

## Research Article

# The Vacuum and Light-Avoided Packaging Ameliorate the Decline in Quality of Whole Chili during Storage

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Vacuum packaging is a superior procedure that could maintain the quality of dried red peppers for a relatively longer period, while the effect of light avoidance was inconsistent. Therefore, this study was to evaluate the effect of vacuum in combination with light avoidance on the storage quality of peppers and to investigate vacuum packaging and vacuum and light-avoided packaging influence the quality characteristics of dried chili peppers during storage. The results indicated that the quality characteristics of peppers gradually deteriorated: redness, extractable color, capsanthin, capsaicinoids, pungency, total phenolic content, and antioxidant capacity decreased, whereas moisture, water activity, and browning index increased as storage progressed. Vacuum packaging inhibited these changes compared with the control. Vacuum and light-avoided packaging further decelerated the quality loss of dried peppers, with the minimum changes in color, pungency, and antioxidant capacity of dried peppers during storage, and these could be due to lower water activity by light avoidance. Therefore, light avoidance may be necessary to further delay the deterioration of dried peppers under vacuum, and vacuum and light-avoided packaging could better retain the quality of dried chili peppers.

## 1. Introduction

Dried red peppers, the dehydrated and storable fruit of certain varieties of red pepper (*Capsicum annuum*), are extensively used in cuisines and the food industry throughout the world as a natural food colorant or a seasoning agent due to their intense red color, pungency, and aroma. China is one of the main producers and consumers of dried peppers in the world, with over 300,000 tons of production in 2018 [1]. In recent years, the increasing requirement for dried peppers has been attributed to the beneficial effects of red peppers on human health due to their high antioxidant content and bioactive constituents [2–4].

The color and pungency of dried peppers are considered to be the main quality parameters in the trade. The intense red color of dried peppers is due to the presence of carotenoid pigments, mainly capsanthin and capsorubin, while the yellow-orange color is mostly derived from xanthophylls (zeaxanthin, violaxanthin, antheraxanthin,  $\beta$ -cryptoxanthin, and capsolutein) and  $\beta$ -carotene [5]. These carotenoids have

been shown to be antioxidants and have important health benefits in inhibiting cancer cells and alleviating various diseases [6, 7]. Pungency is a specific taste of red peppers and is attributed to capsaicinoids. The primary capsaicinoids in red peppers are capsaicin and dihydrocapsaicin, which account for the majority (~90%) of spice pungency [8]. Capsinoids possess the biological properties of being antioxidant, antitumoral, and antiobesity, and are useful in relieving pain and preventing cardiovascular and gastrointestinal diseases [5, 9].

The color, pungency, and phytochemical composition of peppers and their products are susceptible to being affected by light, temperature, moisture, and oxygen during post-harvest and storage [10–14], which work together to cause deterioration through breaking bonds, redox reactions, etc. [15]. Various packaging materials and methods are employed to preserve quality characteristics and to extend the shelf life of products. Compared with packing in jute bags, wrapping with low-density polyethylene bags could retard the disappearance rates of capsaicinoids and

antioxidants in dried chili peppers [16]. Vacuum packaging in polythene bags could maintain the quality of whole chilies for up to 24 months, especially under cold storage [17]. Paprika packed in CO<sub>2</sub> and under vacuum had higher capsanthin concentrations and extractable color (American Spice Trade Association, ASTA) values [18]. Modified atmospheric packaging (MAP) could retain the stable content of phenolic compounds in peppers [19], and MAP and vacuum packaging with PP under refrigerated conditions could maintain the texture, color, ascorbic acid, and marketability of fresh bell peppers for 20 days [20]. In addition, light exposure could induce oxidation of carotenoids and antioxidants acting as a catalyst, but it had inconsistent effects on the peppers' quality in the complete absence of air. Chetti et al. [17] found the loss of color in chili packed in vacuum bags was relatively lower under darkness than under light, while light has few effects on carotenoids and capsaicinoids under vacuum [21, 22].

Vacuum packaging has been found to be a superior technology in retaining the quality of dried red peppers, while the effect of light avoidance still needs to be evaluated in the complete absence of air. Therefore, this study investigated the effects of vacuum and vacuum in combination with light avoidance on the quality of dried chili peppers during storage.

