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Comprehensive Nutritional Analysis, Antioxidant Activities, and Bioactive Compound Characterization from Seven Selected Cereals and Pulses by UHPLC-HRMS/MS

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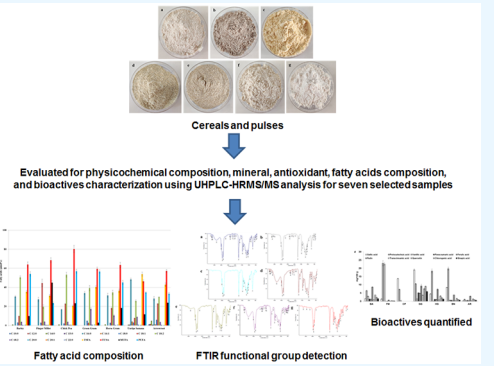
ABSTRACT: Cereals and pulses comprise the largest proportion in a typical Indian diet plate. This research mainly focuses on determining the nutritional composition, bioactive compound characterization, and antioxidant activities of seven selected cereals and pulses. The total carbohydrate content was high in unripe banana (67.65/100 g) and arrowroot (63.76/100 g). Finger millet (44.55 μmol %), chickpea (53.33 μmol %), and green gram (17.40 μmol %) showed high oleic, linoleic, and linolenic acid contents, respectively. The ascorbic acid content was the highest in chickpea and horse gram at 86.83 and 83.76 mg/100 g, respectively. The major phenolics and flavonoids quantified and confirmed using HPLC and UHPLC-HRMS/MS were gallic, protocatechuic, vanillic, para-coumaric, ferulic, chlorogenic, sinapic, and trans-cinnamic acids, rutin, and quercetin. The sample extracts showed dose-dependent antioxidant activity to combat the reactive oxygen species. Hence, these serve as an excellent source for the development of functional food formulations for lowering the risk of various diseases.



INTRODUCTION

Cereals and pulses are a type of grains, where cereals are grasses and pulses are legumes. Cereals are the members of the Gramineae family, whereas the pulses belong to the Leguminosae family and are an excellent source of carbohydrate, dietary fiber, and protein. Cereals and pulses play a predominant role in the diets of developing countries, and the nutritional problem is associated with traditional, complementary foods.[1] Pulses are rich in protein, dietary fiber, and various micronutrients and biologically active substances. Therefore, the Indian Pulse and Grain Association has described pulses as a future major source for nutritional and health benefits.[2]

Cereals, pulses, and related foods has long been advocated as alternative protein and energy sources for infants and young children food products.[1] Among the selected cereals, barley is a good old grain with many health benefits such as weight reduction, decreasing blood pressure, blood cholesterol, and blood glucose in type-2 diabetes, and preventing colon cancer. It is a readily available, low-cost grain and contains both soluble and insoluble fiber, protein, vitamins B and E, the minerals selenium, magnesium, and iron, copper, flavonoids, and anthocyanins.[3] Finger millet is the primary source of dietary fiber and polyphenols, which offers several health benefits such as antidiabetic, antioxidant, hypocholesterolemic, antimicrobial, and protective effects against diet-related chronic diseases.[4]

Pulses are the richest source of dietary fibers and complex carbohydrates, facilitating low-glycemic index foods. Pulses help to lower cholesterol and triglycerides, as leguminous fibers cause hypoglycosuria because they consist of more amylose than amylopectin. The proteins from pulses are readily available, as shown by *in vitro* digestibility.[5] Among the selected pulses, chickpea is a plant known for a long time in Asia. The extract of chickpea is used in the treatment for diarrhea or indigestion. The grains have been used to increase body weight, cure headache, sore throat, and cough.[6] It also has several potential health benefits, such as to treat cardiovascular diseases (CVD), type-2 diabetes, digestive diseases, and cancer.[7] Green gram is rich in proteins and could be considered as a meat alternative for vegetarians. In addition to that, it possesses potential health benefits such as antioxidant, anticancerous, anti-inflammatory, and hypolipi-

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demic activities. Green gram has prebiotic and nutraceutical properties.[8] Horse gram also has enormous potential to cure various diseases such as common cold, throat infection, fever, urinary stones, asthma, bronchitis, and leucoderma. It also contains proteinase inhibitors that could act as anti-inflammatory agents and treat obesity and several degenerative and autoimmune diseases.[9]

Unripe banana (*Musa paradisiaca*) has been used as a source of starch and nutrients since the 13th century.[10] It belongs to the Musaceae family, and unripe bananas are dried, powdered, and commercially available in local markets. The dried powders could serve as potent antioxidants and are rich in vitamin C. Arrowroot is more commonly used in biscuits, porridges, and sweets and is also used to produce bread, ice cream stabilizers, and baby food.[11] The previous studies reported that the arrowroot extract could increase the serum levels of IgM, IgG, and IgA immunoglobulins in mice.[12]

Hence, in view of the present significant attributes to cereals and pulses for food-to-food fortification, mainly for nutrient and mineral deficiencies, there is a need to explore comprehensive scientific data as a prerequisite for further subjects to make food formulations and also to impart value addition, especially under the homestead concept. Accordingly, the present study focuses on investigating the nutritional composition, minerals, pigments, fatty acid profiling (gas chromatography (GC)/mass spectrometry (MS)), Fourier transform infrared (FT-IR), identification, characterizations (high-performance liquid chromatography (HPLC) and ultra-HPLC (UHPLC) high-resolution MS (HRMS)/MS) of bioactive (phenolics and flavonoids) and antioxidant activities of the selected samples.

MATERIALS AND METHODS

Sources of the Raw Material. The selected seven (7) different cereals and pulses, i.e., barley, finger millet, chickpeas, green gram, horse gram, unripe banana, and arrowroot powder, were procured from the local markets of Davangere, Karnataka, India. These selected cereals and pulses were cleaned and roasted in a common household kitchen stove in a low flame until before turning to light brown. Then, these were allowed to cool to room temperature and made into powders separately using a household mixer (Figure S1). The obtained powders were packed in an air-tight container in the dark until further use.

Chemicals. For nutritional analysis, the analytical-grade chemicals were purchased from Sisco Research Laboratory (Bangalore, India). All the phenolic and flavonoid standards, 2,2′-diphenyl-1-picrylhydrazyl (DPPH) and 2,2′-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS), were obtained from Sigma-Aldrich, Bangalore, India. For HPLC analysis, degassed HPLC-grade methanol, acetonitrile, and Milli-Q water were used. Similarly, for UHPLC-HRMS/MS analysis, MS-grade solvents were purchased from JJ Biotech, Bangalore, India.

