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Peanut skins-fortified peanut butters: Effect of processing on the phenolics content, fibre content and antioxidant activity

Yuanyuan Ma [a], William L. Kerr [a], Ruthann B. Swanson [b], James L. Hargrove [b], Ronald B. Pegg [a,*]

[a] Department of Food Science & Technology, College of Agricultural and Environmental Sciences, The University of Georgia, 100 Cedar Street, Athens, GA 30602, USA
[b] Department of Foods and Nutrition, College of Family and Consumer Sciences, The University of Georgia, 305 Sanford Drive, Athens, GA 30602, USA

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ABSTRACT

Incorporation of ground peanut skins (PS) into peanut butter at 1.25%, 2.5%, 3.75%, and 5.0% (w/w) resulted in a marked concentration-dependent increase in both the total phenolics content (TPC) and antioxidant activity. Using dry-blanched PS to illustrate, the TPC increased by 86%, 357%, 533%, and 714%, respectively, compared to the peanut butter control devoid of PS; the total proanthocyanidins content (TPACs) rose by 633%, 1933%, 3500%, and 5033%, respectively. NP-HPLC detection confirmed that the increase in the phenolics content was attributed to the endogenous proanthocyanidins of the PS, which were characterised as dimers to nonamers by NP-HPLC/ESI-MS. FRAP values increased correspondingly by 62%, 387%, 747%, and 829%, while H-ORAC$_{FL}$ values grew by 53%, 247%, 382%, and 415%, respectively. The dietary fibre content of dry-blanched PS was ~55%, with 89–93% being insoluble fibre. Data revealed that PS addition enhances the antioxidant capacity of the peanut butter, permits a "good source of fibre" claim, and offers diversification in the market's product line.

1. Introduction

As demonstrated by George Washington Carver more than a century ago, peanuts (*Arachis hypogaea* L.) are a valuable cash crop to the southern United States. China, India and the U.S.A. account for ~2/3 of the world's peanut production. In 2012 the U.S. peanut production reached 6.7 billion pounds nationwide, generating a farm gate value of $2.3 billion (United States Department of Agriculture, 2013). The potential health benefits associated with eating peanuts are well documented, notably the prevention against cardiovascular disease, type-2 diabetes, cancer, and other degenerative diseases (Isanga & Zhang, 2007). Peanut lipids have largely contributed to these benefits; they are rich in monounsaturated fatty acids (MUFAs), polyunsaturated fatty acids (PUFAs) and phytosterols (Isanga & Zhang, 2007). Additional beneficial nutrients endogenous to peanuts include vitamin E, L-arginine, soluble and insoluble fibre, as well as water- and lipid-soluble natural phenolic antioxidants (Isanga & Zhang, 2007). The nutrients act synergistically with the numerous protective bioactives, making the peanut a desirable nutrient-dense plant-based food.

Besides the kernels, peanut skins (PS), as the other edible part of peanuts, have attracted attention because they are a rich, inexpensive source of potentially health-promoting phenolics and dietary fibre (DF). Phenolic compounds typically concentrate themselves on the outer layers of plants, such as the peel, shell, and hull, to protect the inner core materials. Nepote, Grosso, and Guzman (2002) reported a content of ~159 mg total phenolics/g defatted dry skin, which also exhibited a marked antioxidant activity, as demonstrated by its capacity to inhibit the oxidation of sunflower oil. A study by Yu, Ahmedna, and Goktepe (2005) revealed that PS phenolics are abundant not only in quantity but also in type, which primarily include free-, esterified- and bound-phenolic acids (*i.e.*, caffeic, chlorogenic, ferulic, and *p*-coumaric acids), flavan-3-ols (*i.e.*, (+)-catechin, (−)-epicatechin and their polymers {the proanthocyanidins, PACs}), and stilbenes (*i.e.*, resveratrol). Free phenolic acids are not the predominant phenolics in PS (Yu, Ahmedna, Goktepe, & Dai, 2006); the PACs comprise ~17% by weight of PS (Karchesy & Hemingway, 1986). Six A-type PAC dimers were identified in PS (Lou et al., 1999) and found to inhibit the inflammatory pathway mediated by hyaluronidase-induced release of histamine. A further study by Lou et al. (2004) led to isolation of five oligomeric PACs with potential free radical-scavenging activity from the water-soluble fraction of PS.