## 2. Materials and Methods

**2.1. Sample Preparation.** The Erjingtiao chili pepper (*C. annuum* L.) is a typical representative of Sichuan red peppers and one of the important spices in Sichuan cuisine, with higher colorant power and lower pungency [23]. The fresh Erjingtiao chili pepper was purchased from the local market (Chengdu, China) and was followed by washing, oven-drying (50°C drying oven with aeration, ZFD-7600, North-South Instrument and Equipment, Zhengzhou, China) the products until they reached a constant weight.

Dried chili peppers weighing about 6 kg without mechanical damage, pests, or diseases were chosen and divided into three equal groups. All groups were filled in each pouch with 250 g samples and packed with a packing machine (DZ-800/2S, Xukang, Zhucheng, China). The first group was sealed under normal atmospheric conditions with polyamide/polyethylene films (C), the second was vacuum packed with polyamide/polyethylene (V), and the last was vacuum packed with light-avoided material aluminum polyethylene (V + LA). The samples were stored at 25 ± 2°C and RH of 70–80% for 0, 1, 2, and 6 months, and physicochemical parameters, color, pungency, and antioxidant capacity were measured in triplicate at each storage.

**2.2. Physicochemical Analysis.** The moisture content was determined by drying a known weight of sample to a constant weight in an oven drier (ZFD-7600, Nanbei Instrument, Zhengzhou, China), and water activity ( $a_w$ ) was analyzed by a water activity meter (HD-3A, Huake, Wuxi, China) [23]. The browning index (BI) was estimated as described by Topuz et al. [24] with slight modification. The

chili powder (0.1 ± 0.001 g) was accurately weighed and mixed with 50 mL of distilled water. Then, the mixture was placed in a thermostatic water bath shaker (SHY-A, Chenghui, CHangzhou, China) at 25°C and 140 rpm for 2 h, and centrifuged at 8000 rpm for 8 min. The supernatant was collected and passed through a 0.45 μm PTFE needle filter, and the absorbance value was measured at 420 nm in a UV-Visible spectrophotometer (UV755B, Youke, Shanghai, China).

**2.3. Color Measurement.** The instrumental color of pepper was measured by a CR-400 colorimeter (Minolta Camera Co., Ltd., Osaka, Japan). Before testing, standard white and black plates were used for calibration. The  $L^*$  value (lightness),  $a^*$  value (redness), and  $b^*$  (yellowness) were recorded.

A 0.1 ± 0.001 g of chili powder was extracted with 25 mL of acetone at 25°C and 400 rpm for 3 h. The extract was filtered and diluted 5 times with acetone, and the absorbance values were measured by spectrophotometry at 460 nm. ASTA values were analyzed and calculated according to Topuz et al. [24].

**2.4. Capsanthin Content.** The capsanthin content in chili powders was analyzed as per the method of Kim et al. [4] with minor modifications. A 0.1 ± 0.001 g sample was extracted with 10 mL of acetone under ultrasonication for 2 h at 25°C. The mixture was centrifuged at 8000g for 5 min, and the supernatant was filtered through a 0.45 μm nylon needle filter. Capsanthin analysis was performed using the LC-20A HPLC system (Shimadzu Co., Ltd., Tokyo, Japan). The Shimadzu Shimpack-C18 column (250 mm length × 4.6 mm, 5 μm particle size) was also used. Other conditions were performed based on the method of Kim et al. [4]. The capsanthin standard (ZZ Bio-technology Co., Ltd., Shanghai, China) was dissolved by acetone, and gradient dilution was performed to produce the standard regression equation. The results were converted to dry basis content by moisture content compensation.

**2.5. Capsaicinoids Content and Pungency.** The extraction of the capsaicin and dihydrocapsaicin in paprika powders was carried out according to De Aguiar et al. [8] with slight modification. A 0.1 ± 0.001 g sample was extracted with 10 mL of 80% aqueous methanol by ultrasonication for 2 h. The mixture was centrifuged at 8000 rpm for 5 min, and the supernatant was filtered through a 0.45 μm nylon needle filter. Capsaicinoids were analyzed by a LC-20A HPLC system (Shimadzu Co., Ltd., Tokyo, Japan) equipped with the Shimadzu Shimpack-C18 column (250 mm length × 4.6 mm, 5 μm particle size) set to 360 nm and 35°C. The mobile phase consisted of acetonitrile (A) and water (B) at a flow rate of 1.0 mL/min. The injection volume was 20 μL. The chromatographically pure grades of capsaicin and dihydrocapsaicin standards (Must Bio-technology Co., Chengdu, China) were dissolved by chromatography in methanol and then subjected to gradient dilution to produce the standard regression equation. Pungency was expressed

as Scoville Heat Unit (SHU) and was calculated by the method of Scoville [25].