Nutrient Composition. The moisture content in the prepared powder samples was analyzed by drying at 50 °C overnight in a hot air oven to determine the moisture content with the Da 2a−48 method.[13] The total carbohydrates (phenol sulfuric acid method) and reducing sugars (3,5-dinitrosalicylic acid method) were analyzed using glucose as a standard, and the total protein (Folin−Ciocalteu's reagent) with bovine serum albumin (BSA) as a standard in phosphate buffer extracts of powder samples was determined.[14] The total oil content was determined using hexane at 60 °C for 8 h from dried (moisture-free) powders by following the Soxhlet extraction method Ba 3−38.[13] The ash content was obtained by incinerating the powder samples in a muffle furnace for 8 h at 550 °C in crucibles to ensure complete carbon burning. After cooling to room temperature, the obtained ash was weighed and recorded.[14]

The crude fiber in the dried powder sample was analyzed according to the method reported.[14] Briefly, a known quantity of the sample was boiled for 30 min with 0.255 M sulfuric acid containing boiling chips in a water bath. After cooling to room temperature, the digested sample was passed through a muslin cloth, and the residue was washed several times with distilled water. Furthermore, the residue was boiled for 30 min with 0.313 N sodium hydroxide, and samples were passed through a muslin cloth and washed thrice with distilled water and ethanol. The obtained residue was transferred to a preweighed ashing dish (W_1) and fried in an oven set at 110 °C for about 3 h, and the weight was recorded after cooling (W_2). The moisture-free residue was finally transferred to the muffle furnace set at 550 °C for 8 h, and the weight (W_3) was recorded after cooling. The percentage of crude fiber was calculated as mentioned below,

$$\text{crude fibre (\%)} = \frac{(W_2 - W_1) - (W_3 - W_1)}{\text{weight of the sample}}$$

W_1 = empty weight of the crucible, W_2 = weight of the crucible with the sample, and W_3 = weight of the crucible after ashing.

The total calorie content of the powder samples was calculated as,

Total calorie content = 4 × percentage of proteins + 9 × percentage of lipids + 4 × percentage of carbohydrates.

Color Measurement. The color of the powder samples was measured using a color measurement instrument (Konica Minolta CM-5, VA) by measuring L, a^*, and b^* values.[15] The L values represent the lightness or darkness, the a^* value refers to the red-green color, and the b^* values refer to the blue-yellow color. Based on the obtained L, a^*, and b^* values, the chroma and hue angles were calculated accordingly.

Mineral Analysis. Minerals were estimated by microwave plasma−atomic emission spectroscopy (model MP-AES-4200; Agilent Technologies, Bangalore, India) with a plasma interfaced. A known quantity of the powder samples was incinerated in a muffle furnace for 3 × 90 min at 550 °C to ensure complete carbon burning. The obtained ash was dissolved in aqua regia (3 HCl:1 HNO_3), and the residue-free top layer was used. Iron, zinc, copper, magnesium, potassium, sodium, and calcium were analyzed after diluting with the respective acid solution. The minerals were quantified using reference standard (Agilent Technologies, Bangalore, India) calibration and generated standard curves.

Estimation of Chlorophyll and Carotenoids. The selected powder samples were extracted with acetone in a mortar and pestle (1:10 w/v) and centrifuged at 6000rpm for 10 min, and the supernatant was collected. The absorbance of the collected clear supernatant was measured at 661.5, 663, 645, and 450 nm with a double-beam spectrophotometer (UV-160 A, Shimadzu Corporation, Kyoto, Japan), and the concentrations of chlorophyll a, chlorophyll b, total chlorophyll, and carotenoids were calculated and compared with two standard methods.[16,17]

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Particle Size Distribution. The particle size and particle distribution of the prepared powdered samples were determined using a laser light diffraction instrument (S3500, Microtrac, Largo, Florida), and samples were drawn with vacuum pressure inside the analyzer, and the analysis was carried out.[18] The volume mean diameter has been analyzed (D43) to find the particle size of the powders. The recorded data were analyzed using Flex software with a Microtrac S3500 instrument. The data were generated with a mean of triplicate reads for each sample loaded.

GC/MS Characterization of Individual Fatty Acids. The fatty acid methyl esters (FAME) were characterized in the extracted oil samples to determine the individual fatty acid profile by transesterification, according to the AOCS method no.: Ce 1-62.[13] Briefly, 100 mg of oil was mixed with 1 mL of boron trifluoride (BF_3) in methanol and incubated at 60 °C for 30 min in a water bath. Later, the tubes were immediately transferred to an ice bath for 5 min, 1 mL of hexane and 1 mL of distilled water were added, and the tubes were vortexed for complete mixing. After settling for 10 min at room temperature, the upper layer was collected in a tube containing anhydrous sodium sulfate to remove moisture. Furthermore, the undisturbed top layer of the methyl ester layer free of water and residual particles was transferred to GC vials for GC/MS analyses (Agilent technologies, Milan, Italy). Heptadecanoic acid (C17:0) at 1 mg/mL was added as the internal standard. The GC/MS instrument was connected to a 5977 inert mass spectrometer with the GC column, DB-23 (60 m, 0.25 mm, I.D 0.25 mm film thickness) in the splitless mode (0.5 min), the inlet temperature was 250 °C, and helium as the carrier gas at a flow rate of 1 mL/min was used. The temperature was programmed at 10 °C/min to 300 °C and then isothermal at 300 °C for 5 min. The MS detector was operated in the electron ionization (EI) mode (70eV, 200 mA), in the full-scan mode (m/z 40−400), and also in the selected ion monitoring (SIM) mode (ions at m/z 127, 140, and 256 for heptadecanoic acid as the internal standard).

FT-IR Analysis. The FT-IR spectra of the extracted oils were determined to identify the molecular functional groups of the fatty acids (Tensor II, M/s. Bruker, Germany). FT-IR was performed in the mid-infrared range of 400−4000 cm^{-1} with DLATGS (deuterated L-alanine-doped triglycine sulfate). PC-based data acquisition with OPUS software version 7.5 was used for qualitative transmission. The spectra were compared with the reference identification FT-IR functional group interpretation.[19] The presented FT-IR spectra are representative spectra of three spectra collected.

Extraction and Estimation of Ascorbic Acid Using HPLC. A known quantity of the selected samples was homogenized with 2 mL of methanol and 10 mL of cold extraction solution, containing 3% metaphosphoric acid (MPA) (w/v), 0.05% ethylene diamine tetra acetic acid (EDTA) (w/v), and 0.8% glacial acetic acid (v/v).[20] The homogenate was centrifuged at 8000 rpm for 15 min at 4 °C. The chromatographic analysis of the clear homogenate filtered with a membrane filter (0.22 μm) and ascorbic acid was carried out in a LC 20 AD (M/s Shimadzu Corporation, Kyoto, Japan), a Chromatopak C_{18} (150 mm × 4.6 mm i.d., 5 μ particle size) was used for separation, and the mobile phase was 50 mM dipotassium hydrogen phosphate (K_2HPO_4) adjusted to pH 7 (solvent A) and 100% methanol (solvent B) with a flow rate of 1 mL/min. Ascorbic acid was detected at 254 nm.

Sample Extraction for Bioactives and *In Vitro* Antioxidant Analysis. The extraction efficiency of different solvents, namely distilled water, 80% ethanol, and 70% methanol, was analyzed in selected powder samples.[15] Briefly, about 5 g of powder samples was extracted with 50 mL of solvent using a mortar and pestle and centrifuged at 10 000 rpm for 10 min. The collected supernatant was stored in srew cap vials and used for further analysis.

Determination of the Total Phenolic Content (TPC). The total phenolic content (TPC) of the above-prepared extracts was determined using the Folin−Ciocalteu method.[14] The extract (0.1 mL) was pipetted out into a test tube and made up to 3 mL with distilled water. A total of 0.1−1 mL of the working standard (Gallic acid, 0.1 mg/mL) was taken for preparing the standard curve. Next, 0.5 mL of Folin−Ciocalteau reagent was added into each test tube and incubated for 3 min. Later, 2 mL of 20% Na_2CO_3 solution was added to each tube. The tubes were vortexed and placed in a boiling water bath for exactly 1 min. The absorbance was measured at 650 nm after cooling, and the amount of phenolics present in the samples was obtained by plotting against the standard graph. The TPC was expressed as gallic acid equivalent (GAEq.) in mg/100 g dry weight (DW) powder samples.