PACs are complex flavonoid polymers; their phenolic nature makes them excellent candidates as food antioxidants. The health benefits associated with PACs have been documented in terms of antioxidation, anti-carcinogenesis, and cardiovascular disease prevention (Santos-Buelga & Scalbert, 2000). On the other hand, the anti-nutritional effects associated with PACs, notably inhibition on protein digestibility, are clear in animals consuming large amounts of PACs, but have no established nutritional significance

* Corresponding author. Tel.: +1 706 542 1099; fax: +1 706 542 1050.
E-mail address: rpegg@uga.edu (R.B. Pegg).

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in humans (Santos-Buelga & Scalbert, 2000). In the gut, PACs trigger adaptative responses, such as increasing the secretion of various endogenous proteins (especially salivary proline-rich proteins) and biliary acids, which can improve the absorption and utilisation of ingested proteins. Moreover, tannins bound to proteins have been shown to retain their antioxidant activity and may provide persistent antioxidant activity in the gastrointestinal tract when consumed (Riedl, Carando, Alessio, McCarthy, & Hagerman, 2002).

PS are also rich in DF: the total dietary fibre (TDF) comprises ~45% weight of roasted PS, of which roughly 2.2% is soluble fibre (Shimizu-ibuka et al., 2009). A high daily intake of DF helps lower blood pressure and cholesterol levels, resulting in the reduced risk of coronary heart disease, stroke, hypertension, diabetes and obesity (Anderson et al., 2009). Unfortunately, the average fibre consumption amongst adults in the U.S. is ~15 g a day instead of the recommended 25–30 g. Polyphenols and DF are two well-documented dietary factors in the prevention of chronic diseases, but are usually addressed as separate compounds acting independently in disease prevention. A 2011 study by Saura-Calixto demonstrated a synergistic function of DF and dietary antioxidants, mainly in the development of an antioxidant environment in the colon (Saura-Calixto, 2011).

PS have been used in traditional Chinese medicine for the treatment of chronic haemorrhage and bronchitis (Lou et al., 1999). In contrast, the value of PS in the western world has been recognised only recently. As a major waste of peanut processing, most PS are dumped but a small quantity is added to animal feed. In 1999/2000, over 750,000 tons of PS were generated worldwide based on an estimated 29.1 million tons production of peanuts. The red skin of peanuts comprises 2.0–3.5 weight percent of the kernels. In the United States, the total volume of commercially processed shelled edible-grade peanuts used in primary products was roughly 2000 million pounds during 2011 (United States Department of Agriculture, 2010–2012); hence, 40–70 million pounds of PS were generated.

An increased awareness of the role of dietary antioxidants and fibre in health promotion and disease prevention has led to a high demand for antioxidant and fibre-enriched functional foods. PS are a concentrated source of DF and phenolics; thus, their incorporation into a variety of foods would effectively enhance the fibre content and antioxidant capacity of the resultant product and further provide an inexpensive and abundant source of these dietary bioactives. Despite the tremendous potential benefit of PS as an alternative source of antioxidants and DF, their utilisation as a functional food ingredient in value-added products is lacking.

The objectives of the current study are (i) to assess the effect of processing on the phenolics content (TPC and TPAC content) and antioxidant activity of PS and in value-added peanut butters fortified with PS; (ii) to determine the increased fibre content after incorporating PS into the peanut butter prototypes; and (iii) to characterise the phenolics in PS and the PS-fortified peanut butters.

