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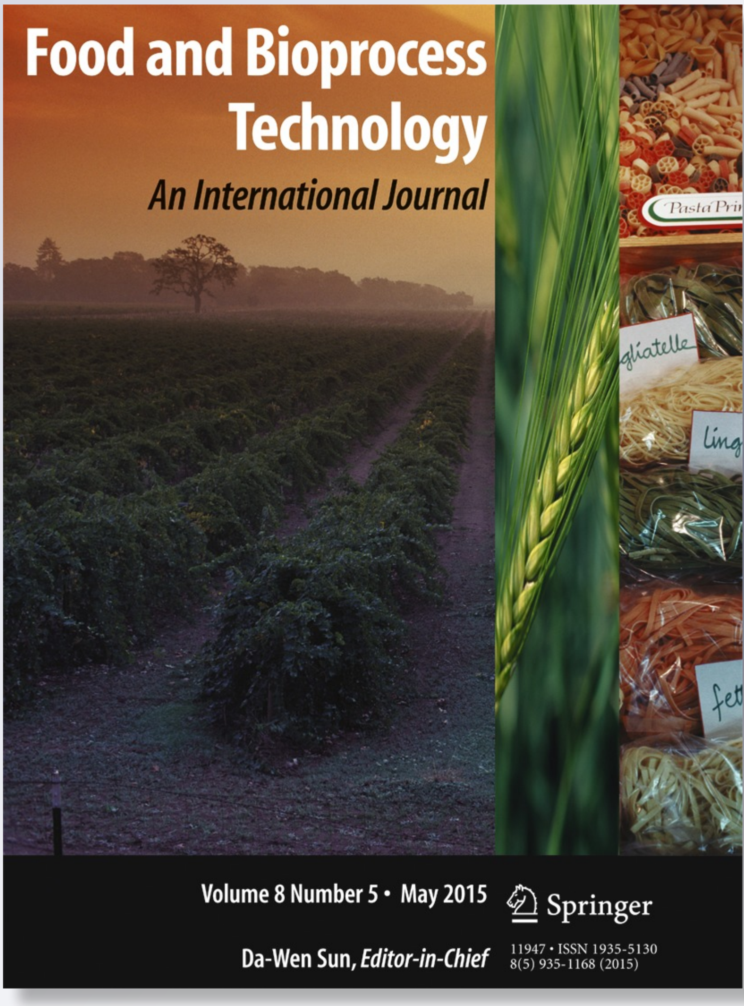


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ORIGINAL PAPER

# Modelling the Influence of Origin, Packing and Storage on Water Activity, Colour and Texture of Almonds, Hazelnuts and Walnuts Using Artificial Neural Networks

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**Abstract** The present work assessed the influence of different factors on some physical and chemical properties of nuts. The factors evaluated were the presence or absence of the inner skin, geographical origin, storage conditions (ambient temperature, in a stove at 30 and 50 °C, in a chamber at 30 and 50 °C and 90 % RH, refrigerated and freezing) and type of package (none, low density polyethylene and low density polyethylene). The fruits studied were almonds, hazelnuts and walnuts from different countries. The properties measured were moisture content, water activity, colour coordinates (L*, a* and b*) and texture parameters (hardness and friability). Experimental data were modelled using neural networks. The results showed that the almonds from Spain and Romania had $a_w$ greater than 0.6, and therefore, its stability was not guaranteed, contrarily to the other samples that presented values of $a_w$ lower than 0.6. The colour coordinate lightness varied from 40.60 to 49.30 in the fresh samples but decreased during storage, indicating darkening. In general, an increase in hardness and friability was observed with the different storage conditions. Neuron weight analysis has shown that the origin was a good predictor for moisture content and texture; whereas, the storage condition was a good predictor for $a_w$ and colour. In conclusion, it was possible to verify that the properties of nuts are very different depending on origin; they are better preserved at lower temperatures and the type of package used did not impact the properties studied.

**Keywords** Colour · Neural network analysis · Nut · Storage · Texture · Water activity

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## Introduction

Tree nuts are used worldwide in confectionary, culinary and bakery food applications due to their desirable flavour attributes and high energy density (Guiné et al. 2014a; Hokmabadi and Sedaghati 2014). Furthermore, they are a compact source of numerous healthful nutrients including monounsaturated fat, α-linolenic acid, protein, arginine, fiber, vitamin E, magnesium and copper, as well as a wealth of healthful phytochemicals such as phytosterols, flavonoids and proanthocyanidins (Bolling et al. 2011; Sweazea et al. 2014).