**2.6. Total Phenolic Content.** The total phenolic content (TPC) of paprika powders was determined spectrophotometrically using the Folin-Ciocalteu method, according to De Aguiar et al. [8] with slight modification. 0.5 mL of 60% aqueous methanol and 1 mL of Folin-Ciocalteu reagent (0.25 mol/L) were added to 0.5 mL of 60% aqueous methanol extract. After 3 min, 2.0 mL of sodium carbonate solution (15%) was added. The mixture was kept in the dark at room temperature for 2 h, then centrifuged at 8000 rpm for 5 min, and the absorbance of the supernatant was measured at 760 nm.

**2.7. DPPH and ABTS Radical Scavenging Rate.** The DPPH (2,2-Diphenyl-1-picrylhydrazyl) radical scavenging rate was determined according to a previous study with optimization [26]. The 4 mL of filtered 60% aqueous methanol extract was mixed with 2 mL of 0.2 mmol/L DPPH ethanol solution. After mixing, it was left in the dark for 30 min. The mixture of absorbance was measured at 517 nm.

The ABTS (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulphonate)) radical scavenging rate was measured as described in a previous study with minor modifications [27]. ABTS reaction solution was mixed with 7 mmol/L aqueous ABTS solution and 2.45 mmol/L potassium persulfate solution (1 : 1) and was kept for 12–16 h in the dark. Dilute with anhydrous ethanol and adjust the absorbance value at 732 nm to  $0.70 \pm 0.02$ . The 0.1 mL of filtered 60% aqueous methanol extract was mixed with 2.9 mL of 60% methanol aqueous solution and 3 mL of ABTS reaction solution, then it was left in the dark for 30 min. The mixture of absorbance was measured at 734 nm. The scavenging rate of DPPH and ABTS was calculated as the following equation:

$$R = \left( 1 - \frac{A_1 - A_2}{A_0} \right) \times 100\%, \quad (1)$$

where,  $R$  means the scavenging rate of DPPH or ABTS,  $A_1$  is the absorbance value of sample with DPPH or ABTS reaction solution,  $A_2$  is the absorbance value of sample with anhydrous ethanol, and  $A_0$  is the absorbance value of 60% aqueous methanol with DPPH or ABTS reaction solution.

**2.8. Total Antioxidant Capacity.** The total antioxidant capacity (TAC) of chili pepper was evaluated by referring to the instructions of the total antioxidant capacity kit with the FRAP (ferric reducing antioxidant power) method. Mix 5  $\mu$ L supernatant of the 60% aqueous methanolic extract after centrifugation and the series concentration standard solution with 180  $\mu$ L of FRAP working solution, respectively. The OD value was measured at 593 nm. A calibration curve was prepared with the series standard. The results were expressed as mol/g, i.e., the antioxidant capacity per gram of the sample was equivalent to the moles of ferrous sulfate heptahydrate.

**2.9. Statistical Analysis.** All measurements were performed in triplicate, and data were expressed as mean  $\pm$  standard deviation. All results were analyzed in a completely randomized design using SPSS software version 25.0 (Chicago, IL, USA). One-way analysis of variance (ANOVA) was performed for physicochemical properties, color, pungency, and antioxidant capacity, with the measured variables as dependent variables, different packaging or storage time as fixed effects, and replicate as a random effect. Fisher's least significant difference (LSD) test was used to determine the statistical significance among the means at a significance level of  $P < 0.05$ .

### 3. Results and Discussion

**3.1. Physicochemical Properties.** Table 1 shows that there are increases in  $a_w$ , moisture content, and BI in all dried chili pepper batches as storage progressed. Among them, the  $V$  and  $V + LA$  groups inhibited the rise of moisture ( $P < 0.05$ ). These were similar to previous study, Chetti et al. [17] found that the moisture content of vacuum-packed stored peppers did not vary much over a 24 m period, while the moisture content of jute bag-stored peppers fluctuated more. These could be related to the residual water and oxygen in packaging. Vacuum packaging is sealed packages with less atmosphere and oxygen level, thus reducing the moisture and  $a_w$ , especially under light avoidance. In this study,  $V + LA$  group had significantly lower  $a_w$  than  $C$  and  $V$  during storage. In agreement with these results, Udomkun et al. [28] found that the moisture content and  $a_w$  of dried papaya packed in polyamide/polyethylene films with 95% light transmissivity was significantly higher than that of aluminum laminated polyethylene film with 0% light transmissivity during storage for 1 to 9 months. These may be associated with the water vapor generated by the light exposure, which increased  $a_w$  of the samples in the package [29, 30].

