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Flavor and Antioxidant Capacity of Peanut Paste and Peanut Butter Supplemented with Peanut Skins

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Abstract: Peanut skins (PS) are a good source of phenolic compounds. This study evaluated antioxidant properties and flavor of peanut paste and peanut butter enhanced with peanut skins. PS were added to both materials in concentrations of 0.0%, 0.5%, 1.0%, 5.0%, 10.0%, 15.0%, and 20.0% (w/w). PS, peanut paste, and peanut butter used in the study had initial total phenolics contents of 158, 12.9, and 14.1 mg GAE/g, respectively. Hydrophilic oxygen radical absorbance capacity (H-ORAC) of peanut skins was 189453 μMol Trolox/100 g and addition of 5% PS increased H-ORAC of peanut paste and peanut butter by 52% to 63%. Descriptive sensory analysis indicated that the addition of 1% PS did not change intensity of descriptors in the sensory profile of either peanut paste or peanut butter. Addition of 5% PS resulted in significant differences in woody, hulls, skins; bitter; and astringent descriptors and 10% PS addition resulted in significant differences in most attributes toward more negative flavor.

Keywords: antioxidants, descriptive sensory analysis, peanut, peanut butter, peanut skins

Practical Application: Peanut skins are a low-value residue material from peanut processing which contain naturally occurring phenolic compounds. The use of this material to improve antioxidant capacity and shelf-life of foods can add value to the material and improve the nutritional value of foods. The improved nutritional qualities and unchanged flavor profile occurring with low levels of peanuts skins in peanut paste and peanut butter suggest potential application of this technology in various food industries.

Introduction

Peanuts are an important crop in many parts of the world. Recent data suggest that production of peanuts in the United States is about 2 million tons (USDA 2011). Peanut skins (testae or seed coat), comprising about 3.0% (w/w) of a peanut seed, are low-value, residue materials resulting from peanut blanching and roasting. Removal of the skin is normally done in preparation for the production of products such as peanut butter. Approximately 60000 tons of peanut skins are accumulated annually in the United States as a result of peanut processing. Peanut skin use is generally limited to animal feeds (Nepote and others 2004; Ha and others 2007). The potential exists for value added use of this material to improve antioxidant capacity and shelf-life of lipid-containing foods.

The concentration of peanut skin tannins from 6 varieties of peanuts ranged from 289 to 468 mg/g (Sanders 1979). Karchesy and Hemingway (1986) reported 17% (w/w) procyanidins in peanuts skins. Lou and others (1999) identified 6 A-type procyanidins from the water-soluble fraction of peanut skin extracts. Procyanidins and other phenolic compounds may provide protection against oxidative stress, which has been implicated in atherosclerosis, diabetes mellitus, chronic inflammation, and some types of cancers in humans (Karadag and others 2009). Procyanidins were reported to have an antihyperglycemic effect in rats with induced diabetes (Pinent and others 2004; El-Alfy and others 2005). Plasma cholesterol levels were reduced in rats fed a diet containing procyanidins (Osakabe and Yamagishi 2009; Shimizu-Ibuka and others 2009). Further, Frankel (1998) reported that phenolic compounds may reduce lipid oxidation in lipid-containing foods. O'Keefe and Wang (2006) evaluated the effect of extracts from peanut skins on the storage stability of ground beef (250 g), and found that 200 to 400 ppm was the optimal concentration of extract to reduce lipid oxidation. Nepote and others (2004) observed that the addition of peanut skin extracts to honey roasted peanuts provided some protection against lipid oxidation.

The oxygen radical absorbance capacity assay (ORAC) is used to determine the inhibition of peroxyl radical induced oxidation in food and biological materials (Karadag and others 2009). ORAC specifically measures peroxyl radical quenching of fluorescence of fluorescein. Ballard and others (2009) reported the ORAC of peanut skins to be as high as 214900 μMol Trolox/100 g. Davis and others (2010) reported that ORAC of peanut skins increased with increased degree of roast.