Estimation of the Total Flavonoid Content (TFC). The TFC of the diluted extracted (1:9) samples of 1 mL was mixed with 1 mL of 2% (w/v) methanolic solution of aluminum chloride ($AlCl_3$) and incubated at room temperature for 15 min. The absorbance of the reaction mixture was read at 430 nm. The results were expressed as rutin equivalent (REq.) in mg/100 g DW powder samples.[21]

HPLC Characterization of Individual Phenolics and Flavonoids. The individual phenolic and flavonoid compounds of the selected samples were extracted with 70% methanol until the macerate becomes colorless and centrifuged to obtain a clear supernatant. The obtained clear filtrate was completely evaporated (Hei-VAP Advantage, Heidolph Instrument GmbH & Co., KG, Schwabach, Germany) to dryness and redissolved in HPLC-grade 70% methanol and filtered through a 0.22 μ filter before HPLC analysis. The separation was performed using a reverse-phase HPLC (Shimadzu LC 20A; Shimadzu Corp.) fitted with a C_{18} column (250 μm × 4.6, 5 μm, Waters Corporation, Milford, MA) and UV detector.[22] A binary solvent system, consisting of filtered Milli-Q water adjusted to pH 2.6 with acetic acid as solvent A and 80% acetonitrile as solvent B, was run for 60 min at a flow rate of 1.2 mL/min. The phenolics and flavonoids were detected at 280 and 320 nm using the standards gallic, protocatechuic, catechin, epicatechin, syringic, para-coumaric, chlorogenic, ferulic, sinapic, and trans-cinnamic acids, rutin, and quercetin.

UHPLC-HRMS/MS Characterization of Major Phenolics and Flavonoids. The UHPLC-HRMS/MS analyses of sample extracts (70% methanol) were performed in a hybrid quadrupole-TOF LC/MS/MS mass spectrometer system (Sciex Triple ToF 5600, Singapore), which was connected to a Kinetex C18 100A (30 mm × 2.1 mm, 1.7 μm) Phenomenex column at a flow rate of 0.4 mL/min. The mobile phase comprises solvent A 0.1% acetic acid in water and solvent B 0.1% acetic acid in acetonitrile and methanol in an 8:2 ratio.[23] The column temperature was maintained at 35 °C, and the mass measurements were taken with a scan range of 200−2000 m/z and an IRDx resolution of 15 000 in the ESI negative

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Table 1. Physicochemical Composition, Color Measurement, and Minerals of the Selected Cereal, Pulse, Unripe Banana, and Arrowroot Samples[a]

parameter (g/100 g DW)	barley	finger millet	chickpea	green gram	horse gram	unripe banana	arrowroot
moisture (%)	1.63 ± 0.09	1.89 ± 0.03	1.45 ± 0.04	4.99 ± 0.05	4.61 ± 0.09	3.03 ± 0.04	7.68 ± 0.15
total carbohydrates	46.10 ± 2.21	59.26 ± 1.81	45.20 ± 2.85	56.52 ± 2.09	56.11 ± 1.18	67.65 ± 2.76	63.76 ± 1.98
total reducing sugars	1.34 ± 0.02	1.66 ± 0.04	1.73 ± 0.14	1.93 ± 0.06	1.98 ± 0.21	1.47 ± 0.14	1.47 ± 0.10
total nonreducing sugars*	44.76 ± 2.20	57.60 ± 1.84	43.47 ± 2.79	54.58 ± 2.11	54.13 ± 1.35	66.18 ± 2.86	62.29 ± 1.87
total protein content	1.32 ± 0.06	0.67 ± 0.07	16.92 ± 0.07	6.21 ± 0.21	11.12 ± 0.43	3.14 ± 0.15	0.01 ± 0.00
total oil content (%)	1.94 ± 0.06	1.69 ± 0.07	7.79 ± 0.06	2.15 ± 0.07	1.27 ± 0.02	0.64 ± 0.02	0.09 ± 0.00
ash content	2.41 ± 0.05	3.47 ± 0.01	2.26 ± 0.06	4.53 ± 0.08	4.38 ± 0.08	3.06 ± 0.23	0.87 ± 0.04
crude fiber	2.69 ± 0.24	4.30 ± 0.20	4.69 ± 0.31	1.98 ± 0.14	5.66 ± 0.43	3.08 ± 0.17	2.31 ± 0.15
calorific value (J/100 g)	207.07 ± 8.24	254.87 ± 7.47	318.64 ± 11.52	270.12 ± 8.76	280.28 ± 4.77	288.91 ± 11.67	255.87 ± 7.88
Color Measurement							
L	83.15 ± 0.08	84.77 ± 0.20	87.32 ± 0.18	76.53 ± 0.48	69.77 ± 0.40	78.05 ± 0.14	94.31 ± 0.19
*a**	1.19 ± 0.04	2.56 ± 0.07	1.24 ± 0.03	2.28 ± 0.10	3.34 ± 0.10	0.72 ± 0.02	−0.02 ± 0.01
*b**	13.18 ± 0.05	22.75 ± 0.14	9.43 ± 0.11	12.96 ± 0.06	8.40 ± 0.09	17.42 ± 0.10	4.24 ± 0.05
chroma	18.63 ± 0.07	32.18 ± 0.20	13.34 ± 0.15	18.32 ± 0.08	11.88 ± 0.12	24.64 ± 0.14	6.00 ± 0.07
hue	1.48 ± 0.00	1.46 ± 0.00	1.44 ± 0.00	1.40 ± 0.01	1.19 ± 0.01	1.53 ± 0.00	−1.57 ± 0.00
Minerals (mg/100 g DW)							
iron	15.87 ± 0.81	14.55 ± 0.72	12.61 ± 0.21	24.44 ± 1.44	12.07 ± 0.13	9.99 ± 0.18	10.84 ± 0.31
zinc	4.00 ± 0.14	11.00 ± 0.40	0.97 ± 0.03	10.71 ± 0.56	0.13 ± 0.02	9.08 ± 0.09	9.06 ± 0.08
calcium	980.06 ± 16.07	1006.83 ± 10.06	765.91 ± 12.63	857.31 ± 13.17	918.27 ± 14.58	801.28 ± 11.17	708.04 ± 11.99
copper	1.34 ± 0.07	3.28 ± 0.09	1.19 ± 0.02	0.76 ± 0.01	0.75 ± 0.07	2.94 ± 0.07	3.89 ± 0.04
magnesium	35.43 ± 0.49	73.12 ± 6.90	52.44 ± 0.57	83.45 ± 2.19	78.66 ± 0.08	40.22 ± 0.59	3.14 ± 0.20
potassium	134.09 ± 0.81	165.85 ± 8.50	346.38 ± 7.10	531.81 ± 7.57	502.78 ± 0.14	422.81 ± 4.88	11.14 ± 0.54
sodium	215.67 ± 18.94	192.47 ± 14.84	337.90 ± 3.48	226.93 ± 3.61	231.40 ± 14.71	190.69 ± 6.72	255.92 ± 18.13

[a]All the values are mean ± SD of three replicates (n = 3) analyzed. * Values are obtained by subtracting the total carbohydrate value with total reducing sugars.

mode. GS1-45, GS2-60, and Curtain GAS (CUR)-40 were used as gases. The duospray ion source was set with an ion spray voltage floating (ISVF) of 4500 at 400 °C. The cycle time was 700 ms, the accumulation time was 250.0 ms, and the accumulation time was 30 min. The information-dependent acquisition (IDA) procedure employed the most intense ion with spread energy at 20% and normal collision energy at 45%. The mass measurements were recorded with a scan range of 200−2000 m/z in the ESI negative mode. The data were analyzed using Peak View 2.1 software (AB SCIEX Triple TOF 5600, Singapore) and equipped with MasterViewTM (Version 1.0, AB SCIEX). The XIC manager tool in Master View was used to detect quasi-molecular weights, mass errors, and isotope patterns of both nontargeted and targeted compounds.

