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Analysis of internal browning of roasted hazelnuts

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Abstract

Formation of internal browning in hazelnuts during roasting and effect of water activity on internal browning were studied. Water activity significantly affected color attributes of roasted hazelnuts. As the water activity increased internal browning also increased. Over the water activity range of study (0.29–0.83), water activity of 0.29 was found to be the least susceptible to internal browning. Roasted samples, having dark-coloured internals contained significantly more total sugar and sucrose than roasted samples, having light coloured internals. Brown centres contained more total sugar and sucrose than light-coloured outer layers. Inner layers of raw hazelnuts contained about 50% more total sugar and sucrose than light-coloured outer layers. Polyphenoloxidase activity could not be detected in the sample roasted at 130°C for 16 and 30 min. These results indicated that internal browning during roasting was due to localised concentration of reactants and, subsequently, the higher non-enzymatic browning reaction rate in the centre of hazelnuts. © 2001 Published by Elsevier Science Ltd.

1. Introduction

Turkey ranks first in hazelnut production with an amount of about 600,000 ton per year, followed by Italy, USA, and Spain. Total export revenue of Turkey from hazelnut and its products is about one billion US$ annually (Akova, 1998).

Roasting is the main step of hazelnut processing. Flavour, colour, texture and appearance of nuts are significantly enhanced by the roasting process. The resulting product is delicate, uniquely nutty and favoured more than raw nuts. Roasting also removes pellicles of hazelnut kernels, inactivates enzymes, and destroys undesirable microorganisms and food contaminants (Atakan & Bostan, 1998; Buckholz, Duan & Stier, 1980; Hashim & Chaveron, 1996; Köksal & Okay, 1996; Mayer, 1985; Özdemir & Devres, 2000a,b; Pattee, Giesbrecht & Islieb, 1995; Richardson & Ebrahem, 1996; Sanders, Vercelotti, Blankenship, Crippen & Civile, 1989).

Colour development, textural changes and aroma formation during roasting are mainly related to drying and non-enzymatic browning (Buckholz et al., 1980; Mayer, 1985; Moss & Otten, 1989; Perren & Escher, 1996a,b). Nonenzymatic browning is a reaction between carbonyl group of reducing sugars and free, uncharged amine groups of amino acids or protein with the loss of one mole of water. The reaction, however, causes a decrease in nutritive value of foods and creates antinutritional properties (Ames, 1988; Jinap, Wan-Rosli, Russly & Nordin, 1998; Labuza & Braisier, 1992; Nicoli, Elizalde, Pitotti & Lerici, 1991; O'Brien & Morrissey, 1989; Troller, 1989). The reaction is dependent on temperature and water activity (a_w) of the food as well as compositional factors, such as concentration and ratio of sugar/amino acids and pH. Compounds such as sulphite, cyanide and mercaptans that might inhibit the reaction, also influence the reaction (Ames; Driscoll & Madamba, 1994; Göğüş, Wectzichha & Lamb, 1998; Labuza & Braisier, 1992; Rapusas & Driscoll, 1995; Saguy & Karel, 1980; Warmbeir, Schnickels & Habuza, 1976).

Colour is an important quality attribute of the dehydrated foods for consumers (Driscoll & Madamba, 1994). Hazelnut is roasted to have a desired product with a range of colours: white, pale-yellow, golden-yellow, dark, and very dark (Özdemir & Devres, 2000a). However, there is problem of internal browning (development of a darker colour inside kernels, namely brown centres, compared to outside colour of the kernels, during roasting). The internal browning makes the product unpleasant for

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consumers, especially when the products are consumed as whole-kernels (Özdemir & Devres, 2000a). Moreover, it leads to underestimation of colour of hazelnut meal, from the outside colour when whole kernels are used for monitoring of roasting degree (Özdemir and Devres, 2000a,b). Internal browning during roasting was also reported for almonds and pecans (King, Halbrook, Fuller & Whithand, 1983), and macadamia nuts (Prichavudhi & Yamamoto, 1965). Hadorn, Keme, Kleinert and Zürcler (1977) also observed internal browning in raw hazelnuts and they related this to the polyphenoloxidase activity. King et al. (1983) reported the possible role of mould activity in internal browning by hydrolysis of protein and carbohydrates to their monomers at high moisture contents. Prichavudhi and Yamamoto (1965) related internal browning in the macadamia nut with an increase in reducing sugar content, through enzymatic hydrolysis of non-reducing sugars at high temperatures (above 37°C) during the initial stages of drying.

