

Article

# Antioxidant Properties of Soybean Oil Supplemented with Ginger and Turmeric Powders

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**Abstract:** Soybean oil has been supplemented with 10% (*w/w*) of ginger and turmeric powders derived from commercial products (GC—commercial ginger and TC—commercial turmeric), freeze-dried rhizomes (freeze-dried ginger (GR) and freeze-dried turmeric rhizome—TR) and peels (freeze-dried ginger peel (GP) and freeze-dried turmeric peel—TP) for developing a functional seasoning with great lipid stability for human consumption. The exhausted ginger and turmeric powders were also recovered and recycled two times to promote a more sustainable process. The antioxidant activity and oxidative stability of oil samples were evaluated respectively by spectrophotometric and Rancimat methods. Folin–Ciocalteu assay and HPLC analysis were also performed to quantify total polyphenols, ginger-derived 6-gingerol and 6-shogaol, and turmeric-derived curcumin. Their antioxidant activity as well as oxidative stability, which non-linearly decreased over cycles because of a strongly reduced phenolic extractability, linearly increased with increasing phenolic yields. Hence, ginger and turmeric can be proposed as healthy spices containing bioactive compounds to control lipid oxidation and improve oil stability. Moreover, the valorization of peels as eco-friendly source of natural antioxidants is a valid strategy for providing added-value to these agro-food wastes.

**Keywords:** lipid oxidation; oxidative stability; Rancimat; natural antioxidants; by-products; phenolic compounds; 6-gingerol; 6-shogaol; curcumin; Zingiberaceae

## 1. Introduction

Soybean oil is rich in polyunsaturated fatty acids such as linoleic and linolenic acids, which are the main responsible of its oxidative instability [1,2], due to the low dissociation energy of their double bonds. In this respect, oxygen plays a leading role in the free radical chain reaction of lipid oxidation mechanism that is explained through the stages of initiation, propagation and termination. The addition of antioxidants as free radical scavengers before the propagation phase, is considered a strategy to counteract the lipid autoxidation and to enhance the lag time until the production of the volatile compounds which are markers of rancidity [3].

Currently, the food industry is moving towards replacing the application of synthetic by natural antioxidants, which are considered safer and healthier [4]. Several studies proposed natural alternatives to BHA (butylated hydroxyanisole) and BHT (butylated hydroxytoluene) for effectively improving the lipid stability of soybean oil [5–15]. Agro-food products, by-products and wastes are rich in bioactive compounds [16–25] thus contributing to the development of new functional ingredients and products [26–28]. Among natural antioxidants derived from plant source, polyphenols are effective in controlling the oil and fat rancidity because they act as free radical scavengers according to two possible mechanisms based on the transfer of hydrogen atom or single electron from antioxidant to oxidant molecules [29]. The Zingiberaceae spices are well known in

the world [30]. Ginger rhizome (*Zingiber officinale*) contains phenolic acids such as gingerols and shogaols [31], which have been associated with antioxidant, anti-inflammatory, anticancer, antimicrobial, antifungal antiviral, antidiabetic, and anti-obesity properties [32–34]. Instead, curcuminoids classified as “other polyphenols”, are the key bioactive compounds of turmeric rhizome (*Curcuma longa*). The main curcuminoid is curcumin [35], the yellow pigment associated with several health benefits [36–40]. In addition, ginger and turmeric peels, which are generally discarded during post-harvest operations, have been previously recovered and proposed as a potential source of natural antioxidants [41]. Although the antioxidant capacity of ginger and turmeric rhizomes and peels has been widely demonstrated by several studies [42–46], none investigated their effect in preventing the lipid oxidation of vegetable oils. Thus, this study is aimed at evaluating the antioxidant activity and oxidative stability of ginger- and turmeric-supplemented soybean oils correlating also their phenolic composition. Moreover, the valorization of peels as a promising antioxidant by-product is proposed.

## 2. Materials and Methods

### 2.1. Materials and Reagents

Refined soybean oil, fresh rhizomes, and commercial spices of ginger and turmeric were purchased by a local market. Butylated hydroxytoluene (BHT), 2,2-diphenyl-1-picrylhydrazyl (DPPH), diammonium salt of the 2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS), ethanol (EtOH), Folin–Ciocalteu's phenol reagent, gallic acid, ginger powder, hydrochloric acid, ( $\pm$ )-6-hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid (Trolox), methanol (MetOH), sodium acetate trihydrate, sodium carbonate, 2,4,6-tripyridyl-s-triazine (TPTZ), Tween80 and HPLC standards were purchased from Sigma-Aldrich (St. Louis, MO, USA).