Antioxidant Assays (AA). Determination of the Total Antioxidant Assay (TAA) by the Phosphomolybdenum Method. The TAA of sample extracts (0.3 mL) was mixed with 28 mm of sodium phosphate and 4 mm of ammonium molybdate. The reaction mixture was incubated at 95 °C for 90 min, and the absorbance of the reaction mixture was read at 695 nm using a double-beam spectrophotometer (UV-160 A, Shimadzu Corporation, Kyoto, Japan) against blank after cooling to room temperature.[24] The TAA was expressed as the number of gram equivalent of ascorbic acid (AA) in mg/100 g DW powder samples.

Ferric Reducing Antioxidant Power (FRAP). The FRAP analysis was performed according to the method reported earlier.[25] The sample extracts in different dilutions of 1 mL were mixed with 2.5 mL of phosphate buffer (0.2 M, pH 6.6) and 2.5 mL of potassium ferricyanide [$K_3Fe(CN)_6$] (1% (w/v)). The mixture was incubated for 30 min at 50 °C. Later, 2.5 mL of 10% trichloroacetic acid (TCA) was added to this mixture and centrifuged at 3000 rpm for 10 min. The upper layer (2.5 mL) was mixed with 2.5 mL of distilled water and 0.5 mL of 0.1% ferric chloride. Finally, the absorbance of the reaction mixture in triplicates was measured at 700 nm. The FRAP was expressed as the number of gram equivalent of ascorbic acid (AA) in mg/100 g DW powder samples.

DPPH Free Radical Scavenging Activity. The DPPH radical scavenging potentials of the extracted samples were performed following the method reported earlier.[26] To different dilutions of sample extracts (1 mL), 1 mL of the DPPH reagent (3.94 mg/100 mL methanol) was added to make up the final volume to 2 mL. The reaction mixture was thoroughly mixed and incubated for 15 min in the dark at room temperature. The absorbance of the reaction mixture was recorded at 517 nm with methanol as the blank.

From the absorbance, % inhibition or % scavenging activity is calculated using this formula,

$$\text{DPPH scavenging activity} = \frac{(\text{OD control} - \text{OD sample})}{\text{OD control}} \times 100$$

ABTS Scavenging Activity Assay. Trolox equivalent antioxidant capacity (TEAC) assay measures the ability of a compound to eliminate or scavenge radicals compared with trolox as an antioxidant.[27] For this assay, 13.2 mg of potassium persulfate was mixed with 20 mL of 7.4 mM ABTS solution (in water) for about 16 h at room temperature in the dark to yield a dark blue−green solution. The solution was diluted with phosphate buffered saline (PBS) (pH 7.4) to attain an

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absorbance value of 0.70 at 734 nm and used for the antioxidant assay within 4 h. The sample extractives at different concentrations were mixed with 2 mL of the diluted ABTS solution for 6 min, followed by the measurement of light absorbance at 734 nm. The radical scavenging activity was represented by the percentage of ABTS scavenging activities of the oil extractives.

Statistical Analysis. All the data were presented in the form of mean ± S.D of three replicates. One-way ANOVA was performed by post hoc Duncan's multiple-range test (DMRT) using SPSS 17 (SPSS Inc., Chicago, IL) for determining significance at $p < 0.05$. The EC_{50} values were calculated using the nonlinear regression.

RESULTS AND DISCUSSION

Determination of the Moisture Content. The selected cereal and pulse samples were analyzed for their moisture content (Table 1). Arrowroot (7.68%) contains a high amount of moisture followed by green gram (4.99%) and horse gram (4.61%). The least moisture percentage was recorded in chickpea, that is, 1.45%, wherein unripe banana, finger millet, and barley contain 3.03, 1.89, and 1.63% of moisture, respectively. The variation in the moisture content of these selected samples depends on the cultivation and storage conditions of the samples.[28] A similar moisture content among five varieties of arrowroot at 5.02−6.53% has been reported.[29]

Nutritional Composition. Table 1 shows the nutritional composition of the selected samples analyzed for their total carbohydrate, total protein, total oil, and crude fiber contents. The total carbohydrate content was high in unripe banana (67.65/100 g) and arrowroot (63.76/100 g) followed by finger millet (59.26/100 g), green gram (56.52/100 g), and horse gram (56.11/100 g). The reducing sugar contents were high in horse gram (1.98/100 g) and green gram (1.93/100 g); however, barley had the least reducing sugars (1.33/100 g). Similarly, the total nonreducing sugars also showed the same trend as total carbohydrates. The total protein content was very high in chickpea (16.92/100 g) and horse gram (11.12/100 g) compared to other samples. The least total protein content was observed in the arrowroot sample (0.01/100 g). Chickpea (7.79%) had a very high content of total oil compared to the other samples. The lowest oil contents were observed in arrowroot (0.09%) and unripe banana (0.64%). Variations in the proximate composition of banana during the four ripening stages and recorded higher carbohydrate content (82.34/100 g) than the present values have been reported.[30] This change in the proximate composition indicates the progress of biochemical reactions and the existence of a climacteric peak between those stages, which is directly linked to ethylene production. Recently, the total lipid content in different varieties of horse gram was analyzed, ranging from 0.91 to 2.29/100 g, which correlated with the present data.[31] The proximate composition of finger millet and horse gram which correlated with the present results has been reported.[32] The reported results of the proximate composition of chickpea and horse gram correlated with the currently analyzed data.[33]

Similarly, the ash content and crude fiber were also analyzed for the studied samples. The highest ash content was recorded in green gram (4.53/100 g) and horse gram (4.38/100 g). However, the least was observed in the arrowroot sample (0.87/100 g). The crude fiber content of the studied samples ranged from 1.98 to 5.66/100 g, wherein the highest was recorded in horse gram and the least was observed in green gram (Table 1). The calorific value of the samples was calculated based on the nutritional composition. Among all, the chickpea (318.64 J/100 g) showed high calorific values, followed by unripe banana (288.91 J/100 g) and arrowroot (255.87 J/100 g). However, the barley sample recorded the least calorific value (207.07 J/100 g) among all the samples selected. Variations in the proximate composition of banana during the ripening stages and the recorded higher ash content (2.10/100 g) than the present values have been reported.[30]

