



## **Nutritional and Spectral Characteristics of *Terminalia* Plants**

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### **Authors' contributions**

*The present work was carried out in collaboration among all authors. Author KSP designed the research study and supervised the whole research work. Author SC collected the plant samples and performed experimental work. Author KPR managed the literature and statistical work. Author EKT generated the mineral data. Author JMG collected the FTIR spectra. Author PMR wrote the and revised manuscript. All the Authors read and approved the final manuscript.*

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### **ABSTRACT**

**Aims:** *Terminalia* spp. is medicinal plants that belong to Combretaceae family, widely used in traditional Ayurvedic medicine. In this work, the nutritional constituents of the leaves, seed kernel and seed coat from four *Terminalia* species (*T. arjuna*, *T. bellirica*, *T. catappa* and *T. chebula*) are reported.

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**Methodology:** Polyphenol and flavonoid contents were analyzed spectrophotometrically by using Folin-Ciocalteu and aluminum chloride as reagents, respectively; mineral contents were quantified by using X-ray fluorescence; and the functional groups of the phytochemicals were investigated by infrared spectroscopy.

**Results:** The total concentration of 20 macro- and micronutrients and heavy metals (viz. P, S, Cl, K, Rb, Mg, Ca, Sr, Ba, Al, Ti, Cr, Mn, Fe, Co, Cu, Zn, Mo, As and Pb), and the total polyphenol and flavonoid contents in the seed kernels ranged from 1754 to 65521 mg/kg, from 2150 to 51100 mg/kg and from 63 to 42300 mg/kg, respectively. Polyphenol and mineral contents for the *Terminalia* spp. seed coats and leaves were also determined. The enrichment in each of aforementioned elements with respect to the soil content was calculated. The vibrational spectra of the leaves and seed coats agreed with a composition rich in lignin, hemicellulose, cutin, pectin and flavonoids, while those of the seed kernels were in accordance with the presence of unsaturated oils, protein, and fiber.

**Conclusion:** Various parts of the four *Terminalia* species under study (*T. arjuna*, *T. bellirica*, *T. catappa* and *T. chebula*) featured high contents of nutrients and polyphenols needed for biological metabolism and human health. In addition, heavy metals were only present at traces level, indicating that these *Terminalia* plants would be safe for medicinal uses.

**Keywords:** *Terminalia*; FTIR; XRF; flavonoid; phenolic; nutrients.

## 1. INTRODUCTION

*Terminalia* genus comprises around 100 species distributed in tropical regions of the world. Trees of this genus, common in plains and low hills in India, are well known as a source of secondary metabolites, such as tannins, cyclic triterpenes and their derivatives, flavonoids, and other aromatics. Tannin-containing cells occur throughout the plant body, particularly in the pericarp of the fruit.

*Terminalia* species are important medicinal plants: they are administered as astringent and purgative, and are used in dropsy, diarrhea, piles, leprosy, and cough treatments [1]. The phytochemical and pharmacological profile of *Terminalia arjuna* (Roxb. ex DC.) Wight & Arn., known as Arjun, has been reported in the review paper by Jain et al. [2]. *T. bellirica* (Gaertn.) Roxb., known as Bahera or Beleric, and *Terminalia chebula* Retz., known as Chebulic myrobalan, are two main constituents of Triphala, traditionally used to treat various gastrointestinal disorders [3], and an evaluation of the pharmacological activities of the latter has been covered in a review paper by Bag et al. [4]. The phytoconstituents and pharmacological benefits of *Terminalia catappa* L., known as Indian-Almond, have been discussed in the review paper by Anand et al. [5].

The antioxidant, antifungal and antibacterial properties of some species of *Terminalia* have

been reported in the literature [6,7,8,9,10]. The chemical composition (proteins, lipids, carbohydrates, starch and fiber) of *T. catappa* fruits from Brazil was reported by dos Santos et al. [11], and the fatty acid composition of *T. catappa* kernels from Benin and Thailand was studied by Ladele et al. [12] and Weerawatanakorn et al. [13]. There is also data on the volatile compounds identified in the fruits and essential oils from *T. arjuna*, *T. catappa* [14, 15,16,17,18,19] and *T. chebula* [20], but other nutritional-related information remains unreported. In this work, a comparative study of the nutritional (i.e. polyphenolic and trace elements) content in the leaves, seed kernel and seed coat from the four *Terminalia* species mentioned above (*T. arjuna*, *T. bellirica*, *T. catappa* and *T. chebula*) are presented.

## 2. MATERIALS AND METHODS

### 2.1 Sampling of Plants

The four *Terminalia* species discussed herein (viz. *T. arjuna* (TA), *T. bellirica* (TB), *T. catappa* (TC) and *T. chebula* (TCh)) grow massively in the Chhattisgarh region, in the center-east of India. They were botanically authenticated with the aid of standard monographs [21]. The leaves and fruits of TA and TB were collected in May 2017 from Raipur city (21.25°N 81.63°E), whereas the leaves and fruits of TC and TCh were collected in December 2017. The near-surface layer of the soils was also sampled.