Although kinetics of colour development during hazelnut roasting have been studied previously (Özdemir & Devres, 2000a,b), there is no information about formation of internal browning and the effect of water activity on the colour development of hazelnuts during roasting. Therefore, in this study, total sugar, sucrose and reducing sugar contents, and polyphenoloxidase activity were determined in raw and roasted samples so as to characterise internal browning, over a water activity range of 0.29–0.83, using the major Turkish hazelnut variety, Tombul. Also the effect of water activity on colour attributes of roasted hazelnuts was investigated.

2. Material and methods

2.1. Sampling and chemical analysis

Sun-dried hazelnuts (season 1998) were supplied from the Hazelnut Research Center (Giresun, Turkey) at the beginning of the harvest season and kept at 4°C until used. The major Turkish variety, Tombul, was used in the study. The samples were cracked using a modified laboratory scale grain miller. Samples were sorted by size and 9–11 mm hazelnut were used in the experiments. The outer layer (thickness of 2–3 mm) of raw hazelnuts and the inner part were analyzed for protein, total oil, total sugar, sucrose and reducing sugar contents separately according to AOAC (1990).

Equilibration of moisture of raw hazelnut kernels was done at 4±0.1°C for 40 days over saturated salt solutions in airtight glass jars, containing 30 g salt and 150 g sample. The jars were then stored at 20±0.1°C for 4 months or until development of a visual microbial growth. After storage, water activity of the raw samples was measured with a water activity instrument (Novasiva, Switzerland), and sugar composition of the samples was determined. Then, the each sample was roasted at 130°C for 30 min, using a forced air pilot scale roaster (Pasillac, APV, UK). After cutting in two the kernels of each roasted sample, they were separated visually as dark-coloured internal centres and light-coloured internal centers. Sugar composition of kernels with dark-coloured internal centres and with light-coloured internal centres was determined for the each sample.

Moreover, the sugar compositions of brown-centre and outer light layers of samples with initial water activity of 0.74 (widely used by the industry for roasting) were also determined using the Lane–Eynon method (AOAC, 1990). Polyphenoloxidase activity in the sample with initial water activity of 0.74 was also measured before and after roasting at 130°C for 16 min and 30 min, as described by Bonvehi and Rousa (1996). Absorbance values obtained by spectrophotometric measurements were used as indications of the enzymatic activity.

2.2. Color measurements

All colour measurements were conducted within 10 days of roasting experiments. The measurements were performed after hand blanching the samples to remove skins. The poor quality hazelnuts were also removed. The colour of all samples was measured using a Minolta Chroma Meter II Reflectance system (Model CR300, Japan). The instrument is a tristimulus colorimeter which measures four specific wavelengths in the visible range, specified by the CIE (Commission Internationale de l'Esclairage).

Outside colours of the 10 randomly selected hazelnut kernels were measured for each sample and referred to as whole-kernel measurements throughout the manuscript. For determination of meal colour, roasted samples were milled to constant particle size. Then the measurements, referred to as ground-state measurements throughout the manuscript, at five different parts of the resulting sample, were conducted. Moreover, 10 randomly-selected hazelnuts were cut into two at the centre and color of the centre of pieces was measured (referred to as cut-kernel measurements throughout the manuscript).

2.3. Statistical analysis

Analysis of variance (ANOVA) and Duncan-test were conducted using SPSS (ver 5.0) statistical analysis at probability of significance (P) lower than 0.05.

3. Results and discussion

3.1. Effect of water activity on colour attributes of roasted hazelnuts

Effect of water activity on L-, a and b-value for the three measurement methods were shown in Fig. 1.