Color Measurement. The color is the basic and first appearance property of the edible samples to be consumed by human beings. The powders of the selected cereal, pulse, unripe banana, and arrowroot samples were subjected to color measurement (Table 1). The arrowroot sample showed a higher (94.31) L value that explains the high brightness or more whiteness of the sample. The lowest L value was observed in horse gram (69.77), indicating the low brightness of the sample. The a^* values were high in horse gram (3.34) and least in arrowroot (−0.02) samples. These a^* values indicate that the horse gram sample contains a reddish/brown color in powder, whereas the arrowroot sample is colorless. The b^* value explains about the blue to yellow chroma; in the studied samples, higher b^* values were observed in finger millet (22.75) with the lowest in arrowroot (4.24) samples. Similarly, the chroma and hue values were high in finger millet and unripe banana samples. The reported variations in color values in chickpea and the L and b^* of the present values of chickpea were correlated.[34] The arrowroot color measurement values were correlated with the color values recorded in five different varieties of arrowroot flours.[29] Overall, the changes in color measurement values could be solely dependent on the enzymatic reactions during developmental changes.[30]

Minerals. Minerals are the most important micronutrients that play a key role in various metabolic processes. In the studied samples, we found some of the major minerals such as iron, zinc, calcium, copper, magnesium, potassium, and sodium (Table 1). The green gram contains high amounts of iron, magnesium, and potassium of 24.22, 83.45, and 531.81 mg/100 g DW, respectively. Zinc (11 mg/100 g DW) and calcium (1006 mg/100 g DW) were observed in high amounts in finger millet. The chickpea contains a high amount of sodium (337.9 mg/100 g DW), and finally, the arrowroot contains a high amount of copper (3.89 mg/100 g DW). Ramashia et al. reported similar mineral profiles in finger millets with an iron content of 14.89 mg/100 g, wherein we recorded 14.55 mg/100 g.[35] Pal et al. reported comparable values pertaining to horse gram varieties for iron, zinc, and calcium contents.[31]

Determination of Chlorophyll and Carotenoids. Table S1 shows the chlorophyll and carotenoids analyzed by Lichtenthaler and Arnon methods, which are primary components in plant-based samples. The total chlorophyll content was high in unripe banana (2.72, 3.07 mg/100 g DW) and least in arrowroot samples (0.20, 0.22 mg/100 g DW). Among all, chlorophyll *a* (chl *a*) was higher in the case of green gram (1.16, 1.28 mg/100 g DW) and chlorophyll *b* (chl *b*) was higher in the unripe banana sample (1.73, 1.93 mg/100 g DW). The highest total carotenoid content was recorded in chickpea (670.48 mg/100 g DW) with the lowest in arrowroot (23.97 mg/100 g DW). When compared to Lichtenthaler, the Arnon method showed a higher chlorophyll content (10−12%). A similar comparative study of the chlorophyll and carotenoid contents in green leafy vegetables, *i.e.*, *Hibuscus sabdariffa* dried using different methods, was conducted, and a

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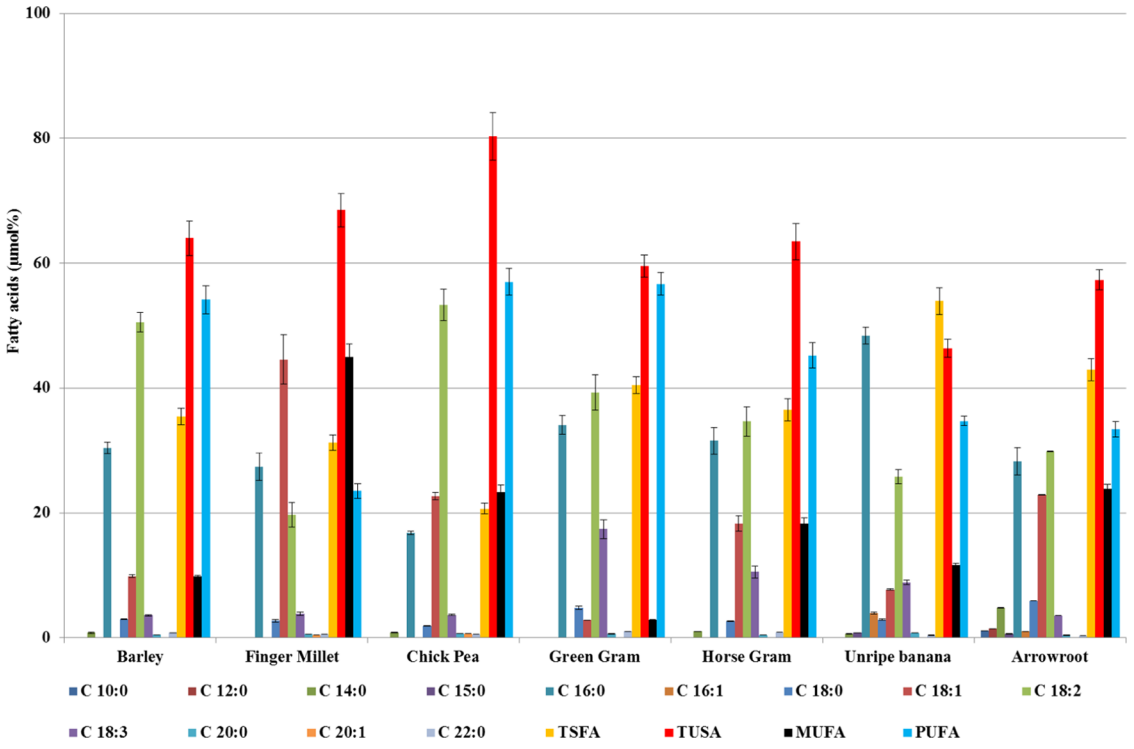

Figure 1. Fatty acid composition quantified using GC/MS of the selected cereal, pulse, unripe banana, and arrowroot samples.

12% difference in the pigment content quantified using two methods was found.[36]

Particle Size Distribution. A particle size analyzer can measure the dimensions of solid particles present in the samples (Table S2). Among the studied samples, the green gram showed the highest particle size of 368 nm with 69.6% volume, followed by the horse gram at 349.9 nm with 100% volume. The lowest size was observed in the arrowroot sample at 14.25 nm with a volume of 31.7%. This difference in particle size may be due to the milling process. It will mainly plays a key role in the development of products and water absorption. Furthermore, this plays a main role in maintaining the quality of the product (dough).[18]

GC/MS Characterization of Individual Fatty Acids. GC/MS analysis was performed to study the individual fatty acids present in the lipid content of the studied samples (Figure 1). The fatty acids were observed in the range of C 10:0−C 22:0. Palmitic acid (C 16:0) is a saturated fatty acid (SFA) observed in high amounts in the unripe banana (47.42 μmol %) sample followed by green gram (34.04 μmol %) and horse gram (31.56 μmol %). The USFAs such as oleic, linoleic, and linolenic acids are present in high amounts in all the studied samples. Among the studied samples, finger millet (44.55 μmol %), chickpea (53.33 μmol %), and green gram (17.40 μmol %) showed high oleic, linoleic, and linolenic amounts, respectively. Compared to SFAs, the contents of USFAs are high in all the samples. Hence, the present samples can act as a source of USFAs. The total saturated fatty acids were high in the unripe banana sample, and the total unsaturated fatty acids were high in the chickpea sample. However, the mono- and polyunsaturated fatty acids were high in finger millet and green pea, respectively. Ramashia et al. reported that finger millets contain high amounts of oleic acid (49.80 mg/100 g) and linoleic acid (24.20 mg/100 g).[35]