2. Materials and methods

2.1. Materials

Dry-blanched PS were provided by Universal Blanchers, LLC (Sylvester, GA). Roasted PS (light, medium, and dark) were a gift from the Golden Peanut Company, LLC (Alpharetta, GA). Peanut paste, flour salt, and hydrogenated vegetable oil (*i.e.*, stabiliser) were supplied by Seabrook Ingredients, Inc. (Edenton, NC). Domino premium pure cane granulated sugar and peanut oil were purchased from Sam's Club (Athens, GA). All solvents and reagents were of analytical (ACS) grade, unless otherwise specified. Methanol, ethanol (95%) and hexanes were purchased from VWR International (Suwanee, GA). Folin & Ciocalteu's phenol reagent, (+)-catechin hydrate, 4-(dimethylamino)cinnamaldehyde (DMAC), fluorescein sodium salt, (±)-6-hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid (Trolox), 2,2′-azobis(2-amidinopropane) dihydrochloride (AAPH), 2,4,6-tri(2-pyridyl)-*s*-triazine (TPTZ), and a TDF assay kit (Cat No. TDF100A-1KT) were acquired from the Sigma–Aldrich Chemical Co. (St. Louis, MO). Procyanidin B2 was purchased from Indofine Chemical Company, Inc. (Hillsborough, NJ) and the PAC standards with a degree of polymerisation (DP) = 2 through DP = 10 were obtained from Planta Analytica (Danbury, CT).

2.2. Peanut butter processing

Seventeen formulations of peanut butter (~45% total fat) were prepared, as described by Ma et al. (2013). To facilitate incorporation, PS and sugar (195 g) were ground together at different ratios prior to inclusion into the prototypes, using an electrically-driven stone grinder (Super Masscolloider CA6-3; Masuko Sangyo Co., Ltd, Kawaguchi-City, Saitama, Japan) fitted with a BA6-80 grit stone assembly. More specifically, 22.9, 45.8, 68.7, and 91.6 g of dry-blanched PS were ground with 195 g of sugar; the PS-sugar powders after sieving were then added to peanut butter formulations to achieve PS addition levels of 1.25%, 2.5%, 3.75%, and 5.0% PS. The particle size distribution of the ground PS preparations, represented by Feret's diameters (*i.e.*, the maximum caliper, measured from the longest distance between any two points along the selection boundary), are reported by Ma et al. (2013). Fortified peanut butters were placed in storage at 4 °C until analysed.

2.3. Extraction of phenolics

Phenolic compounds were extracted from both PS and PS-fortified peanut. Samples were placed in cellulose extraction thimbles (43 mm *i.d.* × 123 mm, VWR International, Suwanee, GA), covered with a plug of glass wool and defatted in a Soxhlet extraction apparatus under reflux for 12–14 h with hexanes. Defatted PS and peanut butters were transferred to 125-mL Erlenmeyer flasks at a mass-to-solvent ratio of 1:10 (w/v) with 80% (v/v) aqueous acetone. Extractions were performed in a gyratory water bath shaker (Model G76; New Brunswick Scientific Company, Inc., New Brunswick, NJ) set at 150 rpm and a temperature of 45 °C for 30 min. The slurry was then filtered by gravity through P8 filter paper (Fisher Scientific Co., Suwanee, GA). The extraction process was repeated twice as described above. All filtrates were pooled and acetone was evaporated with a Büchi Rotavapor R-210 using a V-700 vacuum pump connected to a V-850 vacuum controller (Büchi Corporation, New Castle, DE) at 45 °C. The aqueous residue was frozen and then lyophilised in a FreeZone® 2.5-L bench-top freeze dryer (Labconco Corporation, Kansas City, MO) to ensure all traces of moisture were removed and then stored in amber-glass bottles at −20 °C until further analysed.

2.4. Preparation of desugared peanut butter extracts

The method described by Srivastava et al. (2010) for preparing a polyphenolic extract from blackberries was adapted for peanut butter. Briefly, ~3 g of each crude peanut butter extract were dispersed in 10 mL of deionised water, sonicated to facilitate dissolution, and then applied to the top of a chromatographic column (30 mm *i.d.* × 340 mm, Kontes Glass Inc., Vineland, NJ) packed with Amberlite XAD-16 [(bead size: 20–60 mesh), Sigma–Aldrich] and washed with ~1000 mL of deionised water to remove sugars and organic acids. After the first 800 mL, the Brix reading of the eluent was checked using a digital PAL-1 pocket refractometer (Model