Descriptive sensory analysis (DSA) is a powerful and comprehensive tool used in sensory science to generate quantitative and qualitative data. DSA can be used to evaluate quality control parameters, test the effects of ingredients, aid in evaluating processing methods, and can be correlated with other sensory data

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(Meilgaard and others 1999; McNeil and others 2002; Drake 2007). The peanut lexicon developed by Johnsen and others (1988), with an addition by Sanders and others (1989), describes both desirable and undesirable flavors of peanuts. This lexicon has been used as a common communication tool among researchers and various segments of the peanut industry. The peanut lexicon has been used extensively to relate flavor to maturity and curing (Sanders and others 1989), evaluate roast peanut flavor (Bett and others 1994), off-flavor development during storage (Pattee and others 1999), and processing effects (Schirack and others 2006). Because of the antioxidant activity of phenolic compounds, the goal of this research was to evaluate the nutritional antioxidant properties and flavor of peanut paste and peanut butter enhanced with peanut skins.

Materials and Methods

Roasted peanut skins, peanut paste, and peanut butter from runner-type peanuts were obtained from Jimbo's Jumbos Inc. (Edenton, N.C., U.S.A.). Peanut paste was ground peanuts only while peanut butter contained added salt, sugar, and stabilizer which were less than 5% (w/w) of the peanut butter. Skins were milled using a laboratory grade Wiley Mill (Paul N. Gardner Co., Inc., Pompano Beach, Fla., U.S.A.) fitted with a 0.5 mm sieve. Milled peanut skins were blended into peanut paste and peanut butter in concentrations of 0%, 0.5%, 1.0%, 5.0%, 10.0%, 15%, and 20.0% w/w. All samples were prepared in triplicate and all subsequent analyses were performed in triplicate.

Color

The Hunter L, a, b system was utilized for color measurement. Hunter L (0 = black, 100 = white) and *a* value (*+value = red, −value = green*) were determined using a HunterLab Colorimeter (HunterLab DP-9000^{TM} Reston, Va., U.S.A.).

Sample extraction

Samples for lipophilic (L) and hydrophilic (H) ORAC analyses were extracted using a Dionex (Sunnyvale, Calif., U.S.A.) ASE® 200 Accelerated Solvent Extractor (Prior and others 2003; Davis and others 2010). Approximately 1.0 g of sample was weighed analytically and mixed with 25 g of clean sand. Samples and sand were transferred to a 22 mL extraction cell and extracted with 1:1 hexane:dichloromethane for lipophilic analysis. Lipophilic extracts (peanut skins) were dried using nitrogen and adjusted to a final volume of 10 mL using acetone. Samples and sand were then extracted with 70:29.5:0.5 acetone:water:acetic acid (AWA) and brought to 50 mL final volume with additional AWA in preparation for hydrophilic analysis.

Total phenolics

Total phenolics were determined using the Folin-Ciocalteu method (Waterhouse 2002) with modifications. Gallic acid (Sigma-Aldrich Co., St. Louis, Mo., U.S.A.) standards were prepared with 0.0, 50.0, 100.0, 150.0, 250.0, 500.0 mg/L. Approximately 0.1 mL of standard solution and hydrophilic extract samples were pipetted into test tubes, to which 7.9 mL of deionized water and 0.5 mL Folin reagent (Sigma-Aldrich Co.) were added to each of the standard and sample solutions. After 1 min, 1.5 mL of sodium carbonate solution was added followed by vortexing. The sodium carbonate solution was prepared with 100 g anhydrous sodium carbonate in 400 mL water, which was allowed to sit for 24 h, filtered and brought to a final volume of 1 liter. Standards and samples remained at room temperature for 2 h followed by absorbance measurement at 765 nm using a SAFIRE^2 microplate reader equipped with version 6.1 Magellan reader software (Tecan US, Raleigh, N.C., U.S.A.). Total phenolics were calculated as milligrams of gallic acid equivalents per gram (mg GAE/g).