FT-IR Analysis. FT-IR analysis explains about the functional groups present in the extracted oil samples. The FT-IR transmission spectra of the visible and near-infrared fingerprints are shown in Figure S2. The spectra showed the peaks at 722 and 1093 interpreted by overlapping of the methylene rocking vibrations and attributed to the out-of-plane vibration of *cis*-distribution olefins and stretching C−O−C of the ether group, respectively. Similarly, the peaks at 1118 and 1744 represent the TAG-derived secondary alcohol and C═O stretching mode of lipids. The asymmetric CH_2 stretching modes of the methylene chains in membrane lipids, CH_2 acyl chains, and −C═CH *cis*-double bond stretching at peaks 2854, 2923, and 3009 were correlated with the MUFA content of the oil samples. This oil fatty acid functional group molecular fingerprinting was correlated completely with the *Basella rubra* seed oil FT-IR interpretation.[19]

Estimation of Ascorbic Acid Using HPLC. Ascorbic acid is an important antioxidant compound with high radical scavenging activity, which helps in regulating blood pressure and lowering and removal of cholesterol in arterial walls to prevent arteriosclerosis. The ascorbic acid content of the selected samples was analyzed using HPLC (Figure 2a). The ascorbic acid content was the highest in the chickpea, horse gram, and unripe banana samples at 86.83, 83.76, and 81.88 mg/100 g DW, respectively, whereas, a low content was observed in the green gram (14.43 mg/100 g DW) sample. The finger millet, arrowroot, and barley showed 56.46, 36.92, and 16.13 mg/100 g DW, respectively. The contents of ascorbic acid in the selected samples were comparatively high compared to the reported values, as the ascorbic acid content depends on the soil nutrition, processing conditions, and storage.[35,37] Similarly, variation in the ascorbic acid content in *Talinum triangulare* dried foliage has been recently reported.[15]

Determination of TPCs. Phenolic compounds are plant secondary metabolites with aromatic ring structures that are

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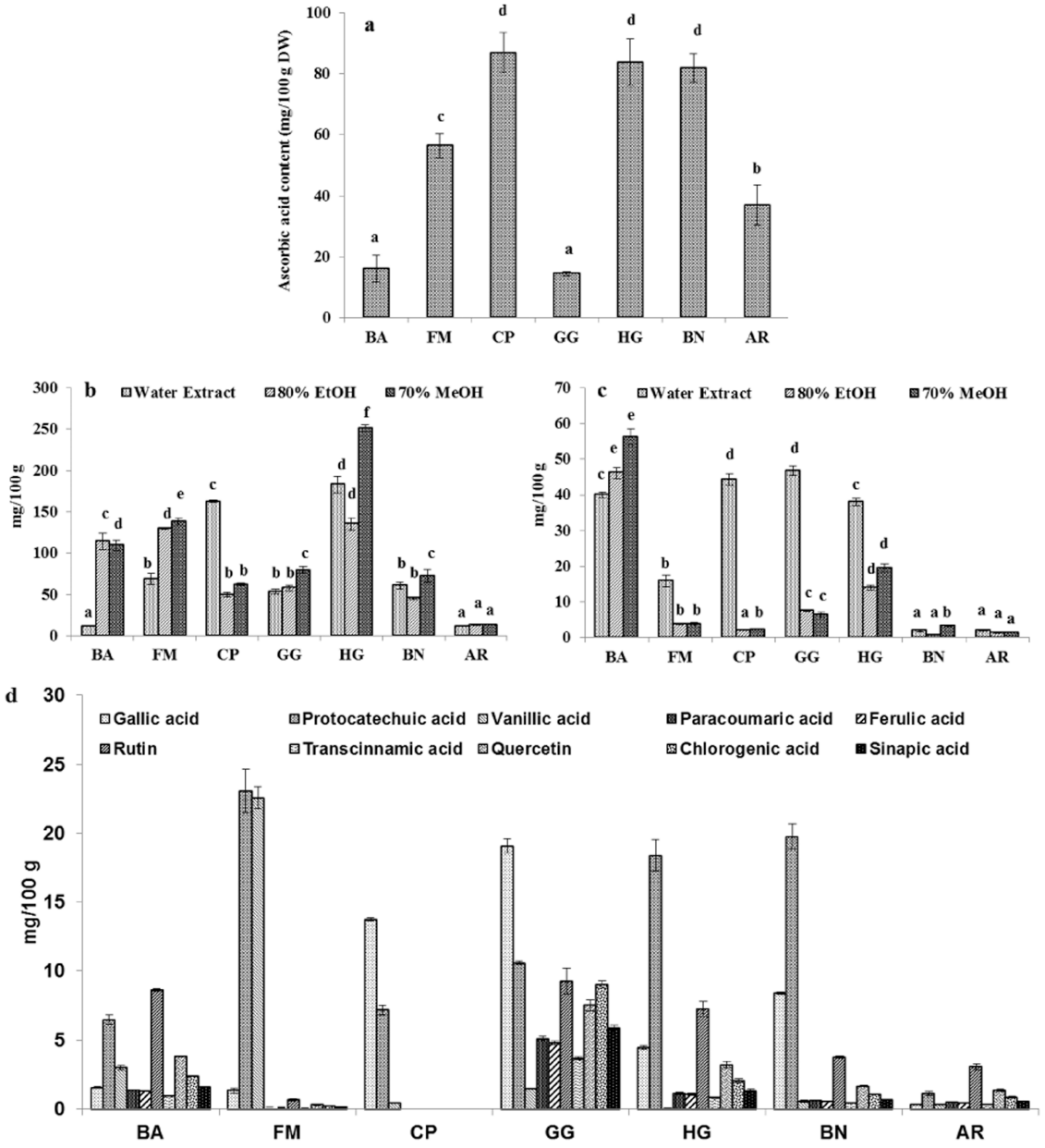

Figure 2. (a) Ascorbic acid content quantified using HPLC, (b) TPC, and (c) TFC and (d) phenolic and flavonoid compounds quantified using HPLC. All the values are mean ± SD of three replicates, and the values with different superscripts for each parameter were significantly different from each other ($p < 0.05$). BA—barley, FM—finger millet, CP—chickpea, GG—green gram, HG—horse gram, BN—banana, and AR—arrowroot.

widely distributed in the plant kingdom and linked with the color, sensory qualities, and antioxidant activities of the foods. The redox potentials of the phenolic compounds are the key factors for their antioxidant properties. The TPCs of the studied samples were analyzed using a spectrophotometric method in three different extracts (distilled water, 80% ethanol, and 70% methanol) to identify the maximum extractability of the antioxidant compounds in different solvents. The extracted horse gram sample using distilled water (183.26 mg/100 g DW), 80% ethanol (135.41 mg/100 g DW), and 70% methanol (251.14 mg/100 g DW) showed the highest TPC followed by finger millet (Figure 2b). The present TPC contents in finger millet extracts were correlated with the four varieties of finger millets reported.[38] However, the lowest TPC was observed in arrowroot extracts with 11.94, 13.31, and 13.46 mg/100 g DW for water, 80% ethanol, and 70% methanol extracts, respectively. Jogihalli et al. reported a higher TPC content (802 mg/100 g) in chickpea compared to the present values (162 mg/100 g), which depends on the extracting solvent, area of cultivation, soil nutritional conditions, genetic factors, farming practice, post-harvest handling, processing, and storage conditions.[34] The TPC content in unripe banana was recorded to be higher compared to the reported values, at 29.68 mg/100 g.[30]