## 2.2 Sample Preparation

The leaves, pericarp, seed coat and seed kernel were manually separated. First, they were cleaned with de-ionized water and dried with a hot air blower. They were sundried in a glass room for one week, further dried for 24 h at 50°C in a hot air oven, and finally stored in glass containers. The plant and soil samples were crushed into fine powder with a mortar and particles of mesh size of  $\leq 100 \mu\text{m}$  were sieved out. They were preserved at -4°C in a deep freezer till the analyses were conducted.

## 2.3 Analyses

Sample weights were measured with a Mettler-Toledo (Columbus, OH, USA) electronic balance. The moisture content of the samples was determined by drying at 105°C in an air oven for 6 h prior to the analysis. All characterization results are presented on a dry weight (dw) basis.

The total phenolic content (TPC) and flavonoid content (Fla) were determined as follows: Firstly, 100 mg of powdered sample was dispersed in 5 mL of an acetone: water (70:30, v/v) solution, which was sonicated in an ultrasonic bath for 20 min at 20°C. Then, 5 mL of fresh acetone: water (70:30, v/v) solution was added to the mixture and the extraction was repeated for 20 min at 20°C. After centrifugation, the supernatant was collected. The total phenolic content of each extract was determined as tannic acid equivalents (TAE) by using the Folin-Ciocalteu reagent [22]. The flavonoid content was determined by the aluminum chloride method as quercetin equivalents (QE) [23].

For macro- and micronutrient analyses, X-ray fluorescence (XRF) technique was chosen, using a Bruker III Tracer SD (T3S2731 (Kennewick, WA, USA) spectrometer equipped with a 4W rhodium anode and Xflash SDD 2028 channels. Standard brown and white cowpea (*Vigna unguiculata* (L.) Walp.) seeds and a soil sample ((NCS DC 73382 CRM) were used for calibration.

The Fourier-Transform Infrared (FTIR) spectra were characterized with a Thermo Scientific (Waltham, MA, USA) Nicolet iS50 spectrometer equipped with an in-built diamond attenuated total reflection (ATR) system. Spectra were collected in the 400-4000  $\text{cm}^{-1}$  spectral range,

with a 1  $\text{cm}^{-1}$  spectral resolution and averaging 64 scans.

All analyses were carried out in triplicate, and mean values are reported.

## 3. RESULTS AND DISCUSSION

### 3.1 Plant Characteristics

The physical characteristics of the leaves and seeds from the four *Terminalia* species are shown in Table 1. The leaves, seeds and seed kernels were colored, with various shapes, as shown in Fig. 1. Micrographs of leaves samples are shown in Fig. 2. The average mass of a single leaf of TA, TB, TC and TCh was 2367±41, 3700±66, 7500±142 and 3767±67 mg, respectively. The mass of a single seed on dry weight basis- was 3885±75, 4373±81, 4762±78 and 5426±102 mg, with a kernel fraction of 3.1%, 11.0%, 8.3% and 2.1%, respectively (provided that seed coats were hard and thick, and accounted for a remarkably high fraction of the seed weight). The water content in the leaves, seed coat and seed kernel ranged from 2.8% to 4.9%.

### 3.2 Phenolic Content

The phenolic content for the four *Terminalia* species is shown in Table 1, with TPC values in the leaves, seed coat and seed kernel ranging from 23900 to 33100 mg/kg, from 22400 to 51100 mg/kg, and from 2150 to 9530 mg/kg, respectively. Similarly, Fla concentration in the leaves, seed coat and seed kernel varied in the 11200–25900 mg/kg, 5300–42300 mg/kg, and 63–2150 mg/kg range, respectively. Plant parts from TCh were found to contain the highest contents of TPC and Fla. The Fla/TPC ratio in the leaves, seed coat and seed kernel showed mean values of 0.69, 0.54 and 0.11, respectively.

For comparison purposes, the TPC contents and Fla contents for leaves and seeds from *T. cattapa* reported herein (Table 1) were slightly lower than those reported by Rajesh et al. [24] for other Indian samples: 38.21 mg TPC/g and 45.65 mg TPC/g, and 41.23 mg Fla/g and 43.86 mg Fla/g, for the leaves and entire seeds (coat + kernel), respectively. For fruits from the same species, Ladele et al. [12] reported a TPC of 35.5 mg/g for samples from Benin, and Weerawatanakorn et al. [13] reported a TPC of 51.1 mg/g for samples from Thailand.