Almonds (*Prunus dulcis* M.) belong to the family *Rosaceae* and are a very important crop worldwide, with a global production of about 1.7 million metric tons (Mandalari et al. 2014). The consumption of almonds has been associated with a multitude of nutritional benefits, including cholesterol-lowering effects, protection against diabetes and potential prebiotic properties (López-Calleja et al. 2014).

Hazelnut (*Corylus avellana* L.) is a popular nut tree worldwide and the fruits are one of the most important raw materials for the pastry and chocolate industry due to their organoleptic characteristics. Furthermore, hazelnuts contribute to human nutrition and health because of their protein, oil, vitamin and mineral contents (Alasalvar et al. 2009; Turan et al. 2015; Yağci and Göğüş 2009).

Walnuts (*Juglans regia* L.) are an excellent source of essential unsaturated fatty acids and tocopherols. Among tree nuts, walnut is one of the two highest ranked nuts for total phenolic content and antioxidant capacity, being very rich in the strong antioxidant hormone, melatonin. Besides, it is also rich in a variety of polyphenolic components, including ellagitannins, quinones, phenyl propanoids and dicarboxylic acid derivatives (Colaric et al. 2005; Grace et al. 2014; Ito et al. 2007; Kornsteiner et al. 2006; Li et al. 2006).

The storage conditions are of major importance so as to maintain the integrity and quality of nuts, and thus, prevent spoilage (Freitas-Silva and Venâncio 2011; Ma et al. 2013). Post-harvest handling and storage might deteriorate both nut nutritional value and taste. It is well known that many factors, such as a high drying temperature or prolonged storage under air and/or at a relatively high temperature and humidity have negative effects on various nut properties (Tsantili et al. 2011). According to Ghirardello et al. (2013), the final quality of nuts is defined by a number of factors, such as lipid oxidation, appearance, texture, flavour, chemical composition, nutritional value and food safety aspects.

The artificial neural network (ANN) approach is a generic technique for mapping non-linear relationships between inputs and outputs without knowing the details of these relationships (Ceylan and Aktaş 2008). ANN modelling resembles a data processing (learning) and decision making system of a human brain and the network function is greatly determined by the connections between elements which mimic human's brain cells and nerves (Çelekli et al. 2012; Khataee and Kasiri 2010). An ANN is generally a parallel interconnected structure consisting of the input layer of neurons (simple processing units), a number of hidden layers and the output layer (Çelekli et al. 2012; Khataee et al. 2011). The first ANN models were proposed in 1943 (McCulloch and Pitts 1943), as a general bio-inspired brain modelling tool. Since then, network models have evolved and been used for different purposes such as pattern recognition, data classification and other data mining tasks. Recently, ANNs have been widely used in modelling food engineering processes of different nature. Huang (2012) applied ANN and image processing techniques for detecting and classifying the quality of areca nuts. Teimouri et al. (2014) used a novel ANN assisted segmentation algorithm for discriminating almond nut and shell from background and shadow, classifying the images into three categories. Omid et al. (2009) developed and tested an intelligent pistachio nut sorting system combining acoustic emissions analysis, principal component analysis and a multilayer feed-forward neural network. Ceylan and Aktaş (2008) presented an application of an ANN for modelling of a hazelnut dryer assisted heat pump. Çelekli et al. (2012) used an ANN model to predict the removal efficiency of Lanaset Red G on walnut husk. Guiné et al. used ANN to model some characteristics of dried apples (Guiné et al. 2014b) and bananas (Guiné et al. 2015).

The aim of the present work was to study the effect of different storage conditions and types of package on some physicochemical properties of commercially available nuts (almond, hazelnut and walnut) from different geographical origins. The properties evaluated at the chemical level were the moisture content and water activity, because these relate directly to stability and microbial safety. The properties evaluated at the physical level were the colour and texture (hardness and friability) because they are associated with organoleptic acceptance and quality perception.

## Materials and Methods

### Sampling

The fruits evaluated in this study were almond, hazelnut and nut, all without the outer shell; but in some cases, they retained the inner skin (seed coat), and in other cases, they did not.

The almond samples were: from Spain with skin (A-SP-s), Portugal with skin (A-PT-s) and from United States with and without skin (A-US-s and A-US-n). The hazelnut samples were: from Spain with skin (H-SP-s), Portugal with skin (H-PT-s) and from Turkey without skin (H-TR-n). The walnut samples were: from Chile with skin (W-CH-s), Portugal with skin (W-PT-s), Romania with skin (W-RO-s) and USA with skin (W-US-s).