Hydrophilic-ORAC

Hydrophilic oxygen radical absorbance capacity (H-ORAC) was determined using the procedure described by Prior and others (2003) and Davis and others (2010). All solutions and samples were prepared using pH 7.4 phosphate buffer. Solutions of 3.12, 6.25, 12.5, 25, and 50 μM of Trolox (Aldrich, Milwaukee, Wis., U.S.A.) were used as the control standards. Approximately 130 μL of standards and hydrophilic extracts were added to a Costar polystyrene flat-bottom black 96 microwell plate (Corning, Acton, Mass., U.S.A.). Sixty micro liters of a 70 nM fluorescein (FL) solution was then added to the wells and then the plate was incubated in the SAFIRE^2 for 15 min at 37 °C. Next 60 μL of a 153 mM 2,2'-azobis (2-amindino-propane) dihydrochloride (AAPH) (Wako, Richmond, Va., U.S.A.) solution was added rapidly. Fluorescence excitation and emission wavelengths were programmed to 485 and 535 nm. H-ORAC was calculated using a regression equation between the concentration of Trolox and the net area under the curve. H-ORAC was reported as Trolox equivalents (μMol Trolox/100 g).

Lipophilic-ORAC

The L-ORAC procedure was carried out as described by Prior and others (2003) and Davis and others (2010). A 7% randomly methylated beta cyclodextrin (RMCD) (Trappsol®; CTD, Inc., High Springs, Fla., U.S.A.) solution was prepared in 50% acetone: 50% water (7% RMCD). Solutions of standards prepared in 7% RMCD ranged from 200 to 1.56 μM of Trolox (Aldrich, Milwaukee, Wis., U.S.A.). Twenty-five μL of standards and lipophilic extracts were added to the 96-microwell plate. One hundred twenty μL 21.5 nM of fluorescein solution (prepared in 75 mM phosphate buffer) was added to samples using a multichannel pipette and incubated in the SAFIRE^2 for 15 min at 37 °C. AAPH was prepared to a final concentration of 70 mM in phosphate buffer and 80 μL of AAPH solution was added rapidly using a multichannel pipette. Data handling and export were the same as reported for H-ORAC. L-ORAC was calculated using a regression equation between the concentration of Trolox and the net area under the curve. L-ORAC was reported as Trolox equivalents (μMol Trolox/100 g).

Descriptive sensory analysis

Evaluation of peanut paste and peanut butter samples were conducted by an experienced descriptive sensory panel (n = 6, females; n = 6, males; >500 h experience) established using the Spectrum^{TM} universal 15-point intensity scale. Panelists used the peanut lexicon described by Johnsen and others (1988) and Sanders and others (1989). To mask color differences of the various concentrations of peanut skins, all samples were presented in 2oz soufflé cups with lids under red lamps. Samples were equilibrated to and served at room temperature (22 °C).

A carboxymethyl cellulose (CMC) rinse (5.5 g/L, TIC Gums, Belcamp, Md., U.S.A.) protocol described by Beecher and others (2008) was used to minimize astringency carryover effects. Panelists were trained an additional 7 h on bitter, astringency, and familiarization with the CMC protocol. After tasting the sample and expectorating, panelists were instructed to rinse with CMC,

Table 1–Intensity of attributes of peanut paste reference containing 2% tannic acid.

Attribute	Intensity
Roast peanutty	4.5
Sweet aromatic	3.0
Dark roast	3.0
Raw beany	2.0
Woody/Hulls/Skins	3.0
Sweet taste	2.5
Bitter	5.0
Astringency	4.0

Table 2–Hunter L and *a* value of peanut paste (PP) and peanut butter (PB) containing peanut skins (PS).

	PP		PB	
% PS	L	*a*	L	*a*
0	49.5a	7.1c	48.5a	8.5d
0.5	48.1b	7.2c	46.8b	8.5d
1.0	47.9c	7.2c	46.5b	8.5d
5.0	41.1d	7.9b	39.0c	8.9c
10.0	35.8e	8.6a	31.9d	9.4b
15.0	32.9f	8.7a	27.9e	9.4b
20.0	29.2g	8.7a	25.3f	9.6a

[a]Means in the same column with different letters are significantly different ($P < 0.05$).

take a sip of water, a bite of cracker, then another sip of water followed by a 2-min timed waiting period. A peanut paste reference (Table 1), containing 2% w/w tannic acid, was used as a warm-up sample prior to evaluating the test samples. Tannic acid, a plant polyphenol, was added for increased bitter and astringent intensity in the reference peanut paste. The order of sample presentation was randomized for 4 replications and all samples were coded with random 3-digit codes.