Fig. 1. *T. arjuna*, *T. billerica*, *T. cattapa* and *T. chebula* (from left to right) leaves, seeds and kernels (from top to bottom)



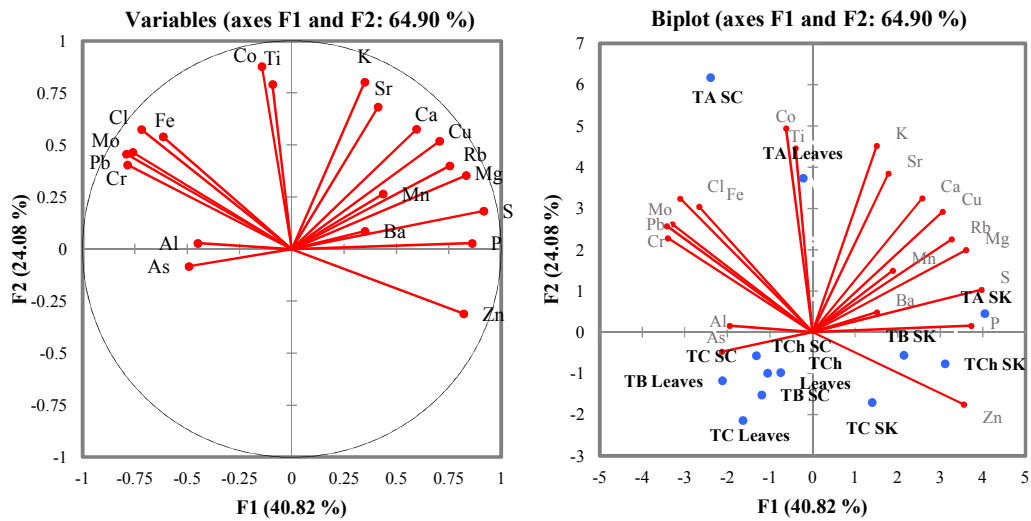
Fig. 2. Micrographs of *T. arjuna*, *T. billerica*, *T. cattapa* and *T. chebula* (from left to right) leaves samples at 50× (upper row) and 500× (lower row)

### 3.3 Macro- and Micronutrients Contents

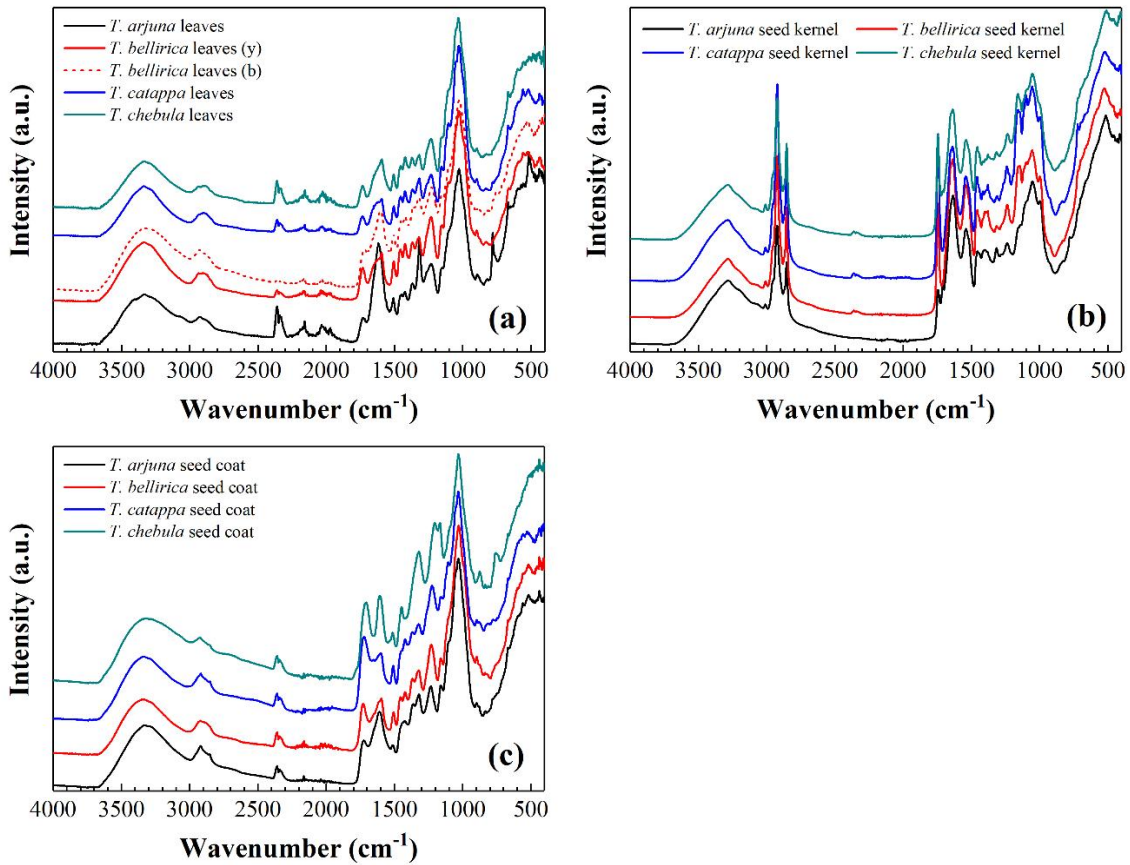
The mineral element concentrations are also presented in Table 1, and a comparison with the values reported in the literature for *T. cattapa* seed kernel is summarized in Table 2. As regards macronutrients, which play a major role in plant physiological processes, P concentration in leaves, seed coat and seed kernel ranged from 51 to 772 mg/kg, from 287 to 1109 mg/kg, and from 3842 to 8171 mg/kg, respectively. Relatively higher concentrations of K were detected, which varied in the 288–9364 mg/kg, 3683–16001 mg/kg, and 4334–13947 mg/kg range in the leaves, seed coats and seed

kernels, respectively. Rubidium, which has chemical properties similar to  $K^+$  in the biological processes [27], showed concentrations in the leaves, seed coats and kernels of 1–16 mg/kg, 9–25 mg/kg, and 13–28 mg/kg, respectively.