### Analysis of Water Activity and Moisture Content

The water activity was measured at 25 °C by a hygrometer (Hygroskop model BT-RS1 from Rotronic, New York, USA) connected to a thermal bath. In all the cases, four measurements were made to calculate the mean value and standard deviation. Moisture content was evaluated by drying in a stove (model WTB from manufacturer Binder, Tuttlingen, Germany) at 105 °C until reaching constant weight. In all the cases, the determinations were made in triplicates.

### Evaluation of Colour

The colour of all samples was measured using a handheld tristimulus colorimeter (chroma meter model CR-400 from manufacturer Konica Minolta, Tokyo, Japan). The parameters measured were the brightness L*, which varies between 0 and 100 (from black to white, respectively), and the coordinates of opposed colour: a* and b*, which vary from – 60 to +60. The redness, a*, assumes negative values for green and positive values for red, while the yellowness, b*, assumes negative values for blue and positive for yellow. The total colour difference (TCD) was the parameter considered for the overall colour difference evaluation, between a sample and the reference (Guiné and Barroca 2012):

$$\mathrm{TCD} = \sqrt{(L_0{*}-L{*})^2 + (a_0{*}-a{*})^2 + (b_0{*}-b{*})^2} \quad (1)$$

where $L_0*$, $a_0*$ and $b_0*$ are the reference values. In all the cases, the number of replicates was 25.

The TCD was used for evaluating the change of colour because it is generally used by many researchers to assess the difference between two colours, often related to a reference sample (Chutintrasri and Noomhorm 2007; Goyeneche et al. 2014; Kačíková et al. 2013; Kara and Erçelebi 2013; Kaushik et al. 2014; Mahdavee Khazaei et al. 2014). The total colour difference is evaluated according to the following scale: trace level difference $\Delta E*=0$–0.5, slight difference $\Delta E*=0.5$–1.5, noticeable difference $\Delta E*=1.5$–3.0, appreciable difference $\Delta E*=3.0$–6.0, large difference $\Delta E*=6.0$–12.0 and very obvious difference $\Delta E*>12.0$ (Chen and Mujundar 2008).

### Texture Analysis

The texture analysis for all samples was made by a texturometer (model TA-XT Plus from manufacturer Stable Micro Systems, Godalming, Surrey, UK). The test performed consisted in measuring the force in compression. The probe used was a Blade Set HDP/BS (Warner- Bratzler). The load cell used was 500 N and the trigger force was 0.15 N. The, pre-test, test and post-test speeds were respectively 1.50, 1.00 and 10.00 mm/s. Twenty measurements were carried for each sample, allowing the assessment of parameters such as the friability and hardness as shown in Fig. 1. For the walnuts, the texture was not accessed due to the high irregularity of the fruits, thus, not allowing a correct evaluation of the textural attributes. The curve force (N) versus distance (mm) allows calculating the hardness (the force at first peak) and friability (the distance of first peak). Hardness is the mechanical strength necessary to crush (Guiné and Marques 2013; Santos et al. 2013). It is important as it ensures the physical integrity of the product, allowing it to support the mechanical stress in the process of packing and transportation. The friability respects to the ease with which the fracture occurs in the products (Almeida 2013; Gharibzahedi et al. 2012).

### Storage Conditions

Different samples of the fruits were stored for 90 days (a) at ambient conditions, (b) in a stove at two temperatures with controlled temperature but no control over relative humidity (RH), (c) in a chamber with controlled temperatures and relative humidity, (d) under refrigeration and (e) frozen.

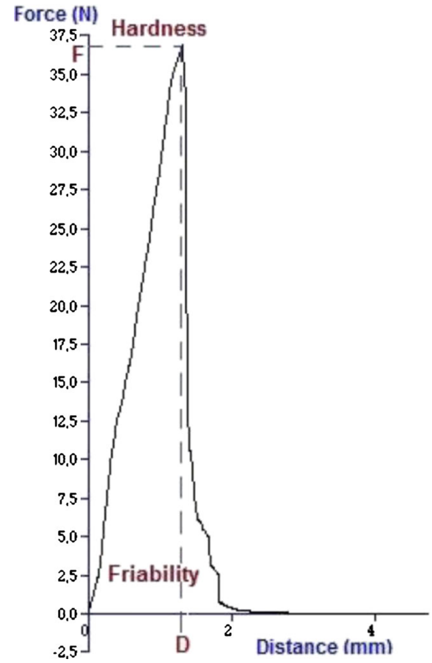



**Fig. 1** Example of graph obtained with the texture measurement

Furthermore, different samples were stored (a) without any package, and with two commercial plastics: (b) low density polyethylene (LDPE) from ALBERPLÁS with 110 μm thickness (density=920–940 kg/m$^3$) and (c) linear low density polyethylene (LLDPE) from MGP minigrip with 40 μm thickness. Table 1 summarizes the conditions used to store all fruits.