Statistical analysis

Analysis of variance (ANOVA) was generated using PROC GLM and comparison of means were made using Duncan's *post hoc* test (SAS version 9.1, Cary, N.C., U.S.A.). Significance was established at $P < 0.05$.

Results and Discussion

The L value for peanut paste and peanut butter containing 0% to 20% (w/w) peanut skins ranged from 49.5 to 29.2 and 48.5 to 25.3, respectively (Table 2). The increase in darkness of both peanut products was directly related to the quantity of skins added. The *a* value (red to green) increased significantly ($P < 0.05$) from 7.1 to 8.7 and 8.5 to 9.6 for peanut paste and peanut butter, respectively, as the concentration of peanut skins increased (Table 2). Stansbury and others (1950) reported dark red extracts from peanut skins. Chukwumah and others (2009) evaluated the relationship between peanut skin color and polyphenolic compounds. The redness of the peanut skin extracts correlated well with the concentration of polyphenolic compounds present in the skins. Compounds, such as procyanidins, found primarily in woody or herbaceous plants, are colorless but may convert to red-brown pigments under atmospheric conditions or under light (Schwartz and others 1996). As such, differences in *a* value were observed in a collected sample of raw skins ($a = 8.8$) and roasted ($a = 10.2$) peanut skins (data not presented), demonstrating an increase in red pigments in roasted skins.

The total phenolics content of peanut skins used in the present study was 158 mg GAE/g. Peanut skin total phenolics have been reported to range from 36 to 280 mg GAE/g (Francisco and Resurreccion 2008). Ballard and others (2009) reported that the total phenolic content of an ethanolic extract of peanut skins was 118 mg GAE/g. The phenolics content of roasted peanut skin extracts using water, methanol, and ethanol solvents was 79.0, 96.7, and 125.0 mg GAE/g, respectively (Yu and others 2005). Yu and others (2006) published total phenolics results of peanut skins removed using direct peel, water blanching, and roasting. Peanuts skins removed by direct peeling (130.8 mg GAE/g) and after roasting (124.3 mg GAE/g) had higher antioxidant activity than skins removed by water blanching (15.1 mg GAE/g). Water blanching of the skin causes phenolic compounds to leach out, resulting in a loss of skin color (Yu and others 2005). Based on data from our study, more than 95% of the phenolic compounds in peanut skins are hydrophilic and supports the concept of possible leaching of these compounds when skins are removed with water blanching. The total phenolic content of peanut paste increased ($P < 0.05$) from 12.9 to 31.9 mg GAE/g across the range of PS added (Table 3) and from 14.1 to 28.1 mg GAE/g in peanut butter samples when peanut skins were added (Table 3). Currently, literature suggests that Americans consume about 1 g of phenolic compounds daily; however, there is no recommended daily dietary intake of phenolic compounds (Scalbert and Williamson 2000; Williamson and Holst 2008).

Table 3– Total phenolics (TP) and oxygen radical absorbance capacity (ORAC) of peanut paste (PP) and peanut butter (PB) containing peanut skins (PS).

	PP		PB	
% PS	TP (GAE/g)	H-ORAC (μMol Trolox /100 g)	TP (GAE/g)	H-ORAC (μMol Trolox /100 g)
0	12.9f	4041f	14.1f	5702f
0.5	13.7ef	4625f	14.7f	6059ef
1.0	14.4e	5846e	15.1e	6547e
5.0	20.8d	7737d	21.5d	8954d
10.0	25.5c	13004c	24.0c	15653c
15.0	27.4b	17145b	25.5b	18071b
20.0	31.9a	20063a	28.1a	20376a

[a]Means in the same column with different letters are significantly different ($P < 0.05$).

Table 4–Oxygen radical absorbance capacity (ORAC) of roasted peanut skins.