Apropos of the secondary macronutrients (viz. S, Mg and Ca), the concentrations of S in the leaves, seed coat and kernel were in the 71–606 mg/kg, 176–545 mg/kg, and 1166–3158 mg/kg intervals, respectively. Magnesium concentrations in the leaves, seed coat and kernel varied from 105 to 1868 mg/kg, from 11 to 1316 mg/kg, and from 828 to 5440 mg/kg, respectively. Calcium concentrations in the



**Fig. 3.** Principal component analysis results: (a) correlation circle; (b) biplot. TA = *T. arjuna*; TB = *T. bellirica*; TC = *T. catappa*; TCh = *T. chebula*; SC = seed coat; SK = seed kernel



**Fig. 4.** ATR-FTIR spectra for (a) leaves, (b) seed kernel, and (c) seed kernel samples from the four species of the *Terminalia* genus under study

Table 1. Physicochemical characteristics of the different parts (leaves, seed kernel and seed coat) from the four *Terminalia* species under study. All mineral concentrations are expressed in mg/kg

Param-eter	<i>Terminalia arjuna</i>			<i>Terminalia bellirica</i>			<i>Terminalia cattapa</i>			<i>Terminalia chebula</i>		
	Leaves	Seed kernel	Seed coat	Leaves	Seed kernel	Seed coat	Leaves	Seed kernel	Seed coat	Leaves	Seed kernel	Seed coat
Color	bright green	pale yellow	brown	light green	light brown	grey	light green-elliptical	light brown	reddish	bright green	pale yellow	brown
Shape	oblong-lanceolate	oblanceolate	winged shape	obovate-elliptical	obovate	broadly	obovate-elliptical	lanceolate-cylindrical	ovoid -compressed	ovate-lanceolate	narrow oblong	ellipsoid
Mass, mg	2367	120	3765	3700	483	3890	7500	393	4369	3767	113	5313
Moisture content, %	3.2	3.8	3.7	3.4	5.0	3.5	4.2	4.8	2.8	4.1	4.9	3.7
TPC, mg/kg	30600	7250	48600	23900	2180	25200	29900	2150	22400	33100	9530	51100
Fla, mg/kg	25900	725	31300	20200	138	5300	11200	63	10700	23200	2150	42300
Fla/TPC	0.85	0.1	0.64	0.85	0.06	0.21	0.37	0.03	0.48	0.7	0.23	0.83
Mg	1868	5440	1316	368	1967	11	105	828	17	121	2666	21
Al	427	-	-	21	-	-	27	-	-	32	-	-
P	51	<b>8171</b>	1109	281	4775	287	128	3842	403	772	6942	665
S	426	<b>3158</b>	545	389	1961	176	71	1166	243	606	2549	342
Cl	<b>3346</b>	-	<b>3287</b>	46	-	304	54	-	917	789	-	173
K	9364	13947	<b>16001</b>	1764	8444	3683	288	4334	8913	6340	7302	7582
Ca	<b>49656</b>	9443	6644	2567	6949	1916	919	2031	699	2071	4500	1014
Rb	16	13	25	8	28	9	1	21	9	8	28	9
Sr	101	28	20	9	15	4	3	2	2	9	15	4
Ba	39	3	1	1	12	1	2	2	2	1	1	1
Ti	-	-	42	-	-	-	-	-	-	-	15	-
Cr	1	-	37	7	-	1	1	-	9	2	-	2
Mn	14	88	63	21	17	3	16	22	28	66	38	9
Fe	179	140	937	229	79	100	127	83	383	179	71	416
Co	6	1	1.5	1	1	1	1	1	1	1	1	1
Cu	15	<b>38</b>	<b>771</b>	2	23	5	2	23	3	1	17	5
Zn	5	53	1	6	22	1	3	53	1	7	59	5
As	1	-	-	2	-	-	1	-	-	1	-	-
Mo	4	-	20	1	-	7	3	-	2	1	-	4
Pb	2	-	11	10	-	1	2	-	2	2	-	1

TPC and Fla stand for total phenolic content and total flavonoid content, respectively. “-“ indicates non-detectable levels

**Table 2. Comparison of mineral compositions of *T. catappa* seed kernel samples reported in the literature**

Element (mg/kg)	(12)*	(25)	(13)	(26)	This work
Na	376.0	-	-	-	-
Mg	7290.9	2364.5	3647	-	5440
P	-	-	8899	161	8171
K	17181.2	-	7311	-	13947
Ca	4150.1	2451	3252	2580	9443
Mn	42.5	-	-	-	88
Fe	161.5	51.4	54	14	140
Cu	46.3	3.78	25	-	38
Zn	96.7	6.2	61	9	53
Pb	18040	-	-	-	-