### 2.2. Preparation of the Ginger and Turmeric Powders

The fresh rhizomes of ginger and turmeric have been manually peeled. The peels and their rhizomes were cut into small pieces, freeze dried at  $-40\text{ }^{\circ}\text{C}$ , and grounded with a water-cooled laboratory mill (IKA Werke M20, Staufen im Breisgau, Germany). After that, a representative portion of samples was dried in a stove at  $101\text{ }^{\circ}\text{C}$  for 24 h in order to measure the dry matter, which was approximately of  $92.58 \pm 3.21\%$  for ginger powders and  $90.19 \pm 0.89\%$  for turmeric powders.

### 2.3. Preparation and Phenolic Extraction of the Ginger- and Turmeric-Supplemented Soybean Oils

Soybean oil has been added with 10% (*w/w*) of ginger and turmeric powders, mixed for 10 min using ULTRA-TURRAX<sup>®</sup> disperser tool, and centrifuged for 10 min at 5000 rpm at  $4\text{ }^{\circ}\text{C}$  by a centrifuge (Hettich Zentrifugen, MOD: Universal 320R, Tuttlingen, Germany). Then, the supernatants consisting of soybean oils supplemented with original powders were recovered (Cycle 1) while the exhausted powders were recycled two times by adding soybean oil and following the same enrichment procedure (Cycle 2 and Cycle 3, respectively). The soybean oil supplemented with ginger and turmeric powders derived from commercial products (GC—commercial ginger and TC—commercial turmeric), freeze-dried rhizomes (freeze-dried ginger (GR) and freeze-dried turmeric rhizome—TR) and peels (freeze-dried ginger peel (GP) and freeze-dried turmeric peel—TP) at three different oil enrichment cycles were stored at  $25\text{ }^{\circ}\text{C}$  under dark conditions for one week. To extract phenolic compounds, a 5 g of oil samples were mixed for three consecutive times with 5 mL of mixture based on MetOH and 10% *v/v* Tween 80 (80:20 *v/v*) through an orbital shaker for 5 min and centrifuged for 20 min at 5000 rpm and  $10\text{ }^{\circ}\text{C}$ . All of the supernatants were collected and freeze-stored before analysis under dark conditions.

#### 2.4. Determination of Total Polyphenols

The content of total polyphenols was detected in the phenolic extracts of oil samples by following the Folin–Ciocalteu colorimetric method [41]. The total phenolic content was expressed as gallic acid equivalents per gram of oil (mg GAE/g).

#### 2.5. HPLC Analysis of 6-Gingerol, 6-Shogaol and Curcumin

The 6-gingerol, 6-shogaol and curcumin were HPLC-quantified in the phenolic extracts of oil samples through a Thermo Finnigan SpectraSystem UV6000LP HPLC system (Thermo Finnigan, San Jose, CA, USA) with diode-array detector and a Supelcosil LC-18 column (Sigma-Aldrich). The samples were injected into the column after filtering with 0.22  $\mu\text{m}$  Millipore cellulose acetate filters (Merck Millipore, Billerica, MA, USA).

The 6-gingerol and 6-shogaol were HPLC-detected as follows [31]: a mobile phase consisting of distilled water (Solvent A) and acetonitrile (Solvent B) at different elution gradient (0–8 min, 50% B; 8–17 min, 50–55% B; 17–32 min, 55–100% B; 32–38 min, 100% B; 38–40 min, 100–45% B; 40–50 min, 45% B; 50–60 min, 45–50% B); flow rate of 0.2 mL/min; injection volume of 10  $\mu\text{L}$ ; column temperature of 30  $^{\circ}\text{C}$ ; chromatographic run time of 60 min.

The curcumin was HPLC-detected as follows [35]: a mobile phase consisting of acetonitrile/0.1% *v/v* acetic acid (Solvent A) and distilled water/0.1% acetic acid (Solvent B) at different gradient elution (0–6 min, 45–35% B; 6–21 min, 35–10% B; 21–27 min, 10% B; 27–30 min, 10–25% B; 30–39 min, 25% B; 39–51 min, 25–45% B; 51–70 min, 45% B); flow rate of 1.2 mL/min; injection volume of 10  $\mu\text{L}$ ; column temperature of 55  $^{\circ}\text{C}$ ; chromatographic run time of 70 min. The content of each compound was expressed as milligram per gram of oil (mg/g).

#### 2.6. Determination of Antioxidant Activity

The antioxidant activity was detected in the phenolic extracts of oil samples by using 2,2-di(4-tert-octylphenyl)-1-picrylhydrazyl (DPPH) and ferric reducing ability of plasma (FRAP) assays according to a previous study [41]. The antioxidant activity was expressed as Trolox equivalents per gram of oil (mg TE/g).