In this study, the BI of the  $V + LA$  group was significantly lower than that of the  $C$  at 6 m ( $P < 0.05$ ). Similarly, Ahad et al. [31] found that the nonenzymatic browning values were higher in walnut kernels without vacuum than under vacuum. Tomato paste stored in the light had a higher BI than that stored in the dark [32]. It may be mainly attributed to the higher moisture and  $a_w$  in the package, which increases the intermolecular freedom and accelerates the oxidation reaction [33].

**3.2. Color Traits.** Capsanthin is the key substance in making chili peppers attractive red. The content of capsanthin and ASTA values are critical indexes to evaluate the color and storage quality of chili peppers [34–36]. However, a gradual discoloration [24], significantly decreases in  $a^*$ , ASTA values and capsanthin content [18, 20, 24] were observed in all paprika samples during storage. Similar to the previous studies, this study found a general increase in  $L^*$  and  $b^*$ , while  $a^*$ , capsanthin content, and ASTA values decreased with storage time in all groups (Figures 1 and 2). The reduction of  $a^*$  and ASTA values may be due to the oxidative

TABLE 1: Changes in  $a_w$ , moisture content, and BI of dried peppers during storage.

	Months	C	V	V + LA
$a_w$	0	0.359 ± 0.001 <sup>Aa</sup>	0.359 ± 0.001 <sup>Aa</sup>	0.359 ± 0.001 <sup>Aa</sup>
	1	0.410 ± 0.001 <sup>Ba</sup>	0.407 ± 0.002 <sup>Ba</sup>	0.374 ± 0.001 <sup>Bb</sup>
	2	0.428 ± 0.001 <sup>Ca</sup>	0.422 ± 0.002 <sup>Ca</sup>	0.387 ± 0.004 <sup>Cb</sup>
	6	0.492 ± 0.003 <sup>Da</sup>	0.450 ± 0.004 <sup>Db</sup>	0.404 ± 0.001 <sup>Dc</sup>
Moisture content (%)	0	10.00 ± 0.01 <sup>Aa</sup>	10.00 ± 0.01 <sup>Aa</sup>	10.00 ± 0.01 <sup>Aa</sup>
	1	10.28 ± 0.18 <sup>Ba</sup>	10.06 ± 0.17 <sup>Aa</sup>	10.03 ± 0.24 <sup>Aa</sup>
	2	10.50 ± 0.06 <sup>Ca</sup>	10.14 ± 0.21 <sup>Ab</sup>	10.08 ± 0.16 <sup>Ab</sup>
	6	15.30 ± 0.19 <sup>Da</sup>	10.28 ± 0.20 <sup>Ab</sup>	10.19 ± 0.16 <sup>Ab</sup>
BI	0	0.193 ± 0.005 <sup>Aa</sup>	0.193 ± 0.005 <sup>Aa</sup>	0.193 ± 0.005 <sup>Aa</sup>
	1	0.202 ± 0.004 <sup>ABa</sup>	0.198 ± 0.002 <sup>ABa</sup>	0.197 ± 0.001 <sup>Aa</sup>
	2	0.204 ± 0.005 <sup>ABa</sup>	0.200 ± 0.006 <sup>ABa</sup>	0.199 ± 0.001 <sup>ABa</sup>
	6	0.206 ± 0.006 <sup>Ba</sup>	0.202 ± 0.003 <sup>Bab</sup>	0.201 ± 0.001 <sup>Bb</sup>

C: control with polyamide/polyethylene film; V: vacuum packaging with polyamide/polyethylene film; and V + LA: vacuum and light-avoided packaging with aluminum polyethylene film.  $a_w$ : water activity; BI: browning index. <sup>A-D</sup> represent the significant difference ( $P < 0.05$ ) of dried pepper stored at different period. <sup>a-c</sup> represent significant differences ( $P < 0.05$ ) of dried pepper stored in different packaging.