\* Defatted kernels

**Table 3. Bio-accumulation factors for mineral nutrients in the different parts of the four *Terminalia* species under study**

Element	<i>Terminalia arjuna</i>			<i>Terminalia bellirica</i>			<i>Terminalia cattapa</i>			<i>Terminalia chebula</i>		
	Leaves	Seed kernel	Seed coat	Leaves	Seed kernel	Seed coat	Leaves	Seed kernel	Seed coat	Leaves	Seed kernel	Seed coat
<b>P</b>	0.4	59.2	8.0	2.0	34.6	2.1	0.9	27.8	2.9	5.6	50.3	4.8
<b>S</b>	2.1	15.3	2.6	1.9	9.5	0.9	0.3	5.6	1.2	2.9	12.3	1.7
<b>Cl</b>	26.3	0.0	25.9	0.4	0	2.4	0.4	0	7.2	6.2	0	1.4
<b>K</b>	6.5	9.7	11.1	1.2	5.9	2.6	0.2	3.0	6.2	4.4	5.1	5.3
<b>Rb</b>	2.3	1.9	3.6	1.2	4.1	1.3	0.1	3.0	1.3	1.2	4.1	1.3
<b>Mg</b>	1.2	3.5	0.9	0.2	1.3	0	0.1	0.5	0	0.1	1.7	0
<b>Ca</b>	7.9	1.5	1.1	0.4	1.1	0.3	0.1	0.3	0.1	0.3	0.7	0.2
<b>Sr</b>	2.1	0.6	0.4	0.2	0.3	0.1	0	0	0	0.2	0.3	0.1
<b>Ba</b>	1.1	0.1	0	0	0.3	0	0.1	0.1	0.1	0	0	0
<b>Cu</b>	0.2	0.5	10.7	0.0	0.3	0.1	0	0.3	0	0	0.2	0.1

leaves, seed coat and kernel were in the 919–49656 mg/kg, 699–6644 mg/kg and 2031–9443 mg/kg ranges, respectively. Strontium showed concentrations in the range of 3-101, 2-20 and 2-28 mg/kg for leaves, seed coat and seed kernel, respectively. Barium was detected in the leaves, seed coat and kernel at 1-39, 1-2 and 1-12 mg/kg concentrations, respectively.

Chloride was detected only in the leaves and seed coat of all *Terminalia* species, ranging from 46 to 3346 and from 173 to 3287 mg/kg, respectively.

Titanium, which stimulates enzyme activities and the uptake of nutrients [28], was detected only in the TA seed coat and TCh seed kernel at low levels, 15 and 42 mg/kg, respectively.

Chromium was identified in the leaves and seed coats of all species at 1-7 and 1-37 mg/kg concentrations, respectively.

Manganese, necessary in the photosynthesis and nitrogen metabolism, was identified in all

parts of the *Terminalia* spp., and varied from 14 to 66, from 3 to 63, and from 17 to 88 mg/kg for the leaves, seed coats and seed kernels, respectively.

Iron, involved in production of chlorophyll, lignin formation, etc., was detected at moderate to high levels, varying from 127-229, 100-937 and 71-140 mg/kg for the leaves, seed coats and kernels, respectively.

Cobalt, an essential component of several enzymes, was detected at low levels (1-6 mg/kg) in all parts of the *Terminalia* species.

Copper, necessary for carbohydrate and nitrogen metabolism, was detected in the leaves, seed coats and kernels of all *Terminalia* species, ranging from 1 to 15 mg/kg, from 3 to 771 mg/kg, and from 17 to 38 mg/kg.

Zinc –an essential component of various enzyme systems for energy production, protein synthesis, and growth regulation– was identified in the

Table 4. Correlation coefficients among the phenolic, flavonoid, macro- and micronutrient contents