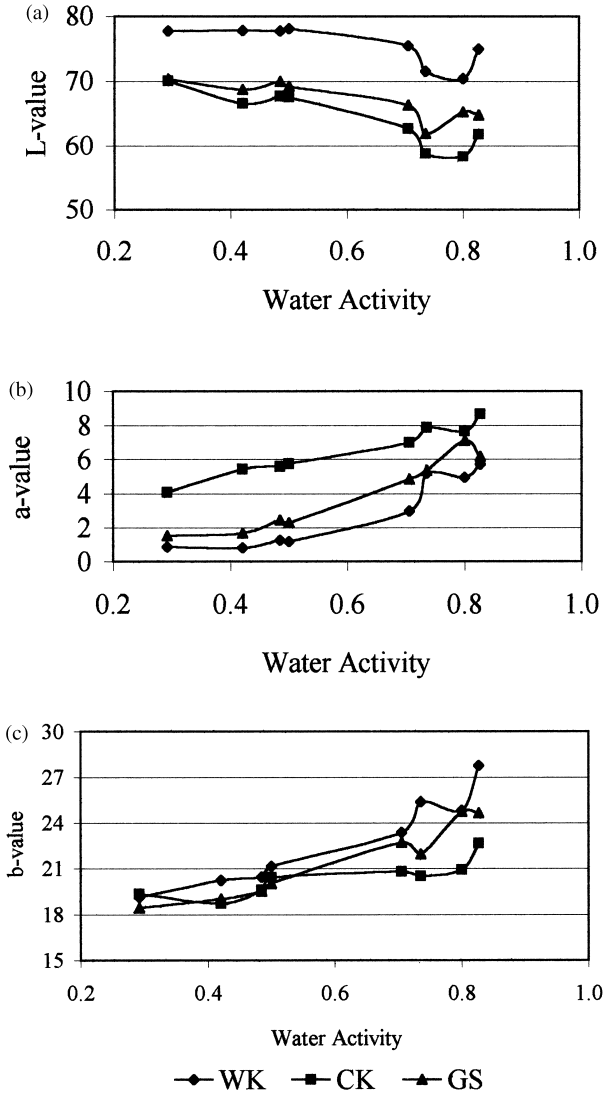

Fig. 1. Effect of water activity on (a) *L*-value, (b) *a*-value and (c) *b*-value of roasted hazelnuts for whole-kernel (WK), cut-kernel (CK) and ground-state (GS) measurements.

ANOVA indicated that there was a significant difference between water activities for *L*-, *a*- and *b*-value ($P < 0.0001$) as shown in Table 1. As the water activity increased, *L*-value decreased while *a* and *b*-value increased in all measurements, indicating higher rate of browning at higher water activities (Fig. 1). Duncan's test showed *L*-values were significantly lower at aw of 0.5 and higher water activities for all measurements (Table 1). *a*-values were significantly higher at 0.5 and higher water activities for whole-kernel measurements, and at 0.42 and higher water activities for ground-state and cut-kernel measurements (Table 1). *b*-Values were significantly higher at 0.48 and higher water activities for whole-kernel and cut-kernel measurements, and at 0.42 and higher water activities for ground-state measurements (Table 1). These results showed that rates of colour change increased above 0.48 water activity in whole-kernel measurements, and at water activities more than 0.42 in ground-state and cut kernel measurements. Higher non-enzymatic browning rates at these water activity levels were also reported in other foods (Labuza, 1980; Leung, 1990; Troller, 1989).

Table 1
Effects of water activity on colour attributes of roasted hazelnut samples for whole-kernel, ground-state and cut kernel measurements

Measurements	a_w	*L*-Value	*a*-Value	*b*-Value
		Mean ± S.D.[ab]	Mean ± S.D.	Mean ± S.D.
Whole-kernel	0.29	77.78±2.35d	0.88±0.50a	19.17±1.59a
	0.42	77.83±2.18d	0.83±0.78a	20.25±1.77ab
	0.48	77.80±2.69d	1.26±0.47a	20.44±0.76ab
	0.50	78.09±1.33d	1.19±0.20a	21.16±1.28bc
	0.71	75.47±2.74cd	3.00±0.96b	23.38±1.47d
	0.74	71.56±4.21ab	5.20±2.15c	25.36±1.44e
	0.80	70.41±5.49a	4.94±2.61c	24.82±1.21e
	0.83	75.00±4.52cd	5.73±2.08c	27.76±1.25f
P[c]		< 0.0001	< 0.0001	< 0.0001
Ground-state	0.29	70.38±0.86c	1.54±0.41a	18.45±0.50a
	0.42	68.78±0.77bc	1.69±0.16a	19.04±0.32ab
	0.48	70.01±2.88c	2.45±0.36b	19.54±0.81bc
	0.50	69.26±1.74bc	2.31±0.35b	20.04±0.30c
	0.71	66.36±1.34a	4.83±0.38c	22.73±0.93e
	0.74	61.92±1.91d	5.33±0.26c	21.94±0.92de
	0.80	65.27±2.11a	7.08±0.64e	24.72±0.97f
	0.83	64.76±2.1a	6.17±0.72d	24.63±0.39f
P		< 0.0001	< 0.0001	< 0.0001
Cut-kernel	0.29	70.09 ±3.23d	4.07±1.23a	19.38±1.28a
	0.42	66.62±5.95bcd	5.43±1.34ab	18.74±1.02a
	0.48	67.72±4.74cd	5.57±1.53bc	19.63±0.53ab
	0.50	67.57±2.45cd	5.74±1.08bcd	20.44±0.91bc
	0.71	62.72±7.66abc	6.98 ±2.53cde	20.80±1.11c
	0.74	58.72±3.12a	7.87±0.91ef	20.51±0.69bc
	0.80	58.29±4.51a	7.65±1.138ef	20.91±0.83c
	0.83	61.78±5.85ab	8.66±1.48f	22.66±1.52d
P		< 0.0001	< 0.0001	< 0.0001