### Artificial Neural Network Modelling

Experimental data were modelled using artificial neural networks trained and simulated in Matlab™ .[1] The ANN used was a feed-forward model, created using Matlab fitnet function. The ANN used Levenberg–Marquartd method for training and the mean squared error method for performance assessment, as in fitnet function default values. The type of nut was encoded as input variables 1–3. The presence of skin was input 4. The country of origin was encoded in inputs 5–10. Storage type was encoded in inputs 11–17 and the type of package accounted for inputs 18–19. Therefore, the number of inputs for the network was 19, when all inputs were available.

The total number of entries in the dataset available was 6,050, each entry corresponding to one experiment. The values of the output variables were not known for all entries, since each line represented a different experiment. Therefore, the number of samples available for training and validating the neural networks was lower and depended on which variable was being studied.

Table summarises the number of samples available for each output variable. As the table shows, the number of valid samples varied but was always large enough for data analysis

[1] Matlab is a registered trademark of Mathworks. www.mathworks.com.

**Table 1** Storage conditions tested

| Description | T[1] (°C) | RH[1] (%) |
|---|---|---|
| B: Before storage | – | – |
| AT: Ambient temperature | 23.4±2.5 | 50.5±6.7 |
| S30: Stove (T controlled but RH not) | 30.0±0.0 | 36.0±3.6 |
| S50: Stove (T controlled but RH not) | 50.0±0.0 | 13.2±1.5 |
| C30: Chamber (T and RH controlled) | 30.0±0.0 | 90.0±0.0 |
| C50: Chamber (T and RH controlled) | 50.0±0.0 | 90.0±0.0 |
| R: Refrigerated | 2.3±1.7 | 48.1±13.3 |
| F: Frozen | -15.4±2.6 | 61.7±6.2 |

All results are mean value±standard deviation

[1] T = temperature, RH = relative humidity

under normal conditions. To facilitate training and analysis of the results, each output variable was processed separately. This simplification does not imply any loss of generality, for it is always possible to simulate a smaller neural network using a larger neural network with sufficient neurons, and it greatly facilitates the analysis of the results. Thus, subsets of the whole dataset were created for each output variable, and the rows which contained no output value for the variable being studied were dropped, leading to the cardinality numbers shown in Table 2.

Each network had just one hidden layer with one neuron and one output neuron. Experimental results showed that fitness of the model was only marginally improved if more neurons were used in the hidden layer, so all the results shown were obtained with networks containing just one neuron in the hidden layer, in order to facilitate the sensitivity analysis. For each run, the Matlab script randomly selected approximately 70 % of the samples for the train subset, 15 % for the validate subset and the remainder samples for the test subset.

## Results and Discussion

### Moisture Content and Water Activity

Figure 2 shows the mean values and the corresponding standard deviation for the moisture content and water activity of the different nuts studied. The results obtained for moisture content revealed that in general the almonds had higher moisture when compared with the other two types of nuts (hazelnuts and walnuts), varying from 4.3 to 6.8 %. Xiao et al. (2014) reported moisture levels in raw almonds with skin of about 5 %. Hosseinpour et al. (2013) reported values of moisture varying from 4.10 to 8.75 % for hazelnut kernels of 12 cultivars. In the present work, the moisture in the hazelnuts varied from 2.3 to 4.2 %. For the walnut samples, the moisture observed varied from 3.0 to 4.5 %. Toğrul and Arslan (2007) reported values of moisture in unshelled walnuts cultivated in Turkey of about 3.6 % and Yang et al. (2013) for raw almonds from California (USA) reported a value of 4.6 %.

**Table 2** Number of samples of the datasets used to study each output variable

| Characteristic | Number of samples in the dataset |
|---|---|
| MC (moisture content) | 609 |
| $a_w$ (water activity) | 820 |
| TCD (total color difference) | 4,708 |
| H (hardness) | 2,761 |
| F (friability) | 2,761 |