	TPC	Fla	Mg	Al	P	S	Cl	K	Ca	Rb	Sr	Ba	Ti	Cr	Mn	Fe	Co	Cu	Zn	As	Mo	Pb	
TPC	1																						
Fla	<b>0.9</b>	1.0																					
Mg	-0.5	-0.4	1.0																				
Al	0.2	0.3	0.1	1.0																			
P	-0.7	-0.6	0.8	-0.3	1.0																		
S	-0.7	-0.6	<b>0.9</b>	-0.2	<b>1.0</b>	1.0																	
Cl	0.5	0.5	0.0	0.6	-0.4	-0.3	1.0																
K	0.1	0.2	0.6	0.1	0.4	0.4	0.5	1.0															
Ca	0.1	0.2	0.3	<b>1.0</b>	-0.1	0.0	0.7	0.3	1.0														
Rb	-0.4	-0.3	0.4	0.0	0.6	0.6	0.2	0.5	0.2	1.0													
Sr	0.1	0.2	0.4	<b>0.9</b>	0.0	0.1	0.7	0.3	<b>1.0</b>	0.2	1.0												
Ba	0.0	0.1	0.2	<b>0.9</b>	-0.1	-0.1	0.6	0.2	<b>1.0</b>	0.2	<b>0.9</b>	1.0											
Ti	0.4	0.3	0.1	-0.1	0.1	0.0	0.6	0.6	0.0	0.5	0.0	-0.2	1.0										
Cr	0.5	0.4	-0.1	-0.1	-0.3	-0.3	0.6	0.5	-0.1	0.2	0.0	-0.2	<b>0.9</b>	1.0									
Mn	-0.1	-0.1	0.6	-0.2	0.5	0.5	0.1	0.6	-0.1	0.2	0.0	-0.2	0.4	0.3	1.0								
Fe	0.7	0.6	-0.2	-0.1	-0.3	-0.3	0.6	0.6	-0.1	0.1	0.0	-0.2	0.8	<b>0.9</b>	0.3	1.0							
Co	0.2	0.3	0.1	<b>1.0</b>	-0.3	-0.2	0.7	0.2	<b>1.0</b>	0.1	<b>1.0</b>	<b>1.0</b>	0.0	0.0	-0.2	0.0	1.0						
Cu	0.4	0.4	0.1	-0.1	-0.1	-0.1	0.6	0.6	0.0	0.4	0.0	-0.1	<b>0.9</b>	<b>1.0</b>	0.4	<b>0.9</b>	0.0	1.0					
Zn	-0.7	-0.6	0.7	-0.2	<b>0.9</b>	<b>0.9</b>	-0.4	0.1	-0.1	0.5	0.0	-0.1	0.0	-0.4	0.4	-0.4	-0.2	-0.2	1.0				
As	0.2	0.3	-0.3	0.4	-0.5	-0.4	0.1	-0.5	0.2	-0.5	0.2	0.2	-0.3	-0.1	-0.1	-0.2	0.3	-0.2	-0.4	1.0			
Mo	0.7	0.5	-0.2	0.0	-0.4	-0.4	0.7	0.5	0.1	0.2	0.1	0.0	0.8	<b>0.9</b>	0.1	<b>0.9</b>	0.1	<b>0.9</b>	-0.5	-0.2	1.0		
Pb	0.5	0.5	-0.2	0.0	-0.4	-0.4	0.5	0.1	0.0	0.0	0.0	-0.1	0.6	0.8	0.2	0.7	0.0	0.7	-0.5	0.5	0.6	1	

TPC and Fla stand for total phenolic content and total flavonoid content, respectively. Values higher than 0.8 have been highlighted in boldface



**Table 5. Main absorption bands in the ATR-FTIR spectra for leaves, seed coats and kernels from four species of the *Terminalia* genus (all wavenumbers are expressed in  $\text{cm}^{-1}$ )**

Leaves				Seed kernel				Seed coat			
<i>T. arjuna</i>	<i>T. bellirica</i>	<i>T. catappa</i>	<i>T. chebula</i>	<i>T. arjuna</i>	<i>T. bellirica</i>	<i>T. catappa</i>	<i>T. chebula</i>	<i>T. arjuna</i>	<i>T. bellirica</i>	<i>T. catappa</i>	<i>T. chebula</i>
3331	3334	3334	3331	3282	3282	3285	3284	3330	3330	3335	3332
				3008	3008	3008	3808				
2924	2929	2901	2924	2923	2923	2921	2923	2918	2918	2917	2924
				2853	2853	2852	2853				
2360	2359	2360	2360					2359	2359	2358	2359
1734b		1733	1734	1743	1745	1744	1744	missing	missing	missing	missing
1716y	1716			missing	missing	missing	missing	1723	1723	1723	1723
				1709			1709				1706
1617b				1634	1636	1637	1636	1623			
1606y	1606	1594	1618	missing	missing	missing	missing	1609	1609	1597	1607
				1538	1540	1541	1540	missing	missing	missing	missing
1508	1506	1505	1508	missing	missing	missing	missing	1508	1508	1507	1509
1449	1447	1455	1449	1455	1456	1457	1456				1446
1420	1423	1422	1421	1402	1399	1415	1416	1424	1424	1418	
1369		1368	1369		1379	1378	1378	1362	1362		
1317	1318	1318	1326	1316	1314	1311	1316	1318	1318	1321	1319
1231	1229	1231	1232	1238	1237	1240	1235	1232	1232	1224	1204
1154y	1154	1155			1144	1158	1156	1158	1158	1159	
		1104			1097	1097	1093			1104	
1027	1027	1027	1027	1050	1054	1054	1053	1031	1031	1031	1030
				997	996	996		missing	missing	missing	missing
893	897	896	893	missing	missing	missing	missing	896	896	896	874
							831		834		835
780			780	778							
668		668	668			720			667		668
557		559	558					558	558	557	

b: black portion; y: yellow portion

leaves, seed coats and kernels, varying from 3 to 7 mg/kg, from 1 to 5 mg/kg, and from 22 to 59 mg/kg, respectively.