[a] Values in the same column with the different lower-case letters (a–f) are significantly different at $P < 0.05$.
[b] S.D., standard deviation.
[c] *P*, probability of significance.

There was a significant difference between measurements for *L*- and *a*-value at all water activities as shown in Table 1. But there was no significant difference in *b*-value between measurements at 0.29, 0.42 and 0.50 water activities (Table 1). The difference between the measurements can be attributed to the internal browning of hazelnuts during roasting, because whole-kernel measurements do not take account of the internal browning in the centres of kernels (Özdemir and Devres, 2000a,b).

Duncan's test indicated that whole-kernel measurements gave significantly higher *L*-values than ground-state and cut-kernel measurements for all water activities (Table 2). The difference between whole-kernel and cut-kernel measurements was less than eight units at 0.29

Table 2
Effects of whole-kernel, ground-state and cut kernel measurements on colour attributes of roasted hazelnut samples with respect to initial water activity level

a_w	Measurements	*L*-Value Mean ± S.D.[ab]	*a*-Value Mean ± S.D.	*b*-Value Mean ± S.D.
0.29	Whole-kernel	77.78±2.52b	0.88±0.50a	19.17±1.59a
	Ground-state	70.38±0.86a	1.54±0.41a	18.45±0.50a
	Cut-kernel	70.09±3.22a	4.07±1.23b	19.38±1.28a
P[c]		< 0.0001	< 0.0001	NS[d]
0.42	Whole-kernel	77.83±2.18b	0.83±0.78a	20.25±1.77b
	Ground-state	68.78±0.77a	1.69±0.16a	19.04±0.32ab
	Cut-kernel	66.62±5.95a	5.43±1.34b	18.74±1.02a
P		< 0.0001	< 0.0001	NS
0.48	Whole-kernel	77.80±2.69b	1.26±0.47a	20.44±0.76b
	Ground-state	70.01±2.88a	2.45±0.36b	19.54±0.81a
	Cut-kernel	67.72±4.74a	5.57±1.53c	19.63 ±0.53a
P		< 0.0001	< 0.0001	0.023
0.50	Whole-kernel	78.09±1.33b	1.19±0.20a	21.16±1.28a
	Ground-state	69.26±1.74a	2.31 ±0.35b	20.04±0.30a
	Cut-kernel	67.57±2.45a	5.74±1.08c	20.44±0.91a
P		0.000	0.000	NS
0.71	Whole-kernel	75.47±2.74b	3.00±0.96a	23.38 ±1.47b
	Ground-state	66.36±1.34a	4.83±0.38a	22.73±0.93b
	Cut-kernel	62.72±7.66a	6.98±2.53b	20.80±1.11a
P		< 0.0001	< 0.0001	< 0.0001
0.74	Whole-kernel	71.56±4.21b	5.20±2.15a	25.36±1.44c
	Ground-state	61.92 ±1.91a	5.33±0.26a	21.94±0.92b
	Cut-kernel	58.72±3.12a	7.87±0.91b	20.51±0.69a
P		< 0.0001	0.001	< 0.0001
0.80	Whole-kernel	70.41±5.49c	4.94±2.61a	24.82±1.21b
	Ground-state	65.27±2.11b	7.08±0.64b	24.72±0.97b
	Cut-kernel	58.29±4.51a	7.65±1.14b	20.91±0.83a
P		< 0.0001	0.009	< 0.0001
0.83	Whole-kernel	75.00±4.52b	5.73±2.08a	27.76±1.25c
	Ground-state	64.76±2.10a	6.17±0.72a	24.63±0.39b
	Cut-kernel	61.78±5.85a	8.66±1.48b	22.66±1.52a
P		< 0.0001	0.002	< 0.0001

[a] Values in the same column with the different lower-case letters (a–c) are significantly different at $P < 0.05$.
[b] S.D., standard deviation.
[c] *P*, probability of significance.
[d] NS, not significant.

water activity only. It ranged from 10.1 to 13.6 at higher water activities (Table 2). The difference between *b*-value of whole-kernel and cut-kernel measurements was more than 2 units at 0.71 and higher water activities.