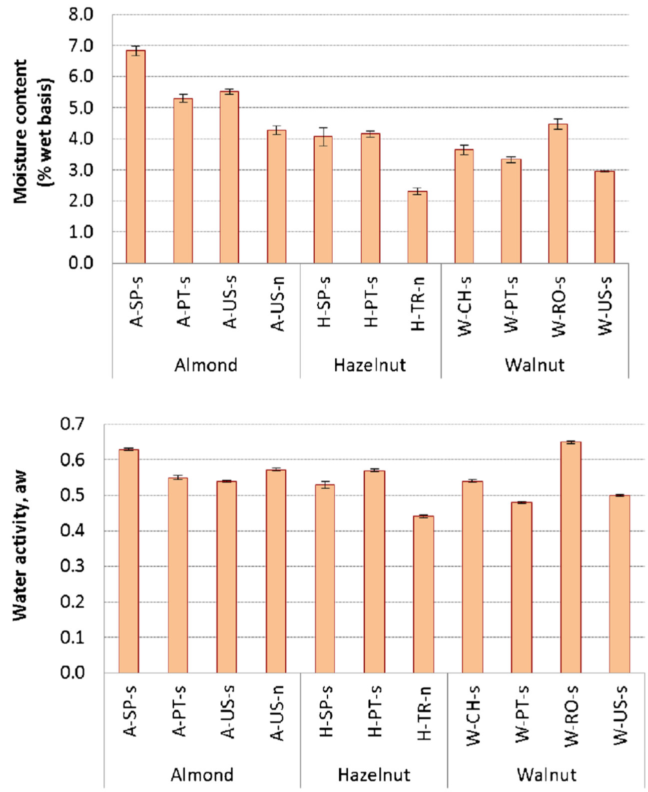



**Fig. 2** Moisture content and water activity of the nuts studied (*A* almonds, *H* hazelnuts, *W* walnuts; country codes: *SP* Spain, *PT* Portugal, *US* United States, *TR* Turkey, *CH* Chile, *RO* Romania; *s* with internal skin, *n* without internal skin)

Furthermore, it was observed that the different geographical origin of the nuts influenced their moisture content. The highest values were registered among the almonds in the sample from Spain (6.8 %), among the hazelnuts in the sample from Portugal (4.2 %) and among the walnuts in the sample from Romania (4.5 %). Finally, it was observed that the absence of the inner skin (the seed coat) contributed for some dehydration, as could be seen comparing the samples of almonds from USA with and without skin, with 5.5 and 4.3 % moisture content, respectively.

Besides the moisture content, also the water activity ($a_w$) is of vital importance to assure the quality of the food products because this relates to the free water that is available to chemical and enzymatic reactions as well as microbial growth. It is reported that for $a_w$ lower than 0.6 practically all microbial activity is neutralized (bacteria, fungi or yeasts). Contrarily, for $a_w$ over 0.6, the chemical and enzymatic reactions start assuming some importance, as for example, the Maillard reactions that produce browning reactions and colour changes and which are favoured at $a_w$ between 0.6 and 0.7 (Guiné 2011). The results in Fig. 2 showed that the almonds from Spain and the walnuts from Romania had $a_w$ greater than 0.6 (0.63 and 0.65, respectively), and therefore, its stability could be compromised due to degradation reactions. However, all other samples presented values of $a_w$ lower than 0.6 and therefore not susceptible to deterioration reactions. Piscopo et al. (2010) reported values of $a_w$ for almond kernels from Italian, French and Spanish cultivars varying from 0.48 to 0.61.

Figure 3 shows the effect of storage conditions and type of package on the moisture content and water activity of the different nuts studied. For this analysis, and to facilitate understanding, the average variation values were calculated for each type of nut, according to the following equation:

$$\mathrm{V}R\ (\%) = \frac{Vi - V_B}{V_B} \times 100 \qquad (2)$$

where VR represents the percentage of variation relatively to the corresponding control sample, which corresponds to storage condition B. It is worth noting that later the influence of each factor affecting nut properties will be discussed separately as the result of the ANN analysis carried out.

The results in Fig. 3 revealed that the storage in the stove at 50 °C originated a high degree of dehydration, decreasing moisture content within the range −62 to −71 %, this trend being similar regardless of the type of nut or the type of package used for storage. It was further observed that the only situations which originated an increase in the moisture level were the storage in the chamber at 30 or 50 °C and 90 % RH, and that this increase was larger in the almonds and hazelnuts packed in LLDPE bags. To note that in some cases no evaluation was undertaken, due to experimental difficulties. Table 3 shows the water vapour permeability of different low density polyethylene films for diverse conditions. The values indicate a clear influence of thickness and storage time on the barrier properties of the low density polyethylene films.