Molybdenum, involved in enzyme systems relating to nitrogen fixation by bacteria, was found at low concentrations in the leaves and seed coats (1-4 and 2–20 mg/kg, respectively). Arsenic was detected in leaves at low levels, 1-2 mg/kg. Pb was found at concentrations of 2-10 and 1-11 mg/kg in the leaves and seed coats, respectively.

The total concentration of 20 elements (i.e. P, S, Cl, K, Rb, Mg, Ca, Sr, Ba, Al, Ti, Cr, Mn, Fe, Co, Cu, Zn, Mo, As and Pb) in the leaves, seed coat and seed kernel of TA, TB, TC and TCh was 65521, 30832 and 40523; 5733, 6510 and 24293; 1754, 11634 and 12408; and 11009, 10254 and 24204 mg/kg, respectively. Remarkably high concentrations of the elements in all parts of TA were detected.

### 3.4 Soil Characteristics and Bioaccumulation Factors

In Chhattisgarh region, red laterites or entisol soils cover 19.5% of the cultivated area and yellow clayey inceptisol soils account for 14.8%, but in Raipur district the latter are the most frequent. These soils are slightly alkaline (mean value, 7.7; range 7.3–8.0), and show electrical conductivities (EC) in the range of 465–523  $\mu\text{S}/\text{cm}$ , with a mean value of 495  $\mu\text{S}/\text{cm}$  (indicating an appreciable accumulation of salts).

The concentration in major and minor elements in the surface soil varied in the 114-141 mg/kg range for Cl (mean value, 127 mg/kg); 119–162 mg/kg for P (mean value, 138 mg/kg); 179–240 mg/kg for S (mean value, 207 mg/kg); 6.0–9.0 mg/kg for As (mean value, 7.5 mg/kg); 1339–1510 mg/kg for K (mean value, 1438 mg/kg); 5.8–8.0 mg/kg for Rb (mean value, 6.9 mg/kg); 1450-1623 for Mg (mean value, 1545 mg/kg); 5880–6710 for Ca (mean value, 6304 mg/kg); 44–58 for Sr (mean value, 50 mg/kg); 29–45 for Ba (mean value, 37 mg/kg); 5460–7050 for Ti (mean value, 6412 mg/kg); 113–150 for Cr (mean value, 128 mg/kg); 1370–1660 for Mn (mean value, 1510 mg/kg); 17460–20123 for Fe (mean value, 18818 mg/kg); 29–38 for Co (mean value, 32 mg/kg); 66–82 for Cu (mean value, 72 mg/kg); 73–96 for Zn (mean value, 87 mg/kg); 1.0–1.8 for Mo (mean value, 1.4 mg/kg) and 2–29 mg/kg for Pb (mean value, 15.5 mg/kg). They were found to occur in the following increasing

order: Mo < Rb < As < Pb < Co < Ba < Sr < Cu < Zn < Cl  $\approx$  Cr < P < S < K < Mn < Mg < Ca < Ti < Fe.

The K/P ratio (=10.4) was in good agreement with the ratio obtained from potassium and phosphorus values (=10.5) reported by Awanish et al. [29].

The bioaccumulation factor (BAF) is a ratio of the concentration of an element in the plant to the concentration of that element in soil, and depends on several factors, such as plant genotype, bioavailability of metals, soil quality, climatic condition, agronomic management, etc. BAFs are reported in Table 3. Several nutrients (K, P, Cl, S, Ca) were hyperaccumulated by the four *Terminalia* species, with the highest hyperaccumulation of K, P, Cl and S for *T. arjuna*.

### 3.5 Statistical Analysis of Phenolic, Macro- and Micronutrients Contents in Seed Kernel

The correlation coefficients of the elements for the *Terminalia* spp. seed kernels are presented in Table 4. TPC showed a good correlation with the Fla, P, S, Mg and Zn contents, which exhibited high positive correlations with each other. Strong statistical correlations were found among P, S, K, Mg, Ca, Sr, Mn, Fe and Cu, indicating their accumulation as cofactor elements.

A principal component analysis was also conducted for the mineral constituents. The percentage of variability represented by the first two factors was ca. 65%. The correlation circle (Fig. 3a) shows a projection of the initial variables in the factors space. In the biplot (Fig. 3b), i.e., a simultaneous representation of variables and observations in the PCA space, it could be observed that the characteristics of *T. arjuna* seed coat and seed kernel were unique. The seed coat and leaves from *T. bellirica*, *T. cattapa* and *T. chebula* would share common characteristics, and so would the seed kernels from the four species.

### 3.6 Vibrational Characterization

The ATR-FTIR spectra for leaves, seed coat and seed kernel samples from the four species of the *Terminalia* genus under study are depicted in Fig. 4. The corresponding bands are summarized in Table 5.