#### 2.7. Determination of Oxidative Stability

The oxidative stability of oil samples was monitored by the official Rancimat method [47] and was compared with that of soybean oils without antioxidants (C) and with butylated hydroxytoluene (BHT) as synthetic antioxidant reference for vegetable oils at the maximum concentration of 0.02% *w/w* according to Codex Alimentarius [48]. After weighing 3 g, the oil sample was loaded into the Rancimat instrument (Metrohm, model 743, Herisau, Switzerland), which measured over time the water conductivity at 110  $^{\circ}\text{C}$  temperature and 20 L  $\text{h}^{-1}$  air flow [2]. The oxidative stability was expressed as the induction period (IP) corresponding to the time (h) at which the water conductivity ( $\mu\text{S min}^{-1}$ ) starts increasing because of the production of compounds involved in the lipid oxidation.

#### 2.8. Statistical Analysis

All of the data obtained from three replicates ( $n = 3$ ) were presented as mean  $\pm$  standard deviation (SD) and analyzed by a one-way analysis of variance (ANOVA) through the PROC GLM of SAS<sup>®</sup> 9.3 software packaging. Differences among means with  $p < 0.05$  were considered as statistically significant differences in accordance with Bonferroni test. Linear regression analysis was performed by correlating the DPPH, FRAP, and IP values with the contents of 6-gingerol and curcumin using the PROC REG and PROC CORR of SAS<sup>®</sup> 9.3 software packaging.

### 3. Results and Discussion

#### 3.1. Phenolic Characterization of Ginger- and Turmeric-Supplemented Soybean Oils

The total phenolic content was not detected in the refined soybean oil used as control, in agreement with other authors [49], even if a total phenolic content respectively of 0.13 mM GAE/L [50], 200 mg GAE/kg [10,11], 139 mg GAE/g [12] was determined in other commercial soybean oils. This could be associated not only to the different conditions of phenolic extraction, but also to the commercial refining grade, since it has been demonstrated that the total phenolic content of crude soybean oil decreased during refining steps [51]. In this study, the total phenolic content of soybean oil increased only after adding ginger and turmeric powders containing mostly ginger-derived 6-gingerol and 6-shogaol, and turmeric-derived curcumin [31,35,41]. The high oil solubility of these hydrophobic compounds has been confirmed by several studies [7,52]. Based on the comparison between 6-gingerol content of ginger-supplemented oils at the first cycle (Table 1) with that of the corresponding powders analyzed in our previous study [41], the solubility in oil for all three ginger powders was 100%. Comparing curcumin content of turmeric-supplemented oils at the first cycle (Table 1) with that of the corresponding powders analyzed in our previous study [41], the oil solubility for commercial product (43%) was significantly lower than that for freeze-dried powders derived from rhizomes and peels (89% and 100%, respectively). In this regard, the food processing could affect the bioaccessibility and bioavailability of phytonutrients [53].

The contents of total polyphenols as well as bioactive compounds statistically differed ( $p < 0.001$ ) among oil samples at each enrichment cycle (Table 1). The phenolic amounts of all samples in the three oil enrichment cycles were much higher than those found in soybean oils flavored with thyme and rosemary extracts (0.93 and 0.38 mg GAE/g, respectively; [10]), and with microwave-assisted olive leaf extract (0.07 mg GAE/g; [8]) for effectively preventing the lipid oxidation. Soybean oils with turmeric powders such as TR and TP had the greatest total phenolic content at Cycle 1 and Cycle 2, while TC and GP achieved the best phenolic yields at Cycle 3. The powder preparation and structure could affect the bioactive compounds extractability during oil enrichment [53]. In this regard, turmeric-supplemented soybean oils especially TR and TP achieved the maximum yields of total polyphenols and curcumin in the first two cycles probably due to the high lipid solubility of turmeric-derived curcumin [54]. In fact, the percentage reductions of phenolic and curcumin contents in TR (respectively 50% and 77% at Cycle 2; 79% and 97% at Cycle 3) and TP (respectively 58% and 76% at Cycle 2; 81% and 96% at Cycle 3) enhanced at increasing oil enrichment cycles and were higher than those of TC (respectively 19% and 18% at Cycle 2; 33% and 40% at Cycle 3). Regarding ginger-supplemented soybean oils, the amounts of total polyphenols and 6-gingerol were greater in GP whose percentage reductions (respectively 21% and 30% at Cycle 2; 38% and 79% at Cycle 3) were lower than those of GR (respectively 46% and 83% at Cycle 2; 66% and 100% at Cycle 3) and GC (respectively 32% and 70% at Cycle 2; 52% and 96% at Cycle 3). Instead, the 6-shogaol content was stable over cycles. The 6-shogaol is a compound derived from fresh ginger during thermal drying or storage [31]. The highest 6-shogaol amount in GC could be attributed to the different drying procedure [45].