Duncan's test also indicated that there was no significant difference between *a*-value of whole-kernel and ground-state measurements at water activities of 0.29, 0.42, 0.71, 0.74 and 0.83 (Table 2). The difference between *a*-values of whole-kernel and cut-kernel measurements was less than 3 units at 0.29, 0.71 and higher water activities. However, the range of *a*-values at 0.29 water activity was from 0.83 to 4.07, while from 3.0 to 8.66 for 0.71 and higher water activities. Since the higher *a*-value indicates higher redness, and, subsequently, a higher internal browning in the roasted hazelnuts, these results indicated a lower level of internal browning at 0.29 water activity. This may be attributed to decreased mobility of substrates and, subsequently, a lower rate of non-enzymatic browning at lower water activities (Labuza, 1980; Troller, 1990). At industrially used storage water activity levels, around 0.74, (approx. 6% moisture content), hazelnuts were most susceptible to internal browning. Therefore, a pre-drying prior to roasting would be useful in controlling internal browning of roasted hazelnuts. A pre-drying stage during roasting was also suggested by Perren and Escher (1996b) to increase shelf life of roasted hazelnuts.

3.2. Polyphenoloxidase activity in raw and roasted hazelnuts

Polyphenoloxidase activity was observed in samples (0.74 initial water activity) in raw state but no activity of the enzyme was observed in samples after roasting at 130°C for 16 and 30 min (Fig. 2). These results suggest that enzymatic activity in internal browning of roasted hazelnuts is very low. Enzymes responsible for enzymatic-browning were reported to be denatured at high process temperatures (Driscoll & Madamba, 1994; Troller, 1989). Moreover, Lopez, Pique, Ferran, Romero, Boatella and Garcia (1997) reported that heat stability of polyphenoloxidase is low, and browning of hazelnuts during drying of hazelnuts at a temperature range of 30–70°C is due to nonenzymatic browning.

3.3. Composition changes in internally browned samples

Total sugar, sucrose and reducing sugar contents of raw hazelnuts, equilibrated over different relative humidities, and roasted hazelnuts, having light-coloured internal centres and having dark-coloured internal centres are given in Table 3. Reducing sugars were found only in raw samples of 0.83 and 0.80 water activity. Reducing sugars may be formed through enzymatic hydrolysis of non-reducing sugars during storage (Keme et al., 1983a,b; King et al., 1983; Prichavudhi & Yamamoto, 1965). A visible mould growth at 0.80 and 0.83 a_w was also observed within 15 days during storage at 20°C which may contribute to breakdown of carbohydrates into their monomers at high moisture content (King et al., 1983).

There were significant differences between total sugar and sucrose contents of raw hazelnuts, roasted hazelnuts, having light-coloured centres, and roasted hazelnuts, having dark-coloured centres at each water activity level (Table 3). Dark-coloured samples showed equal to or significantly higher total sugar and sucrose contents at

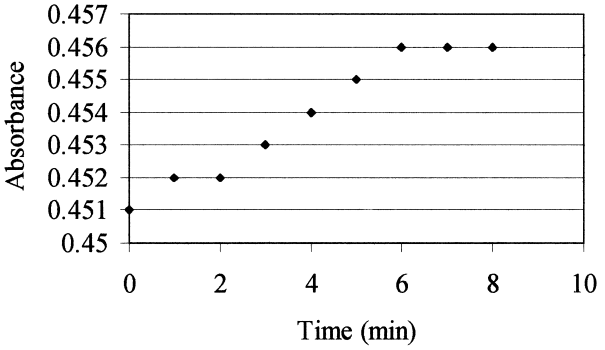

Fig. 2. Activity of polyphenoloxidase in raw hazelnut with 0.74 water activity.

all water activities (Table 3). Moreover, brown centres in the roasted sample, with initial water activity of 0.74, showed significantly more total sugar and sucrose than light-coloured outer layers (Table 3). These results suggested that brown centres were formed through non-enzymatic browning reactions between sugar components, and other kernel constituent. Similarly, Prichavudhi and Yamamoto (1965) reported that brown centres in the macadamia nut had higher levels of reducing sugar than the light outer layers. They postulated that high drying temperature increased reducing sugar content through enzymatic hydrolysis of non-reducing sugars. Subsequently, upon heat treatment, such as roasting, brown centres are formed through non-enzymatic browning reactions between reducing sugars, amino acids and other kernel constituents (Prichavudhi & Yamamoto, 1965). These results also supported by Göğüş et al. (1998) who discussed effect of temperature and moisture gradient within food during processing, and localisation of constituent within the food. They reported that all these factors may cause different non-enzymatic reaction rates in different sections of the samples, and subsequently localised browning.