Peaks at around  $3330\text{ cm}^{-1}$  (OH stretching) corresponded to typical characteristic absorption from cellulose [30]. Peaks at around  $2920\text{ cm}^{-1}$  ( $-\text{CH}_2$  aldehydic symmetrical stretching) and at  $2853\text{ cm}^{-1}$  ( $-\text{CH}$  stretching) indicated the presence of cutine and wax. Peaks at ca.  $1740$  and at around  $1370\text{ cm}^{-1}$  were indicative of hemicellulose, specifically of  $\text{C}=\text{O}$  stretching ( $1734\text{ cm}^{-1}$ ) and  $-\text{CH}_3$  symmetric deformation ( $1379\text{-}1362\text{ cm}^{-1}$ ). Prominent bands in the  $1340$  to  $890\text{ cm}^{-1}$  region were also attributed to cellulose: At  $1336\text{ cm}^{-1}$  ( $\delta\text{CH}$  in-plane), at  $1321\text{-}1311\text{ cm}^{-1}$  (C-H vibration), at around  $1150\text{ cm}^{-1}$  ( $\nu\text{C-O-C}$  in bridge, asymmetric), at  $1031\text{-}1027\text{ cm}^{-1}$  ( $\nu\text{C-O}$  or  $-\text{C-O-C}$  stretching) and at ca.  $896\text{ cm}^{-1}$  ( $\nu\text{C-O-C}$  in bridge, symmetric, characteristic of the glycosidic ring in cellulose). The presence of pectin was indicated by peaks associated with  $\text{COO}^-$  asymmetric and  $\text{O-CH}_3$  stretching (at  $1457\text{-}1447\text{ cm}^{-1}$ ) for calcium pectate and with  $-\text{CH}_3$  distortion ( $1240\text{-}1229\text{ cm}^{-1}$ ) for pectic ester. The band that appeared at around  $1424\text{ cm}^{-1}$  can be attributed either to cellulose ( $\rho\text{CH}_2$ , sym.) or to symmetric stretching vibration for calcium pectate [31]. Bands at around  $831\text{ cm}^{-1}$  were due to aromatic C-H out-of-plane bending or to C-O-C deformation and suggested the presence of D-Glc pyranoside configurations. Bands at  $780\text{ cm}^{-1}$ , assigned to  $\text{O-C}=\text{O}$  in-plane deformation or to a  $\text{CH}_2$  rocking deformation, were attributed to phenolic components.

For samples from leaves and seed coat, two bands attributed to lignin could be observed: the band of the aromatic ring stretching of the lignin ( $1606\text{ cm}^{-1}$ ), which appeared at  $1618\text{-}1594\text{ cm}^{-1}$ ; and the band of the aromatic skeletal vibration ( $\text{C}=\text{C}$  aromatic symmetrical stretching), at  $1509\text{-}1505\text{ cm}^{-1}$ .

Seed kernel samples showed strong characteristic bands at around  $1744\text{ cm}^{-1}$ ,  $1636\text{ cm}^{-1}$ , and  $1540\text{ cm}^{-1}$ . The band at  $1744\text{ cm}^{-1}$ , assigned to  $\text{C}=\text{O}$  (non-conjugated moieties vibrations) could be associated to the stretching vibration of the ester carbonyl functional groups of the triglycerides. The peak obtained at around  $1636\text{ cm}^{-1}$  could be characteristic of  $\text{C}=\text{C}$  absorption cellulose when it is cross-linked and dehydrated, but it may also be assigned to amide  $\text{N-H}$  &  $\text{C}=\text{O}$  stretching from mucilage [32] or to an enrichment in unsaturated oils. The presence of this band, typical of the vinyl group, would justify the quantitative presence of unsaturated oils in the kernel of all the seeds under study.

*Analysis of band maxima positions.* The absorption bands that occur at  $3330\text{ cm}^{-1}$  for

seed coat and leaves samples appeared shifted to  $3380\text{ cm}^{-1}$  for kernel samples. The absorption band at  $1723\text{ cm}^{-1}$  found in seed coats was shifted to  $1743\text{ cm}^{-1}$  in seed kernels. As regards the band that occurred at  $1053\text{ cm}^{-1}$  for kernel samples, shifts to  $1031\text{ cm}^{-1}$  for seed coats and to  $1027\text{ cm}^{-1}$  for leaves were observed. The band at  $558\text{ cm}^{-1}$  was absent in seed kernel samples.

Results from the FTIR spectra of leaves and seed coats showed that they are rich in lignin, hemicellulose, cutin, pectin and flavonoids, while unsaturated oils, protein, and fiber would be the main constituents of the seed kernels.

#### 4. CONCLUSIONS

The nutritional potential of four *Terminalia* species (*T. arjuna*, *T. bellirica*, *T. catappa* and *T. chebula*) was investigated with a view to their valorization as a new source of nutrients. All the species examined, especially *T. arjuna*, showed high concentrations of phenols and macro- and micronutrients. The highest TPC and Fla contents occurred in the seed coats and leaves. P, S, K, and Rb appeared hyperaccumulated in the four *Terminalia* species. The differences in the FTIR spectra for the seed kernels, seed coats and leaves have been referred to the different contents in some components (unsaturated oils, lignin and flavonoids).

#### CONSENT

It is not applicable.

#### ETHICAL APPROVAL

It is not applicable.

#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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