**Table 1.** The phenolic contents in ginger- and turmeric-supplemented soybean oils at different oil enrichment cycles <sup>1</sup>.

Parameters	Soybean Oils <sup>2</sup>					
	GC	GP	GR	TC	TP	TR
<b>Cycle 1</b>						
Total polyphenols (mg GAE/g)	3.00 ± 0.08 <sup>b</sup>	3.59 ± 0.25 <sup>b</sup>	3.75 ± 0.10 <sup>b</sup>	3.46 ± 0.30 <sup>b</sup>	9.19 ± 0.73 <sup>a</sup>	8.53 ± 0.44 <sup>a</sup>
6-gingerol (µg/g)	1156.50 ± 7.00 <sup>c</sup>	1734.58 ± 17.00 <sup>a</sup>	1352.44 ± 11.00 <sup>b</sup>	–	–	–
6-shogaol (µg/g)	37.43 ± 1.00 <sup>a</sup>	20.00 ± 0.60 <sup>b</sup>	13.75 ± 0.50 <sup>c</sup>	–	–	–
Curcumin (µg/g)	–	–	–	1112.62 ± 5.00 <sup>a</sup>	5890.77 ± 18.00 <sup>b</sup>	6032.87 ± 23.00 <sup>c</sup>
<b>Cycle 2</b>						
Total polyphenols (mg GAE/g)	2.05 ± 0.08 <sup>c</sup>	2.97 ± 0.27 <sup>b</sup>	1.92 ± 0.12 <sup>c</sup>	2.82 ± 0.08 <sup>b</sup>	3.91 ± 0.34 <sup>a</sup>	4.30 ± 0.25 <sup>a</sup>
6-gingerol (µg/g)	342.68 ± 3.00 <sup>b</sup>	1213.93 ± 12.00 <sup>a</sup>	226.74 ± 4.00 <sup>c</sup>	–	–	–
6-shogaol (µg/g)	37.79 ± 0.70 <sup>a</sup>	19.15 ± 0.50 <sup>b</sup>	11.97 ± 0.30 <sup>c</sup>	–	–	–
Curcumin (µg/g)	–	–	–	908.57 ± 9.00 <sup>c</sup>	1427.36 ± 15.00 <sup>a</sup>	1380.96 ± 13.00 <sup>b</sup>
<b>Cycle 3</b>						
Total polyphenols (mg GAE/g)	1.45 ± 0.04 <sup>c</sup>	2.24 ± 0.07 <sup>a</sup>	1.29 ± 0.01 <sup>c</sup>	2.33 ± 0.04 <sup>a</sup>	1.78 ± 0.10 <sup>b</sup>	1.83 ± 0.03 <sup>b</sup>
6-gingerol (µg/g)	44.84 ± 0.30 <sup>b</sup>	364.10 ± 6.00 <sup>a</sup>	3.22 ± 0.03 <sup>c</sup>	–	–	–
6-shogaol (µg/g)	36.68 ± 0.20 <sup>a</sup>	19.49 ± 0.10 <sup>b</sup>	8.64 ± 0.04 <sup>c</sup>	–	–	–
Curcumin (µg/g)	–	–	–	369.58 ± 7.00 <sup>a</sup>	263.49 ± 3.00 <sup>b</sup>	158.59 ± 2.00 <sup>c</sup>

<sup>1</sup> Cycle 1: oil enrichment with original powders; Cycle 2: oil enrichment with exhausted and one time recycled powders; Cycle 3: oil enrichment with exhausted and two times recycled powders. <sup>2</sup> GC: commercial ginger; GP: freeze-dried ginger peel; GR: freeze-dried ginger rhizome; TC: commercial turmeric; TP: freeze-dried turmeric peel; TR: freeze-dried turmeric rhizome. <sup>a-c</sup> Data are presented as mean ± SD ( $n = 3$ ). Values with different letters within rows are statistically different ( $p < 0.05$ ) as determined by Bonferroni test.