Estimation of TFC. The TFC of the studied samples was analyzed by a spectrophotometric method. The distilled water−green gram extract (46.69 mg/100 g DW) showed a high TFC (Figure 2c). Similarly, the barley in 70% methanol (56.21 mg/100 g DW) and 80% ethanol (46.2 mg/100 g DW) extracts showed a high content of TFC. However, the lowest

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Table 2. Identification of Major Phenolic and Flavonoid Compounds with MS/MS Fragment Ions of the Selected Cereal, Pulse, Unripe Banana, and Arrowroot Samples[a]

compound name	empirical formula	$[M-H]^-$	MS/MS fragment ion	barley	finger millet	chickpea	green gram	horse gram	unripe banana	arrowroot
quercetin galloyl glucoside	$C_{28}H_{23}O_{16}$	615	329, 217, 144	−	−	−	−	−	−	+
rutin	$C_{27}H_{29}O_{16}$	609	300, 271, 255	+	+	−	+	+	+	−
jasmonyl dicitrate	$C_{24}H_{34}O_{17}$	593	547, 325	−	−	+	−	+	+	−
isoquercitrin	$C_{21}H_{20}O_{12}$	463	300, 301, 271	−	+	+	+	+	−	−
epicatechin glucoside	$C_{21}H_{24}O_{11}$	451	289, 245, 203	−	+	+	−	+	−	+
quercetin rhamnoside	$C_{21}H_{19}O_{11}$	447	152, 108	−	−	+	−	+	−	−
quercetin-3-xyloside	$C_{20}H_{17}O_{11}$	433	279, 153	+	−	+	+	+	+	+
chlorogenic acid	$C_{16}H_{18}O_9$	353	183, 167	+	+	−	+	+	+	+
p-coumarylquinic acid	$C_{16}H_{18}O_8$	337	122, 96	−	+	−	−	−	+	−
vanillic acid hexoside	$C_{14}H_{18}O_9$	329	211, 193, 167	+	+	+	−	+	+	+
coumaric acid hexoside	$C_{15}H_{12}O_9$	325	183, 119	+	−	+	+	+	+	+
epigallocatechin	$C_{15}H_{14}O_7$	305	137, 125, 109	−	−	−	−	+	−	−
epicatechin	$C_{15}H_{14}O_6$	289	245, 221, 125	+	+	−	+	+	−	−
apigenin	$C_{15}H_{10}O_3$	269	225, 156	−	−	+	−	−	+	+
sinapic acid	$C_{11}H_{12}O_5$	223	179, 133	−	−	−	−	+	−	−
jasmonic acid	$C_{12}H_{18}O_3$	209	165, 141	−	−	−	−	−	−	+
syringic acid	$C_9H_{10}O_5$	197	169, 150	−	−	−	−	−	−	+
ferulic acid	$C_{10}H_{10}O_4$	193	149, 134, 133	−	+	−	−	+	−	+
citric acid	$C_6H_8O_7$	191	173, 129, 111	+	+	+	+	+	+	+
hydroxy gallic acid	$C_7H_6O_5$	187	169, 125	+	+	+	−	−	+	+
ethyl protocatechuate	$C_9H_{10}O_4$	181	163, 135	−	+	−	−	−	−	+
gallic acid	$C_7H_6O_5$	169	125, 71	−	+	−	−	+	−	+
vanillic acid	$C_8H_8O_4$	167	123, 108	−	+	−	−	+	−	+
O-coumaric acid	$C_9H_8O_3$	163	119	−	−	−	−	+	−	−
protocatechuic acid	$C_7H_6O_4$	153	108, 81	−	+	+	−	−	−	−
trans-cinnamic acid	$C_9H_8O_2$	147	102, 87	−	+	+	−	+	−	−

[a](+ indicates detected, and − indicates not detected).

TFC in all the three different extracts was observed in the arrowroot at 2.00, 1.25, and 1.36 mg/100 g. A higher flavonoid content (44 mg/100 g) in chickpea extracts was recorded in the present study, comparable with the values reported in aqueous methanol acid extracts (23 mg/100 g).[39] Similarly, the finger millet TFC content of the present study was slightly low compared to the values reported.[39]

HPLC Characterization of Individual Phenolics and Flavonoids. The HPLC separation of individual phenolics and flavonoids in the selected samples is shown in Figure 2d. The green gram showed majorly all the phenolic and flavonoid compounds in higher quantities compared to other samples. The major phenolics and flavonoids identified and quantified in all the samples were gallic, protocatechuic, vanillic, para-coumaric, ferulic, chlorogenic, sinapic, and trans-cinnamic acids, rutin, and quercetin. A similar trend of phenolics compounds was characterized in finger millets, and the quantity was in correlation with the present study.[38]

UHPLC-HRMS/MS Characterization of Major Phenolics and Flavonoids. Table 2 shows the major phenolic and flavonoid compounds identified in the selected cereal, pulse, unripe banana, and arrowroot samples were represented. A total of 25 compounds were detected that were confirmed with the MS/MS fragmented ion patterns. The electrospray ionization (ESI)-MS signals that gave deprotonated ions m/z 609, m/z 463, m/z 353, m/z 223, m/z 193, m/z 191, and m/z 147 were identified as rutin, isoquercitrin, chlorogenic acid, sinapic acid, ferulic acid, citric acid, and trans-cinnamic acid, respectively. Similar fragmentation of phenolics and flavonoids using UHPLC-HRMS/MS was performed in Kainth fruit and *Pterocarpus santalinus* hydroalchoholic extracts.[23,40]

Antioxidant Assays (AA). Determination of the Total Antioxidant Assay by the Phosphomolybdenum Method. All the selected sample extracts were used for the determination of phosphomolybdenum complexes by the samples to know the antioxidant capacities (Figure 3a). The method is mainly based on the reduction of Mo(VI) to Mo(V) by antioxidant compounds, which in turn form a green color complex that has absorbance maxima at 695 nm. The samples were extracted with three different solvents such as distilled water, 80% ethanol, and 70% methanol to know which solvent can extract the maximum amount of antioxidant compounds from the samples. The horse gram showed the highest antioxidant activity in all the three different extracts prepared at 725.16, 509.29, and 481.77 mg/100 g. Similarly, the lowest activity was shown by the three arrowroot sample extracts. This high antioxidant activity of the horse gram has also been correlated to the reported values in 12 different samples tested ranging from 526 to 676 μM TE/g.[31] The antioxidant activity of the samples is attributed to the phenolic and flavonoid contents in the present study. Similarly, correlation of the phenolic and flavonoid contents in *T. triangulare* dried foliage to antioxidant activity has been recently reported.[15]

Ferric Reducing Antioxidant Power (FRAP). The FRAP assay is one of the most sensitive methods to check the antioxidant activity of the samples to reduce ferric ions. The three horse gram sample extracts (151.51, 149.07, and 270.08 mg/100 g of distilled water, 80% ethanol, and 70% methanol extracts, respectively) showed the highest antioxidant activity