Özdemir and Devres (2000a) reported that concentrations of reactants in the inner part of the raw hazelnuts might also contribute to internal browning. So as to investigate localisation of reactants, protein, total sugar, sucrose, and total oil contents were determined in the outer and inner layers of raw hazelnuts. The results showed that there was a significant difference between inner and outer layer ($P < 0.0001$) in terms of the reactants. Total oil content was significantly lower in outer layer of kernels (56.27%, d.b.) than inner layers (66.07%, d.b.). Protein contents of outer layers (20.1%, d.b.) were significantly higher than inner layer (14.4%, d.b.). Total sugar (1.45%, d.b.) and sucrose contents (1.38%, d.b.) of outer layers were significantly lower than those of inner layers. Inner layers contained about 50% more total sugar (2.84%, d.b.) and sucrose (2.69%, d.b.). These results indicated that the limiting reactant was sugar which concentrated in the inner layer of raw hazelnuts. Therefore, non-enzymatic browning reactions may develop faster in the inner parts of the kernels. This observation coincides with the pattern of internal browning which develops in the inner parts of the hazelnut, while colour of outer parts remains similar to that of the surface of roasted hazelnuts.

Table 3
Effects of initial water activity and roasting on the total sugar, sucrose and reducing sugar contents of raw hazelnuts and roasted hazelnuts, with light-coloured internal centres and with dark-coloured internal centres

a_w	Sample	Total sugar (%)	Sucrose (%)	Reducing sugar (%)
		Mean±S.D.[ab]	Mean±S.D.	Mean±S.D.
0.83	Raw	6.07±0.01a	2.99±0.10a	2.92 ±0.02
	Light[c]	2.51±0.01c	2.39±0.01b	
	Dark[d]	4.22±0.06b	4.01±0.01a	
	P[e]	< 0.0001	0.004	
0.8	Raw	6.75±0.02c	5.25±0.18c	1.22±0.17
	Light	8.03±0.03b	7.63±0.03b	
	Dark	11.63±0.31a	11.05±0.30a	
	P	< 0.0001	< 0.0001	
0.74	Raw	3.58±0.02b	3.40±0.02b	
	Light	2.49±0.03d	2.37±0.032d	
	Dark	3.59±0.01b	3.41±0.01b	
	Brown-centre	5.90±0.04a	5.60±0.04a	
	Light-coloured outer	3.46±0.02c	3.29±0.02c	
	P	< 0.0001	< 0.0001	
0.71	Raw	4.95±0.03a	4.70±0.03a	
	Light	4.15±0.01b	3.94±0.01b	
	Dark	4.19±0.07b	3.98±0.01b	
	P	< 0.0001	< 0.0001	
0.58	Raw	3.87±0.04b	3.67±0.03b	
	Light	3.57±0.01c	3.39±0.01c	
	Dark	4.22±0.02a	4.01±0.02a	
	P	< 0.0001	< 0.0001	
0.48	Raw	3.62±0.02c	3.44±0.02c	
	Light	3.82±0.02b	3.63±0.02b	
	Dark	4.25±0.03a	4.03±0.02a	
	P	< 0.0001	< 0.0001	
0.42	Raw	4.13±0.01a	3.92±0.01a	
	Light	3.72±0.01c	3.53±0.01c	
	Dark	3.76±0.03b	3.57±0.03b	
	P	< 0.0001	< 0.0001	
0.29	Raw	4.15±0.01a	3.95±0.01a	
	Light	3.85±0.02b	3.66±0.02b	
	Dark	3.90±0.08b	3.71±0.08b	
	P	< 0.0001	< 0.0001	

[a] Values in the same column with the different lower-case letters (a–c) are significantly different at $P < 0.05$.
[b] S.D., Standard deviation.
[c] Light; roasted hazelnuts, having light-coloured internal centres.
[d] Dark; roasted hazelnuts, having dark-coloured internal centres.
[e] *P*, probability of significance.

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