degradation of capsanthin and capsorubin by light, temperature, humidity, and other factors [37, 38]. Capsanthin and capsorubin may react with active oxygen to promote the breaking of their conjugated double bonds, resulting in epoxides and peroxides that cause the discoloration of peppers [15, 39, 40], while capsanthin could be transformed into orange or yellow pigments in the process of photo-degradation, such as zeaxanthin, lutein,  $\beta$ -carotene, and other carotenoid substances [41, 42]. Moreover, the oxidative degradation process of yellow carotenoids is relatively slow [6]. Therefore,  $a^*$  of dried peppers was decreased while  $b^*$  increased during storage.

Figures 1 and 2 show that the V and V + LA groups inhibited the decline in  $a^*$ , ASTA values, and capsanthin content as well as the increase in  $L^*$  and  $b^*$  values during storage, compared with the C group. Among them, the V + LA group had the highest  $a^*$ , ASTA values, and capsanthin content throughout the storage period, but the lowest  $L^*$  and  $b^*$  values. This packaging could inhibit the degradation of capsanthin and other carotenoids through isolating them from oxygen, moisture, and light and better maintain  $a^*$  and ASTA values. These were similar to previous studies. Ordonez-Santos et al. [18] found paprika packed in CO<sub>2</sub> and under vacuum had higher capsanthin concentrations and ASTA values in both types of samples. The whole chili in vacuum-packed bags had higher ASTA values compared with jute bags, and the loss of color was relatively higher in vacuum-packed bags under light exposure [17]. However, light has little effect on carotenoids in the complete absence of air [21].

### 3.3. Capsaicin, Dihydrocapsaicin Content, and Pungency.

Capsaicin and dihydrocapsaicin are the two primary capsaicinoids, accounting for approximately 90% of the capsaicinoids in chili peppers, which are responsible for the majority of spice pungency [43–45]. The content of capsaicin and dihydrocapsaicin as well as pungency in all treatment groups were stable in the first 2 months, then gradually reduced, and the capsaicinoids content and pungency at 6 m in all groups were significantly lower than at 0 m (Figure 3,

$P < 0.05$ ). Previous studies found similar results: capsaicinoids had relatively stable properties, and the levels of capsaicin and dihydrocapsaicin were essentially maintained for 90 days [16], but capsaicin content started declining from the sixth month [17].

The anaerobic environment of vacuum packaging is conducive to inhibiting the loss of capsaicinoids' content and pungency. Peppers stored in nitrogen or vacuum had higher capsaicinoids content than those stored in oxygen [17, 46], and the disappearance rate of capsaicinoids and antioxidants was lower in peppers packed in LDPE (low-density polyethylene bags) that blocked oxygen and water vapor than in jute bags [16]. Similar to the previous studies, our study found the vacuum packaging delayed the loss of capsaicin, dihydrocapsaicin content and pungency in dried chili peppers during storage, especially at 6 m, and their level in V + LA was significantly higher than that in V (Figure 3). The above data indicated that light avoidance could decelerate the loss of color, capsaicinoids, and pungency of dried peppers under vacuum. However, this was inconsistent with some studies. Schweiggert et al. [22] reported that there was no difference in the stability of capsaicinoids in chili powders between storage under light exposure and in the dark. Chetti et al. [17] also found light had little effect on the degradation of capsaicinoids, while it increased the loss of color in the complete absence of air. This discrepancy could be related to the higher stability of capsaicinoids compared with carotenoid pigments. Furthermore, the moisture content and  $a_w$  in dried peppers should be considered: there was no significant difference in moisture content between light and dark treatments [17], whereas the V + LA group had lower  $a_w$  than V in this study, which may inhibit the degradation of carotenoids and capsaicinoids.