### 3.2. Antioxidant Activity of Ginger- and Turmeric-Supplemented Soybean Oils

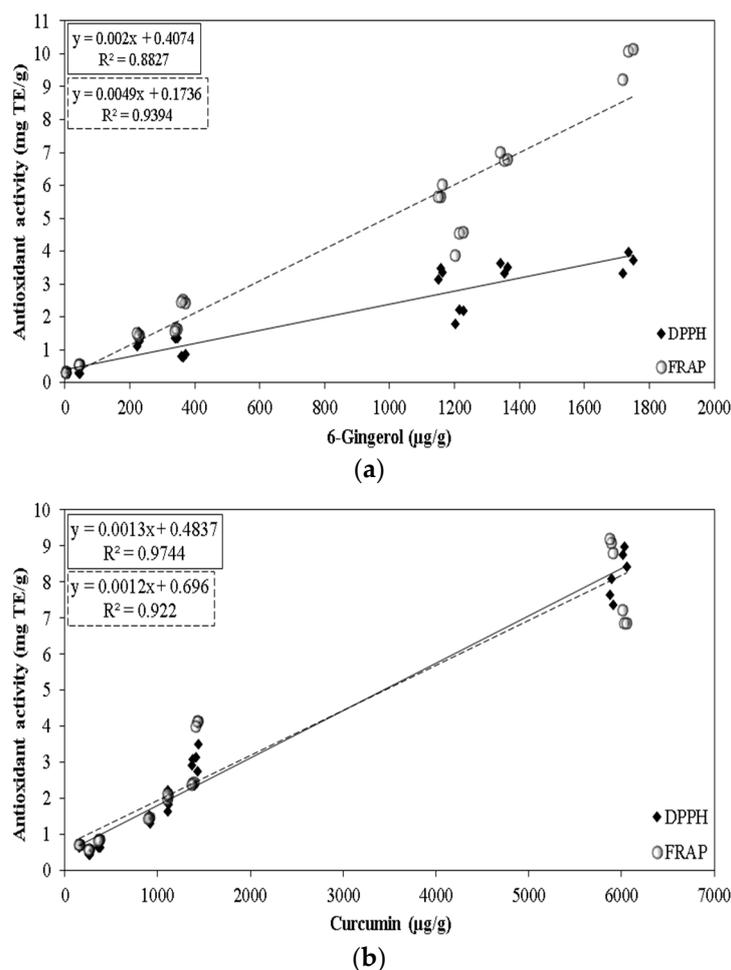
As previously reported, the total phenolic content, and also the antioxidant activity of the refined soybean oil, used as control, was not detected (0.00 mg TE/g), as confirmed by DPPH and FRAP assays. Therefore, the antioxidant activity increased only after adding ginger and turmeric powders containing natural antioxidants [41–46]. The antioxidant activity was significantly different ( $p < 0.001$ ) among oil samples at each enrichment cycle in both DPPH and FRAP assays (Table 2). All of the ginger- and turmeric-supplemented soybean oils lead to greater antioxidant properties than the reference BHT. Oil samples such as TR and TP had the highest antioxidant activity at Cycle 1 and Cycle 2, while GP achieved the best performance at Cycle 3 because of their greater phenolic yields (Table 1). In this regard, previous research works associated the antioxidant activity of agro-food products and wastes with their phenolic composition [6,17,22,25,27,28]. Other authors found that phytochemicals such as 6-gingerol and curcumin contributed to the antioxidant effects of ginger and turmeric rhizomes and peels [41]. Regarding turmeric-supplemented soybean oils, TR and TP had greater antioxidant activity than that of TC only for the first two cycles where achieved the highest yields of total polyphenols and curcumin (Table 1). The percentage reductions of DPPH and FRAP values in TR (respectively 68% and 66% at Cycle 2; 92% and 90% at Cycle 3) and TP (respectively 59% and 55% at Cycle 2, 94% at Cycle 3) were higher than those of TC (respectively 25% and 30% at Cycle 2, 67% and 59% at Cycle 3) as a consequence of their greater percentage reductions of curcumin content (Table 1). Regarding ginger-supplemented soybean oils, GP, which achieved the best antioxidant performance at each oil enrichment cycle, had lower percentage reductions of DPPH and FRAP values (respectively 44% and 56% at Cycle 2; 78% and 75% at Cycle 3) than those of GR (respectively 63% and 79% at Cycle 2; 91% and 96% at Cycle 3) and GC (respectively 59% and 72% at Cycle 2; 92% and 91% at Cycle 3) as a consequence of its lower percentage reduction of 6-gingerol content (Table 1). Moreover, a previous study confirmed that the antioxidant activity of ginger peel was greater than that of the corresponding peeled rhizome because of its higher content of total polyphenols and 6-gingerol [41].

As shown in Figure 1, the positive correlation between antioxidant and phenolic values in soybean oils with ginger ( $r = 0.94$ ,  $r^2 = 0.88$ ;  $r = 0.97$ ,  $r^2 = 0.94$ ) and turmeric ( $r = 0.99$ ,  $r^2 = 0.97$ ;  $r = 0.96$ ,  $r^2 = 0.92$ ) powders was statistically relevant ( $p < 0.001$ ). Hence, the antioxidant activity linearly increased at increasing concentration of 6-gingerol and curcumin as confirmed by several studies [43,44,55].

**Table 2.** The antioxidant activity of soybean oils with butylated hydroxytolueneas (BHT) as well as ginger and turmeric powders at different oil enrichment cycles <sup>1</sup>.