The storage methods that originated in general lower changes in moisture levels were refrigeration and freezing, followed by ambient temperature (Fig. 3). García-Pascual et al. (2003) studied the influence of storage conditions on the moisture of shelled almonds of four varieties (three Spanish and one from California, USA). They investigated the storage at two temperatures (8 and 36 °C) and two packaging atmospheres (air and $N_2$) throughout a period of 9 months. Their results revealed that no significant differences were observed between air and nitrogen packaging in terms of moisture content at the end of the study and that almonds stored in their shells at ambient temperature maintained their

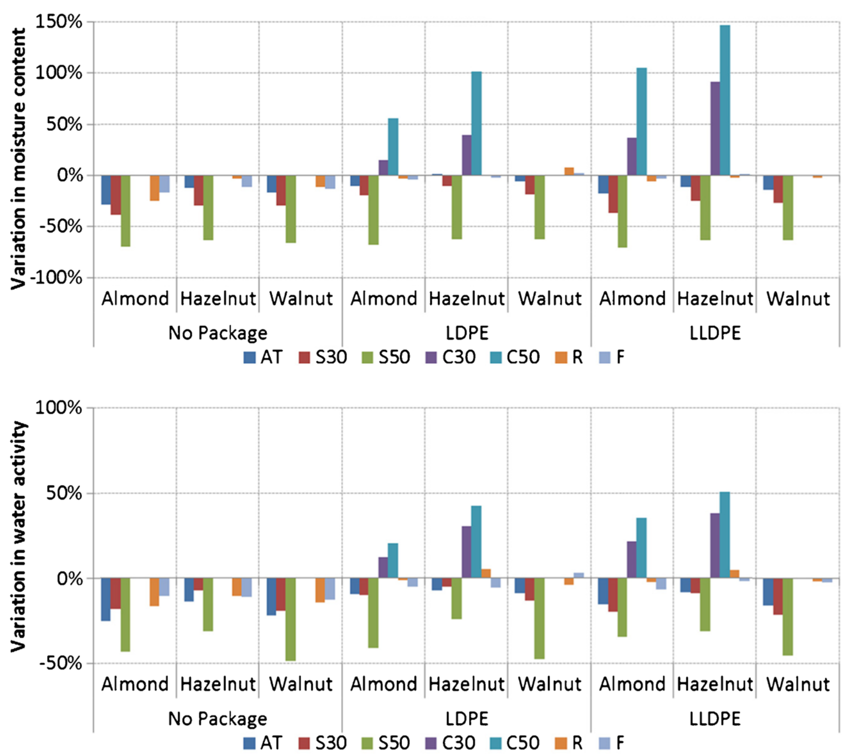



**Fig. 3** Average variation of moisture content and water activity during storage (storage: *AT* ambient temperature, *S30* storage at high temperature 30 °C, *S50* storage at high temperature 50 °C, *C30* chamber at high temperature 30 °C and 90 % RH, *C50* chamber at high temperature 50 °C and 90 % RH, *R* refrigerated, *F* frozen)

**Table 3** Permeability of low density polyethylene films

| Film thickness (μm) | Storage time (days) | Permeability ($mg.cm^{-2}.kPa^{-1}$) | |
|---|---|---|---|
| | | Storage at ~30 °C, ~90 % RH | Storage at ~50 °C, ~90 % RH |
| 25 | 30 | 0.522 | 1.105 |
| 25 | 60 | 1.044 | 2.213 |
| 25 | 90 | 1.566 | 3.317 |
| 40 | 30 | 0.326 | 0.691 |
| 40 | 60 | 0.653 | 1.383 |
| 40 | 90 | 0.979 | 2.075 |
| 70 | 30 | 0.186 | 0.395 |
| 70 | 60 | 0.373 | 0.789 |
| 70 | 90 | 0.559 | 1.185 |
| 110 | 30 | 0.119 | 0.251 |
| 110 | 60 | 0.237 | 0.503 |
| 110 | 90 | 0.356 | 0.753 |

quality. Ghirardello et al. (2013) studied the effect of one year storage under different conditions on the moisture content of hazelnut kernels. The storage conditions tested were ambient temperature and refrigeration at 4 °C and 55 % relative humidity, with or without modified atmosphere (1 % oxygen and 99 % nitrogen). Their results showed that the in-shell hazelnuts stored at ambient temperature increased moisture content in 26 %, whereas the shelled hazelnuts stored under $N_2$ and refrigerated practically did not alter their moisture content (variations of +1 % and −1 %, respectively).

Figure 3 also showed that the trends observed for moisture content were similar to those obtained for water activity, and once again, refrigeration and freezing produced the lowest changes as compared to the control nuts.

The moisture variations in the nuts along time were used to estimate the diffusion coefficients for water loss or uptake according to the Fick's second law equation for diffusion (Guiné et al. 2013). Figure 4 shows those values of diffusivity for the three types of nuts originating from Portugal. The results indicated that in general LLDPE offered a very poor protection for the nuts, and the LDPE, although slightly better, also did not prevent the moisture alterations, particularly, for the storage at the highest temperature (50 °C) both in the stove and in the chamber.