### 3.4. Total Phenolic Content and Antioxidant Capacity.

The red pepper contains a number of substances with antioxidant properties, such as carotenoids, capsaicinoids, phenolics, flavonoids, and vitamins [12, 47, 48], which play an important role in protecting against oxidative damage caused by free radicals [38, 49] and are essential for human

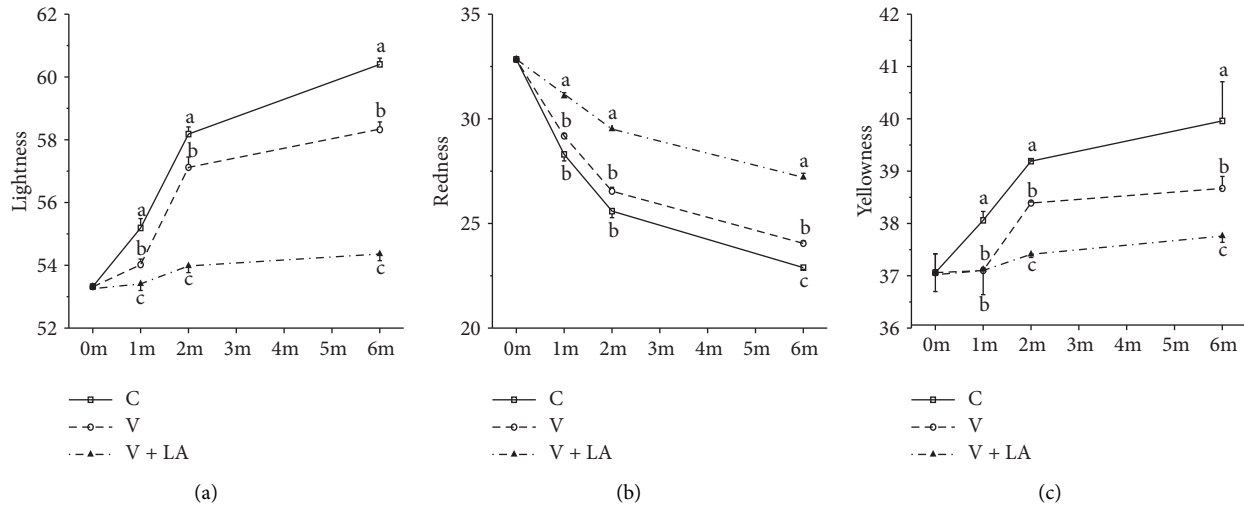


FIGURE 1: Effect of different packaging on lightness, (a) redness, (b) and yellowness (c) of the dried whole chili during storage. C: Control with polyamide/polyethylene film; V: vacuum packaging with polyamide/polyethylene film; V + LA: vacuum and light-avoided packaging with aluminum polyethylene film. a–c represent the significant difference ( $P < 0.05$ ) of dried pepper stored in different packaging.

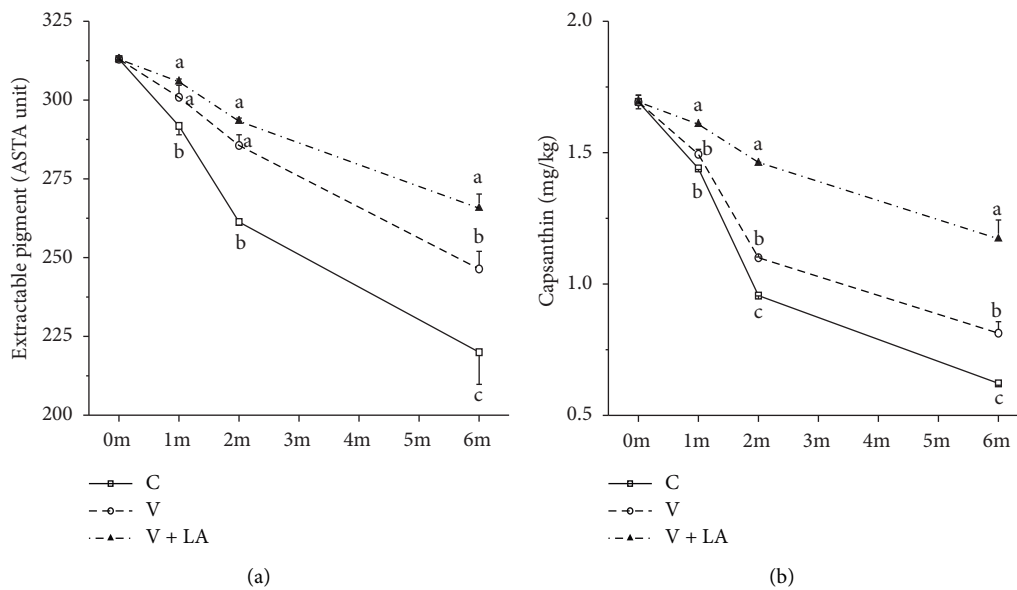


FIGURE 2: Influence of different packaging on ASTA unit (a) and capsanthin content (b) of the dried whole chili during storage. C: control with polyamide/polyethylene film; V: vacuum packaging with polyamide/polyethylene film; V + LA: vacuum and light-avoided packaging with aluminum polyethylene film. ASTA: American Spice Trade Association. a–c represent the significant difference ( $P < 0.05$ ) of dried pepper stored in different packaging.