Parameters	Soybean Oils <sup>2</sup>						
	BHT	GC	GP	GR	TC	TP	TR
<b>Cycle 1</b>							
DPPH (mg TE/g)	0.15 ± 0.01 <sup>e</sup>	3.30 ± 0.17 <sup>c</sup>	3.64 ± 0.33 <sup>c</sup>	3.46 ± 0.15 <sup>c</sup>	1.89 ± 0.29 <sup>d</sup>	7.69 ± 0.38 <sup>b</sup>	8.71 ± 0.28 <sup>a</sup>
FRAP (mg TE/g)	1.32 ± 0.03 <sup>f</sup>	5.76 ± 0.22 <sup>d</sup>	9.80 ± 0.51 <sup>a</sup>	6.83 ± 0.14 <sup>c</sup>	2.06 ± 0.08 <sup>e</sup>	9.03 ± 0.20 <sup>b</sup>	6.98 ± 0.20 <sup>c</sup>
<b>Cycle 2</b>							
DPPH (mg TE/g)	0.15 ± 0.01 <sup>d</sup>	1.34 ± 0.01 <sup>bc</sup>	2.04 ± 0.25 <sup>b</sup>	1.28 ± 0.22 <sup>c</sup>	1.41 ± 0.11 <sup>bc</sup>	3.13 ± 0.39 <sup>a</sup>	2.79 ± 0.37 <sup>a</sup>
FRAP (mg TE/g)	1.32 ± 0.03 <sup>c</sup>	1.59 ± 0.05 <sup>c</sup>	4.32 ± 0.40 <sup>a</sup>	1.43 ± 0.05 <sup>c</sup>	1.44 ± 0.01 <sup>c</sup>	4.09 ± 0.08 <sup>a</sup>	2.41 ± 0.02 <sup>b</sup>
<b>Cycle 3</b>							
DPPH (mg TE/g)	0.15 ± 0.01 <sup>e</sup>	0.25 ± 0.01 <sup>de</sup>	0.79 ± 0.05 <sup>a</sup>	0.30 ± 0.02 <sup>d</sup>	0.63 ± 0.01 <sup>b</sup>	0.44 ± 0.01 <sup>c</sup>	0.70 ± 0.07 <sup>ab</sup>
FRAP (mg TE/g)	1.32 ± 0.03 <sup>b</sup>	0.53 ± 0.02 <sup>e</sup>	2.45 ± 0.05 <sup>a</sup>	0.29 ± 0.01 <sup>f</sup>	0.84 ± 0.01 <sup>c</sup>	0.57 ± 0.01 <sup>e</sup>	0.71 ± 0.01 <sup>d</sup>

<sup>1</sup> Cycle 1: oil enrichment with original powders; Cycle 2: oil enrichment with exhausted and one time recycled powders; Cycle 3: oil enrichment with exhausted and two times recycled powders. <sup>2</sup> BHT: 0.02% *w/w* butylated hydroxytolueneas synthetic reference antioxidant; GC: commercial ginger; GP: freeze-dried ginger peel; GR: freeze-dried ginger rhizome; TC: commercial turmeric; TP: freeze-dried turmeric peel; TR: freeze-dried turmeric rhizome. <sup>a-f</sup> Data are presented as mean ± SD (*n* = 3). Values with different letters within rows are statistically different (*p* < 0.05) as determined by Bonferroni test.



**Figure 1.** Correlation of the antioxidant activity (2,2-diphenyl-1-picrylhydrazyl—DPPH, ferric reducing ability of plasma—FRAP) with the concentration of 6-gingerol and curcumin in soybean oils supplemented with ginger (a) and turmeric (b) powders, respectively.

### 3.3. Oxidative Stability of Ginger- and Turmeric-Supplemented Soybean Oils

In a previous study [56], it has been analyzed the peroxide value (PV) and induction period (IP) for respectively monitoring the primary and secondary phases of lipid oxidation of the same commercial refined soybean oil with/without BHT and ginger and turmeric powders during storage at 62 °C for 28 days. The PV parameter increased slowly in the first 14 days and quickly at the end, while the IP index linearly decreased with increasing days of storage under accelerated oxidation conditions. Moreover, the presence of synthetic and natural antioxidants contributed to significantly decreasing the PV values and increasing the IP values of the soybean oil during thermal storage. In this study, the IP index was considered as reference parameter for not only comparing the oxidative stability of soybean oil samples but also indirectly evaluating the antioxidant effectiveness of ginger and turmeric powders. The averaged IP values were significantly different ( $p < 0.001$ ) among oil samples at each enrichment cycle (Table 3). The ginger- and turmeric-supplemented soybean oils had higher IP values than the control C and the reference antioxidant BHT only at Cycle 1 probably due to a more than 50% percentage reduction of their phenolic contents and antioxidant activities at Cycle 2 and Cycle 3 (Tables 1 and 2, respectively). Generally, the degradation of phenolic antioxidants in oils and fats during thermal storage contributed to the formation of the lipid oxidation-related products [57]. Moreover, some authors showed that the curcuminoids contents in turmeric extracts subjected to different extraction temperatures (60–90 °C) and times (15–180 min) achieved the maximum at 90 °C for 60 min but decreased under extended heating [58]. Hence, phenolic compounds such as 6-gingerol