	μMol Trolox /100 g $\pm$ SD
Lipophilic	5617 $\pm$ 223
Hydrophilic	189453 $\pm$ 6963
Total	195070

L-ORAC of roasted peanut skins was 5617 $\pm$ 223 μMol Trolox/100 g, while the H-ORAC was 189453 $\pm$ 6963 μMol Trolox/100 g (Table 4). These values are similar to data published by Ballard and others (2009) and Davis and others (2010). ORAC reported for roasted almond skins ranged from 80300 to 108000 μMol Trolox/100 g (Garrido and others 2008). Davis and others (2010) found that roasting for longer times increased antioxidant activity of peanuts, peanut flour, and skins. Increase in ORAC with roasting may be related to an increase in Maillard reaction products which, in addition to phenolic compounds, have antioxidant activity.

The USDA (2007) report of the ORAC of approximately 277 foods included raw peanuts as 3166 μMol Trolox/100 g. The report does not state whether or not the skin was intact; however, based on published data, it is likely that the skin was removed.

Table 5–Descriptive sensory analysis of peanut paste containing peanut skins (PS).

% of PS	Roast peanutty	Sweet aromatic	Dark roast	Raw beany	Woody/ Hulls/Skins	Sweet taste	Bitter	Astringency
0	4.4a	3.0a	3.0a	2.0b	3.2d	2.7ab	2.8d	2.7d
0.5	4.6a	3.2a	3.0a	2.0b	3.3cd	2.9a	2.9cd	3.0cd
1.0	4.3a	3.0a	3.0a	2.0b	3.4cd	2.6ab	2.9cd	2.7d
5.0	4.3a	2.9ab	2.9a	2.1b	3.6c	2.5b	3.3c	3.4c
10.0	3.5b	2.6b	2.9a	2.2b	4.6b	2.2c	4.3b	4.4b
15.0	2.7c	2.2c	2.8a	2.1b	4.9b	1.9d	4.6b	4.3b
20.0	1.9d	1.6d	2.9a	2.4a	5.7a	1.5e	5.3a	5.3a

[a]Means in the same column with different letters are significantly different ($P < 0.05$).

Table 6–Descriptive sensory analysis of peanut butter containing peanut skins (PS).

% of PS	Roast peanutty	Sweet aromatic	Dark roast	Raw beany	Woody/ Hulls/Skins	Sweet taste	Bitter	Astringency
0	4.0ab	2.9ab	2.9a	1.9a	3.0cd	2.8a	2.3d	1.6d
0.5	4.0ab	3.0ab	2.9a	2.0a	2.9d	3.0a	2.5cd	1.8d
1.0	4.2a	3.1a	2.9a	2.1a	3.3cd	3.1a	2.6cd	1.9d
5.0	3.9ab	3.0ab	2.9a	2.2a	3.6c	2.8a	3.1c	2.6c
10.0	3.5b	2.7bc	2.8a	2.2a	4.3b	2.7a	4.1b	3.5b
15.0	2.8c	2.5c	2.9a	2.2a	4.3b	2.6a	4.4b	3.7b
20.0	2.2d	2.0d	2.9a	2.1a	5.3a	1.9b	5.2a	4.3a

[a]Means in the same column with different letters are significantly different ($P < 0.05$).

Davis and others (2010) reported slightly higher ORAC for raw blanched peanuts (3750 μMol Trolox/100 g). ORAC for almonds was reported as 4454 μMol Trolox/100 g (USDA 2007). The antioxidant activity of nuts is reduced when the skin is removed (Schmitzer and others 2011). Nuts such as walnuts and pecans, typically eaten with the skin intact, have an ORAC of 13541 and 17940, μMol Trolox/100 g, respectively (USDA 2007). The ORAC of peanut paste and peanut butter increased as the concentration of peanut skins increased, and ranged from 4041 to 20063 and 5702 to 20376 μMol Trolox/100 g, respectively (Table 3). According to the USDA database (2007), peanut butter has an antioxidant capacity of 3432 μMol Trolox/100 g. The differences in antioxidant capacity for peanut butter in the database and the value in this study (5702 μMol Trolox/100 g) may be related to peanut production area and market type used in the products as well as process parameters, and/or composition of the 2 products.