## Colour

The results in Fig. 5 relate to the colour coordinates, lightness (L*), redness (a*) and yellowness (b*) of the nuts studied, according to type of nut and geographical origin. It could be observed from the results obtained that lightness, or sometimes also called brightness, as well as redness varied considerably between the samples with skin and those without skin. The samples without skin (A-USA-n and H-TR-n) were much lighter, with larger values of L* (78.10 and 75.20, respectively) and less reddish (with values of a* equal to 1.60 and 4.80, respectively). Regarding the colour coordinate b*, there were no pronounced differences between the nuts with and without the skin.

The colour coordinate lightness (L* in Fig. 5) varied between 40.60 and 49.30 for the nuts with the inner skin, and no evident differences were observed among the different origins or even the different types of nut. The results reported by Özdemir and Devres (2000) show values of L* between 80 and 85 for hazelnuts from Turkey without the seed coat, thus higher than the value found in this work for the sample H-TR-n, also without the seed coat (75.20). Mexis and Kontominas (2009) reported a value for the lightness of hazelnut kernels of 30.45, which is lower than those reported in this study for the hazelnuts with skin (42.60 and 47.20).

As to redness (a* in Fig. 5), it presented very similar values among the three different almonds studied with the skin (around 17), and this trend was also observed for the two hazelnuts with skin (around 18). However, for the walnuts the values were lower (varying in the range 8.90–11.50), thus indicating a less intense red coloration. Özdemir and Devres (2000) found values of a* for Turkish hazelnuts without skin between −1 and +1, therefore lower than the value of a* of sample H-TR-n (4.80). Mexis and Kontominas (2009)

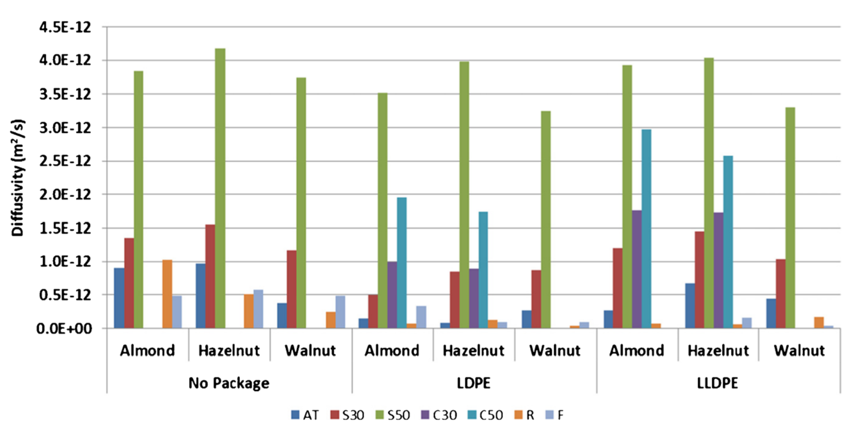



**Fig. 4** Diffusion coefficient for moisture loss or uptake for the nuts from Portugal with internal skin (storage: *AT* ambient temperature, *S30* storage at high temperature 30 °C, *S50* storage at high temperature 50 °C, *C30* chamber at high temperature 30 °C and 90 % RH, *C50* chamber at high temperature 50 °C and 90 % RH, *R* refrigerated, *F* frozen)