and curcumin at low concentrations could be more susceptible to thermal degradation during our Rancimat test carried out at 110 °C, thus, contributing to a further increased oxidation rate of ginger- and turmeric-soybean oils, respectively (Table 3). As regards cycle 1, the IP increments of soybean oils supplemented with antioxidant powders were as follows: TR (+5.4 h) > TP (+4.09 h) > GP (+3.5 h) > GR (+2.9 h) > GC (+2.7 h) > TC (+1.4 h) > BHT (+0.33 h). These values were much greater than those of soybean oils containing low (0.1 mg/g) and high (1.50 mg/g) dosage of red chicory extract (+0.8 h and +1.4 h, respectively) at the same experimental conditions of Rancimat instrument [5]. Moreover, they were also higher than those of soybean oils supplemented with 300 mg/kg of olive leaf extract (+0.5 h) [9], 1823 mg/kg of olive leaf extract encapsulated by Arabic gum (+0.4 h) [4] or 1000 mg/kg of protein hydrolyzates isolate from cow's intestine (+2.2 h) [8]. Among oil samples, TR and TP had the highest oxidative stability at Cycle 1, while GP achieved the best performance at Cycle 2 and Cycle 3 as a consequence of their greater phenolic contents (Table 1) and antioxidant activity (Table 2). In this regard, several authors confirmed the high correlation between the oxidative stability of soybean oils added with plant extracts and their phenolic composition [5–15]. Regarding turmeric-supplemented soybean oils, TR and TP showed greater oxidative stability than that of TC only for the first two cycles where achieved the highest contents of total polyphenols and curcumin (Table 1). In this regard, the decrease in the oxidation rate of soybean oil after adding curcumin extracted from turmeric rhizome has been previously observed [7].

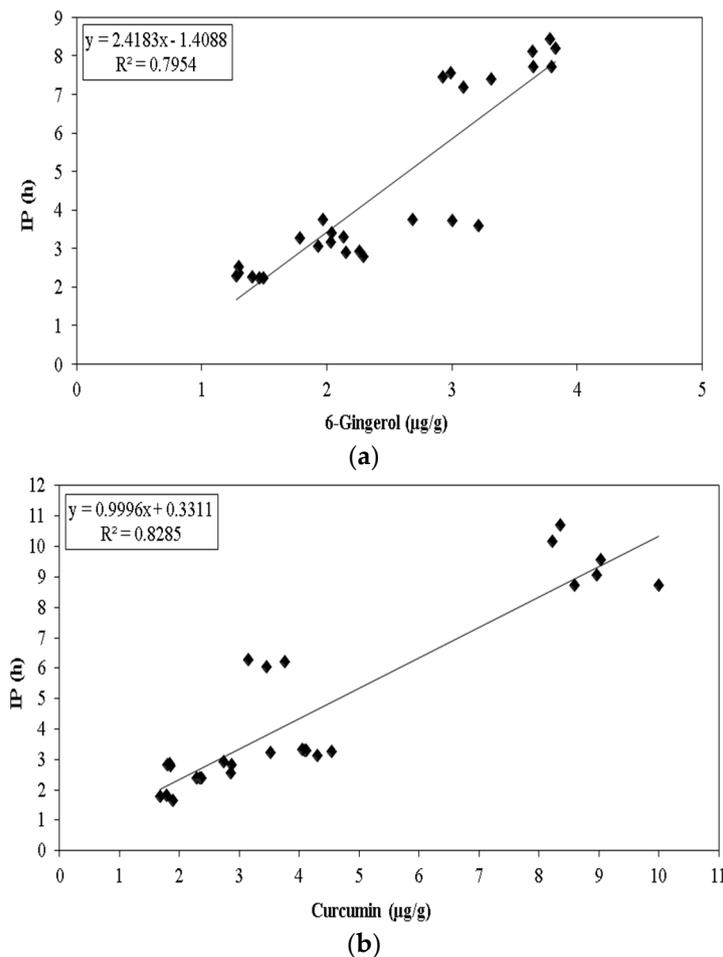
However, the percentage reductions of IP values in TR (68% at Cycle 2 and 72% at Cycle 3) and TP (63% at Cycle 2 and 81% at Cycle 3) were higher than those in TC (56% at Cycle 2 and 62% at Cycle 3) as a consequence of their greater percentage reductions of curcumin content (Table 1) and antioxidant activity (Table 2). Regarding ginger-supplemented soybean oils, GP, which showed the best oxidative stability at each oil enrichment cycle, had lower IP percentage reductions (55% at Cycle 2 and 65% at Cycle 3) than those of GR (58% at Cycle 2 and 68% at Cycle 3) and GC (53% at Cycle 2 and 70% at Cycle 3) as a consequence of its lower percentage reduction of 6-gingerol content (Table 1) and antioxidant activity (Table 2).