The addition of peanut skins at greater than 5% resulted in a decrease in roast peanutty intensity and an increase in woody/hulls/skins, bitter, and astringency intensities in peanut paste and peanut butter (Table 5 and 6). The panel did not detect differences ($P > 0.05$) in roast peanutty intensity among samples through 5.0% skins in peanut paste but differences were detected at 10% added skins (Table 5). Similar results were found in peanut butter (Table 6). Peanut skin is approximately 3% of the weight of unblanched peanuts and the results from this study provide evidence that slightly more than the normal peanut skin weight may be added to peanut paste without discernable difference in flavor.

Sweet aromatic is described as aromatics associated with sweet material such as caramel, vanilla, molasses, and fruit (Johnsen and others 1988). For peanut paste, sweet aromatic did not become significantly ($P < 0.05$) lower until 10% skins were added. The same was observed for peanut butter. Wood/hulls/skins is associated with base peanut character (absence of fragrant top notes) and related to dry wood, peanut hulls, and skins (Johnsen and others 1988). Since peanut skins are more closely associated with woody/hulls/skins, the addition of this material to peanut paste and peanut butter should and did increase base peanut and woody notes. Johnson (2007) used DSA to describe the flavor and aroma profile of almond skins as toasted and toasted:nutty, respectively. Panelists described the skins as having "mild flavor and aroma" but descriptors such as woody and earthy were also reported. The authors suggested that almond skins could be added to foods with little change to flavor but did not report studies to demonstrate that fact.

Astringency intensity was higher in peanut paste than in peanut butter (Table 5 and 6). Viscosity of a material has an effect on perceived astringency. Smith and others (1996) demonstrated that astringency intensity decreased as viscosity was increased in aqueous solutions of grape seed tannins using CMC. Similarly, Peleg and Noble (1999) reported that perceived astringency decreased with increasing viscosity of cranberry juice using CMC. Pectin has also been reported to reduce perceived astringency in catechin-based solutions (Hayashi and others 2005). Buck (2010) measured the yield stress, the minimum shear stress required to initiate flow, of peanut paste and commercial peanut butter. Peanut paste ranged from 0.58 to 2.02 kPa, while peanut butter had a yield stress of 10.61 kPa. Because of the addition of stabilizer to prevent oil separation, peanut butter is typically more viscous than peanut paste. The lower perceived astringency in peanut butter in the current study compared to peanut paste may be in part a result of increased viscosity. Sucrose has been previously demonstrated to reduce perceived astringency, likely due to concentration and viscosity of the test solution (Lyman and Green 1990; Breslin and others 1993).

Procyanidins have been identified as one of the key compounds in foods responsible for bitterness (Lopez and others 2007). The molecular structure of phenolic compounds can affect bitter and astringency perception. Low-molecular weight (<500) phenolic compounds tend to be bitter, while higher-molecular weight (>500) compounds tend to be astringent (Lea and Arnold 1978; Robichaud and Noble 1990; Noble 1994; Peleg and others 1999). Low molecular weight compounds with antioxidant activity found in peanut skins, such as caffeic acid (MW = 180), chlorogenic acid (MW = 354), ellagic acid (MW = 302), and procyanidin monomers (MW = 289) may be perceived as bitter (Peleg and others 1999; Yu and others 2005). While A-type procyanidin dimers (MW = 575), B-type procyanidin dimers (MW = 577), A-type procyanidin trimers (MW = 863), B-type procyanidin trimers (MW = 865), A-type procyanidin tetramers (MW = 1149), and B-type procyanidin tetramers (MW = 1151) found

in peanut skins tend to be more astringent (Peleg and others 1999; Yu and others 2005). The mechanism of astringency perception may result from a decrease in saliva lubrication caused by binding and precipitation of proteins (Charlton and others 2002; Jöbstl and others 2004). Increases in bitter and astringent may be described as unpleasant sensory attributes, which can negatively correlate with consumer liking (Young and others 2005).

Conclusion

The addition of peanut skins to peanut paste and peanut butter significantly increased the total phenolics and ORAC. Addition of 5% (w/w) of peanut skins to peanut paste and peanut butter resulted in increase of peanut skin related flavors and 10% skins resulted in overall reduced flavor. This study indicated a potential limited application for peanut skins in peanut paste and peanut butter and perhaps in other products to improve nutritional quality.

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