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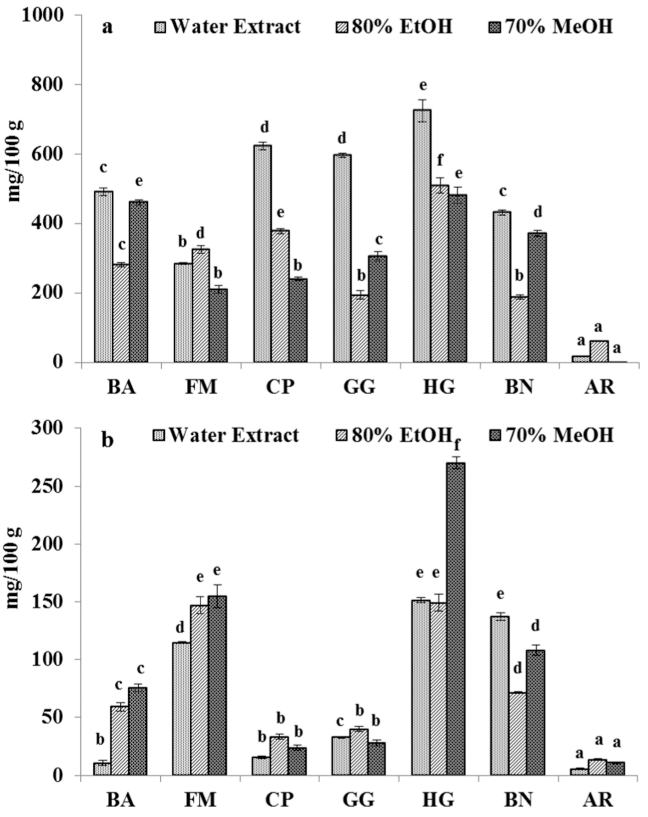

Figure 3. (a) TAA and FRAP activity of the selected cereals, pulses, unripe banana, and arrowroot samples. All the values are mean ± SD of three replicates, and the values with different superscripts for each parameter were significantly different from each other ($p < 0.05$). BA—barley, FM—finger millet, CP—chickpea, GG—green gram, HG—horse gram, BN—banana, and AR—arrowroot.

followed by the finger millet extracts (Figure 3b). The lowest activity was observed in the arrowroot sample in distilled water, 80% ethanol, and 70% methanol extracts with 5.55, 13.64, and 10.92 mg/100 g, respectively. Parikh and Patel reported a higher FRAP activity compared with the present study in chickpea (238 mg/100 g), green gram (209 mg/100 g), and horse gram (822 mg/100 g) samples.[41] These higher values are due to the extraction process, and the results were expressed as trolox equivalents.[41]

DPPH Free Radical Scavenging Activity. DPPH is one of the widely used rapid and reproducible primary analyses to know the antioxidant activity of samples. The reduction of DPPH free radicals by the various extracts in different solvents could reduce the color of DPPH to colorless based on the content of antioxidant capacity of the extracts, which will be read at 517 nm. Among the studied samples, the distilled water (EC_{50} = 1.23 mg/mL) and 70% methanolic extracts (EC_{50} = 4.22 mg/mL) of horse gram and the 80% ethanolic extract (EC_{50} = 1.62 mg/mL) of finger millet showed the highest antioxidant activity compared to other samples (Table 3). Similarly, the lower activity was showed by the distilled water extract (EC_{50} = 35.5 mg/mL) of barley and 70% methanol (EC_{50} = 9.85 mg/mL) and 80% ethanolic extracts (EC_{50} = 9.96 mg/mL) of the arrowroot sample. The chickpea extract showed a higher (EC_{50} = 6.67 mg/mL) DPPH radical scavenging activity compared to the earlier reports.[34] Parikh and Patel reported the DPPH radical scavenging activity of chickpea (72.9 mg/100 g), green gram (186 mg/100 g), and horse gram (112 mg/100 g) samples.[41]

ABTS Radical Scavenging Activity Assay. The ABTS is one of the most important activities to analyze the radical scavenging activity of samples. Among all the studied samples, the three horse gram extracts such as distilled water (EC_{50} = 0.31 mg/mL), 70% methanol (EC_{50} = 0.49 mg/mL), and 80% ethanol (EC_{50} = 0.39 mg/mL) extracts showed the highest radical scavenging activity (Table 3). The arrowroot showed the lowest EC_{50} ABTS radical scavenging activity of 2.10, 3.32, and 4.7 mg/mL in 70% methanol, 80% ethanol, and distilled water extracts, respectively. Parikh and Patel reported the ABTS radical scavenging activity of chickpea (20.5 mg/100 g), green gram (12 mg/100 g), and horse gram (156 mg/100 g) samples.[41]

CONCLUSIONS

The frequently consumed cereals, pulses, unripe banana, and arrowroot were investigated for their nutritional composition, bioactivity profiling, and antioxidant activities. In view of their regular consumption in our daily diet, they are worthy to use in the development of functional food formulations. They possess nutritionally rich physicochemical and mineral contents, which is an asset of these selected samples for consumption as an affordable and sustainable alternative source. Both distilled water and aqueous ethanol extracts exhibited good bioactive profiles and antioxidant activities, which possess high free radical scavenging potentials. Hence, it is of merit to provide comprehensive research data to identify nutritional and nutraceutical potentials of these samples for the development of value-added ready-to-eat functional food formulations.

Table 3. DPPH and ABTS Radical Scavenging Activity of the Selected Cereal, Pulse, Unripe Banana, and Arrowroot Samples (EC_{50} = mg/mL)[a]

	DPPH (EC_{50} = mg/mL)			ABTS (EC_{50} = mg/mL)		
sample	water extract	80% ethanol	70% methanol	water extract	80% ethanol	70% methanol
barley	35.50 ± 0.50	6.27 ± 0.13	9.35 ± 0.05	2.07 ± 0.05	0.78 ± 0.04	0.69 ± 0.03
finger millet	4.40 ± 0.10	1.62 ± 0.03	1.77 ± 0.04	0.36 ± 0.01	0.42 ± 0.01	0.62 ± 0.01
chickpea	6.67 ± 0.12	9.85 ± 0.05	9.80 ± 0.06	0.50 ± 0.01	1.79 ± 0.06	0.88 ± 0.03
green gram	8.73 ± 0.21	9.77 ± 0.03	7.57 ± 0.25	0.63 ± 0.03	1.26 ± 0.01	1.04 ± 0.03
horse gram	4.22 ± 0.03	4.15 ± 0.05	1.23 ± 0.06	0.31 ± 0.01	0.39 ± 0.02	0.49 ± 0.01
unripe banana	7.57 ± 0.15	8.32 ± 0.14	4.43 ± 0.21	0.87 ± 0.02	1.24 ± 0.01	0.89 ± 0.03
arrowroot	6.77 ± 0.06	9.96 ± 0.01	9.85 ± 0.05	4.70 ± 0.11	3.32 ± 0.21	2.10 ± 0.09

[a]All the values are mean ± SD of three replicates (n = 3) analyzed.

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■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c03767.

Comparison of the chlorophyll content using two methods in the selected cereal, pulse, unripe banana, and arrowroot samples (mg/100 g DW), particle size and volume distribution of the selected cereal, pulse, unripe banana, and arrowroot samples, and FT-IR molecular fingerprinting of the fatty acid functional group identification (PDF)

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Author Contributions

The authors H.C.C. and S.S.K. planned, designed, and performed the experiments and interpreted the research data. H.C.C. and S.S.K. performed data analysis and statistical analysis and drafted the article sections. S.J. and P.M. fully supervised the study plan, methodology, data execution, interpretation, and article corrections. All the authors read and approved the final article.

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Notes

The authors declare no competing financial interest.

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