As shown in Figure 2, the positive correlation between IP and HPLC values in soybean oils with ginger ( $r = 0.87$ ,  $r^2 = 0.79$ ) and turmeric ( $r = 0.93$ ,  $r^2 = 0.82$ ) powders was statistically relevant ( $p < 0.001$ ). The IP values linearly increased at increasing contents of bioactive compounds.

**Table 3.** The oxidative stability index of soybean oils with/without antioxidant powders at different oil enrichment cycles <sup>1</sup>.

Parameters	Soybean Oils <sup>2</sup>							
	C	BHT	GC	GP	GR	TC	TP	TR
<b>Cycle 1</b>								
IP (h)	4.71 ± 0.10 <sup>f</sup>	5.04 ± 0.36 <sup>f</sup>	7.39 ± 0.19 <sup>d</sup>	8.25 ± 0.16 <sup>cd</sup>	7.60 ± 0.18 <sup>bc</sup>	6.15 ± 0.13 <sup>e</sup>	8.80 ± 0.18 <sup>b</sup>	10.11 ± 0.58 <sup>a</sup>
<b>Cycle 2</b>								
IP (h)	4.71 ± 0.10 <sup>a</sup>	5.04 ± 0.36 <sup>a</sup>	3.50 ± 0.24 <sup>b</sup>	3.70 ± 0.08 <sup>b</sup>	3.17 ± 0.11 <sup>bc</sup>	2.73 ± 0.19 <sup>c</sup>	3.23 ± 0.04 <sup>bc</sup>	3.21 ± 0.10 <sup>bc</sup>
<b>Cycle 3</b>								
IP (h)	4.71 ± 0.10 <sup>a</sup>	5.04 ± 0.36 <sup>a</sup>	2.24 ± 0.02 <sup>d</sup>	2.87 ± 0.07 <sup>b</sup>	2.40 ± 0.12 <sup>cd</sup>	2.36 ± 0.01 <sup>cd</sup>	1.72 ± 0.10 <sup>e</sup>	2.79 ± 0.03 <sup>bc</sup>

<sup>1</sup> Cycle 1: oil enrichment with original powders; Cycle 2: oil enrichment with exhausted and one time recycled powders; Cycle 3: oil enrichment with exhausted and two times recycled powders. <sup>2</sup> C: control without antioxidant addition; BHT: 0.02% *w/w* butylated hydroxytolueneas synthetic reference antioxidant; GC: commercial ginger; GP: freeze-dried ginger peel; GR: freeze-dried ginger rhizome; TC: commercial turmeric; TP: freeze-dried turmeric peel; TR: freeze-dried turmeric rhizome. <sup>a-f</sup> Data are presented as mean ± SD (*n* = 3). Values with different letters within rows are statistically different (*p* < 0.05) as determined by Bonferroni test.



**Figure 2.** Correlation of the oxidative stability index (IP) with the concentration of 6-gingerol and curcumin in soybean oils supplemented with ginger (a) and turmeric (b) powders, respectively.

#### 4. Conclusions

The supplementation of ginger and turmeric powders significantly contributed to the antioxidant properties of soybean oil by providing highly lipid-soluble bioactive compounds such as 6-gingerol and curcumin, respectively. The antioxidant activity and oxidative stability of ginger- and turmeric-supplemented soybean oils non-linearly decreased over cycles but linearly increased with increasing phenolic yields. In addition, the powders obtained by freeze-drying were the most effective in improving the antioxidant properties of soybean oil only at the first enrichment cycle when occurred the maximum phenolic extractability. Hence, ginger and turmeric rhizomes and peels have been valorized as a natural source of antioxidants to be proposed not only as additives/preservatives towards the oil oxidation by replacing synthetic additives but also as functional food ingredients. Moreover, the recovery and recycle of peels meet the current request for more eco-friendly and sustainable production. Further analysis could be performed to deeply investigate the physical and chemical changes such as color, viscosity, fatty acid composition, and the quality parameters for monitoring during storage the formation of the primary and secondary oxidation products in ginger- and turmeric-supplemented soybean oils.

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**Sample Availability:** Samples of the compounds are available from the authors.

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