 ± 140	236 ± 8	-	-
EEK	1498 ± 112	-	616 ± 20	314 ± 100	-
OSU	1517 ± 114	5711 ± 337	456 ± 15	567 ± 101	827 ± 87
YAR	821 ± 232	7799 ± 459	521 ± 17	-	-
SAF	2993 ± 232	9319 ± 547	453 ± 15	-	825 ± 97
EWE-Y	2246 ± 169	4112 ± 243	347 ± 11	417 ± 92	612 ± 76

the high concentration of Na in most of them. There is a need to have controlled and acceptable dose rates if medicinal plants will continue to serve the people in Africa as potent and reliable drugs for these diseases. The results did not show any important reason to discourage the use, but some of them must be modified to prevent the attendant negative effects of their use.

Vegetables of pharmacological and medicinal importance in southwestern Nigeria were also analyzed using PIXE and PIGE techniques. The results show the presence of elements that are of immense importance in human metabolism. Many of the toxic elements were not detected. Thus, human dietary intakes, which include vegetables, are extremely important as sources of vital elements needed for growth and development. In particular, African unique vegetables with their unique therapeutic effects proved to be very useful for human consumption both as food and medicine. The agreement between our measurements and the certified values in NBS SRM 1547 peach leaves and SRM 1572 citrus leaves was very good, establishing good accuracy of our measurements.

ACKNOWLEDGMENTS

S. O. Olabanji, has carried out this work with the support of the ICTP program for training and research in Italian Laboratories, Trieste, Italy. The work was concluded under the ICTP Associateship program. He wishes to express his profound appreciation and gratitude to G. Furlan and O. Turra. He also wishes to thank the International Centre for Theoretical Physics, Trieste, for hospitality. He is grateful to Obafemi Awolowo University, Ile-Ife, Nigeria for granting the leave. The authors wish to thank I. Motti for dexterous operation of the CN accelerator. The assistance of G. Manente in sample preparation is appreciated. They would also like to thank C. Baiocchi for his assistance with PIGE electronics and the Ge (Li) detector.

REFERENCES

1. M. Rombi, *Phytotherapy*, Herbal Health Publisher, Surrey (1982).
2. M. Yoshiki and O. Nagayo, *Chem. Pharm. Bull.* **32,** 591 (1984).
3. E. A. V. Prasad and V. Raghu, *Environ. Geochem. Health,* **16,** 76 (1994).
4. E. J. Underwood, *Trace Elements in Human and Animal Nutrition,* 4Th ed., Academic, New York (1977).
5. Van C. W. Epehuijsen, Growing Native Vegetables in Nigeria, FAO of United Nations, Rome (1974).
6. J. M. Dalziel, *The Useful Plants of West Tropical Africa,* Crown Agents for Overseas Governments and Administration, London (1955).
7. O. L. Oke, *J. A. Diet Assoc* **53,** 130 (1968).
8. E. T. Ifon and O. Bassin, *Food Chem.* **5,** 231 (1980).
9. V. A. Oyenga, *Nigeria's Food and Teading Stuffs,* Ibadan University Press, Ibadan (1968).
10. Z. O. Gbile, *Vernacular Names of Nigerian Plants in Yoraba,* University Press, Ibadan, Nigeria.
11. S. O. Olabanji, R. Cherubini, S. Fazinic, G. Moschini, and D. Zafiropoulos, *Nucl. Instr. and Methods in Phys. Res.* **B56/57,** 726 (1991).
12. S. O. Olabanji, A. M. I. Haque, S. Fazinic, R. Cherubini, and G. Moschini, *J. Radioanal. Nucl. Chem.* **177,** 243 (1994).
13. A. P. Grimanis, M. Vassilaki-Grimani, D. Alexiou, and C. Papadatos, Proceedings of Nuclear Activation Techniques in Life Sciences IAEA-SM-227/26, p. 241 (1979).
14. H. J. M. Bowen, Environmental Chemistry of the Elements, Academic, London p. 144 (1979).
15. L. Hallberg, L. Garby, R. Suwanik, and E. Bjorn-Rasmussen, *Am. J. Clin. Nutr.* **27,** 826 (1974).
16. M. Layrisse, C. Martinez-Torres, and M. Gonzalez, *Am. J. Clin. Nutr.* **27,** 152 (1974).