health [2]. However, the carotenoids, phenolic compounds, and other antioxidants in chili peppers are susceptible to degrading by oxygen, light, temperature, moisture, and other factors during storage [10, 17, 18], which causes the reduction of antioxidant capacity [50]. Consistent with these studies, Figure 4 indicated that the TPC, TAC, DPPH, and ABTS scavenging capacity in peppers decreased significantly with the prolonging of storage time. The V + LA and V groups inhibited their decline compared to the C group, and the V + LA packaging had the best effect, with the highest TPC, TAC, DPPH, and ABTS scavenging capacity at 6 m

compared with other groups ( $P < 0.05$ ). These were similar to previous studies. Ornelas-Paz et al. [19] found that the content of phenolic compounds remained stable in the modified atmosphere packaging of peppers; vacuum-packed walnuts had higher TPC, DPPH, and ABTS scavenging capacities than other packaging [31]. In addition, the TPC and DPPH scavenging capacities of radish microgreens were not significantly affected by light exposure for 16 days, but they tended to have higher levels under dark storage conditions [51]. Thus, vacuum and light-avoided packaging could better protect phenolics from oxidation and

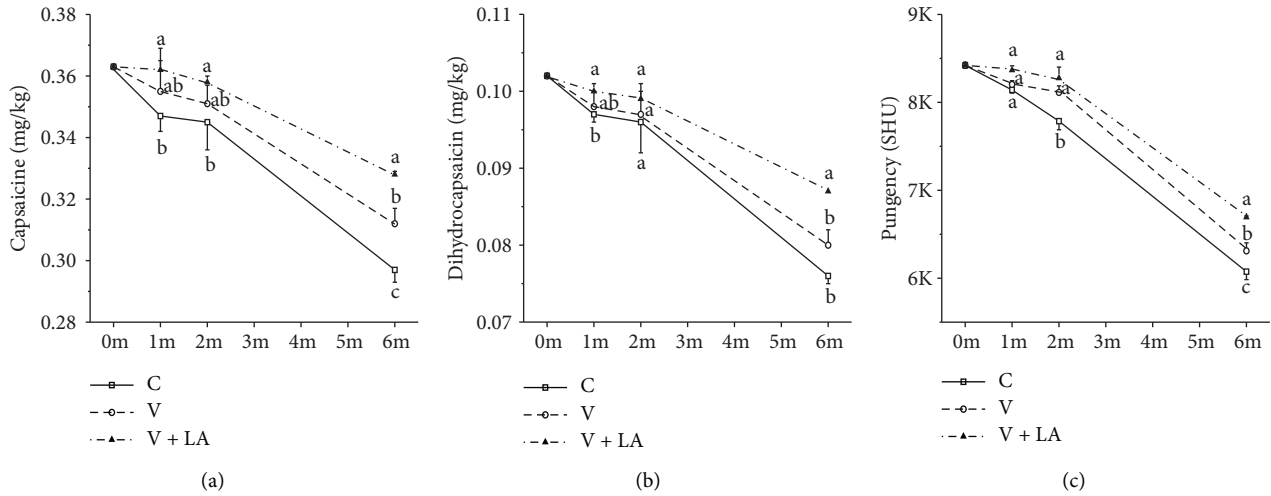


FIGURE 3: Effect of different packaging on capsaicin content, (a) dihydrocapsaicin content, (b) and pungency (c) of the dried peppers during storage. C: control with polyamide/polyethylene film; V: vacuum packaging with polyamide/polyethylene film; V + LA: vacuum and light-avoided packaging with aluminum polyethylene film. SHU: Scoville Heat Unit. a-c represent the significant difference ( $P < 0.05$ ) of dried pepper stored in different packaging.

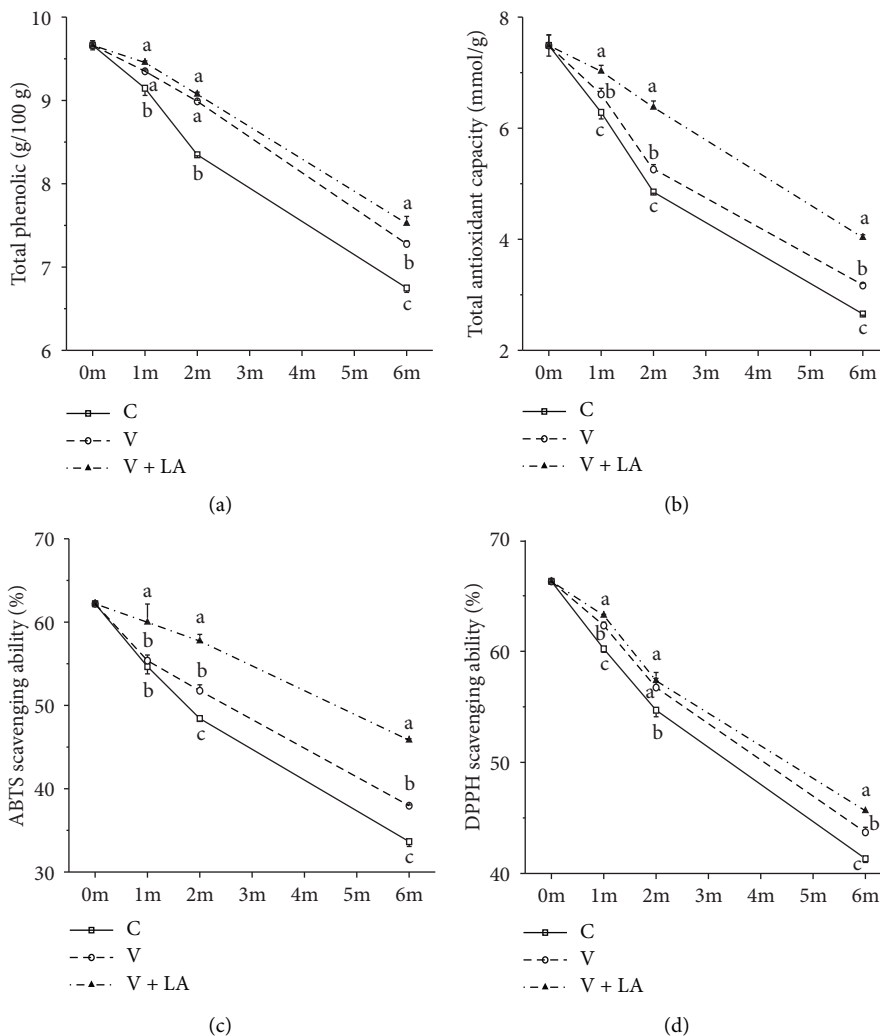


FIGURE 4: Effect of different packaging on total phenolic content, (a) total antioxidant capacity, (b) ABTS, (c) and DPPH (d) scavenging activity of the dried peppers during storage. C: control with polyamide/polyethylene film; V: vacuum packaging with polyamide/polyethylene film; V + LA: vacuum and light-avoided packaging with aluminum polyethylene film. a-c represent the significant difference ( $P < 0.05$ ) of dried pepper stored in different packaging.

degradation due to environmental factors during long-term storage.

#### 4. Conclusion

In summary, the quality characteristics of dried chili peppers gradually declined: color, pungency, and antioxidant capacity decreased, whereas moisture, water activity, and browning index increased as storage progressed. Vacuum packaging ameliorated the deterioration of quality characteristics, and vacuum and light-avoided packaging further reduced the quality loss with the least percent changes in color, pungency, antioxidant capacity, and water activity of dried peppers during storage. Hence, the vacuum combined with light-avoided packaging was considered superior to vacuum packaging in terms of quality parameters, and it was shown to be an effective method for delaying the deterioration of dried chili peppers during storage.

#### Data Availability

The data used to support the findings of this study are included within the article.

#### Conflicts of Interest

The authors declare that they have no conflicts of interest.

#### Authors' Contributions

Investigation, methodology, and writing of the original draft were carried out by Qi Chen. Methodology, data curation, reviewing, and editing were performed by Wensheng Qi. Methodology and formal analysis were carried out by Jiakuan Peng. Renyong Tang supervised the study. Dayu Liu validated the study. Conceptualization, project administration, and reviewing were performed by Xiulan Guo. Qi Chen and Wensheng Qi contributed equally to this work.

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