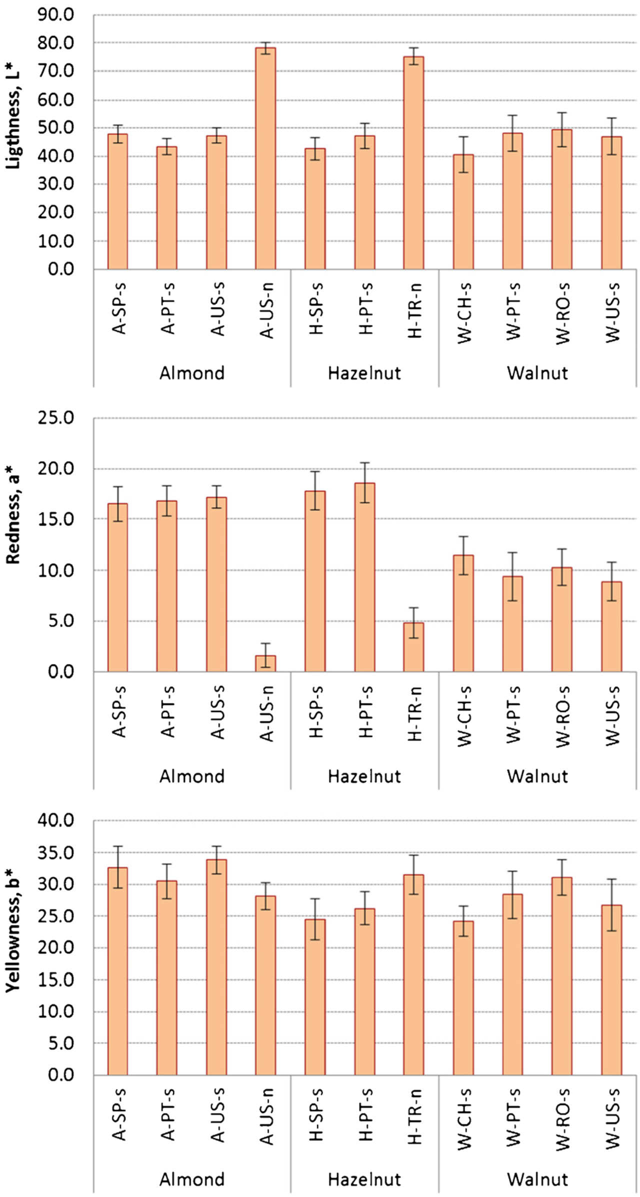



**Fig. 5** Colour coordinates of the nuts studied: lightness (*L**), redness (*a**) and yellowness (*b**;*A* almonds, *H* hazelnuts, *W* walnuts; country codes: *SP* Spain, *PT* Portugal, *US* United States, *TR* Turkey, *CH* Chile, *RO* Romania; *s* with internal skin, *n* without internal skin))

presented a value for the redness of hazelnuts equal to 6.84, which is lower than those reported in this study for the hazelnuts with skin (17.80 and 18.60).

Regarding yellowness (b* in Fig. 5), the degree of variability was greater, with values ranging from 24.50 to 33.90. Özdemir and Devres (2000) found values of b* for Turkish hazelnuts without skin ranging from 1.0 to 1.5, which are much lower than the value found in the present work for the corresponding sample, H-TR-n (31.60), thus indicating that the kernels studied in this work had a much stronger yellow coloration. Mexis and Kontominas (2009) found a value for yellowness of 8.70 for hazelnut kernels, being lower than those reported in this study for the hazelnuts with skin (24.50 and 26.20).

Figure 6 shows the average variation of the colour coordinates during storage under different conditions and with different types of package. The results indicate that there was in all cases a decrease in lightness, indicative of darkening, this being particularly important for the storage in the chamber at 50 °C and 90 % RH and in the stove at 50 °C. In fact, this temperature is quite high and therefore favours oxidation and Maillard reactions, thus leading to the formation of compounds with a brown coloration. Christopoulos and Tsantili (2011) evaluated the influence of storage conditions (temperatures of 1 and 20 °C and atmosphere: air, $N_2$ and $CO_2$) on the colour of walnut kernels. Their results indicated that during storage browning occurred with a corresponding decrease in lightness and hue. However, storage at temperature and packaging under $N_2$ or $CO_2$ prevented browning. Furthermore, the loss in colour was attributed to a decrease in the amount of antioxidants that was observed during storage. Mexis and Kontominas (2010) studied the storage of raw shelled unpeeled almond kernels packed in LDPE pouches. Their results showed that for the samples packaged in PET//LDPE under $N_2$ and stored in the dark at 20 °C small changes were recorded in all three colour parameters (L∗, a∗ and b∗; whereas, the corresponding changes for the samples packaged in the high barrier film (LDPE/EVOH/LDPE) were insignificant. The most pronounced changes in almond colour were observed for almonds packed under $N_2$ in both packaging materials and stored at 20 °C under light. L∗ and b∗ parameters decreased significantly with a parallel increase of a∗ values after 12 months of storage. A similar trend but to a lesser extent was observed for almonds stored under light at 4 °C.

The results in Fig. 6 also revealed that redness increased in almost all cases, with just the exception of the frozen walnuts, where a* decreased 8 %. This increase in redness was particularly intense for the storage of the almonds in the chamber at 50 °C and 90 % RH (with a* increasing by 159 and 189 %, respectively, for LDPE and LLDPE bags). Regarding the yellowness, the variations did not present a constant trend, increasing with some storage methods (as for example, the almonds in the chamber at 30 °C and 90 % RH) and decreasing with others (as for example, the storage of all nuts in the stove at 30 °C or in the chamber at 50 °C and 90 % RH).

Finally, comparing the effect of the different storage conditions (Fig. 6), it was concluded that all colour coordinates changed less for the nuts stored under refrigeration and freezing. Moscetti et al. (2012) evaluated the colour of unripe fresh unshelled hazelnuts during storage under modified atmospheres and concluded that there was no change in colour in the hazelnuts stored under nitrogen at 4 or 10 °C throughout the entire storage period. Also Mencarelli et al. (2008), found similar results for dried hazelnuts. They

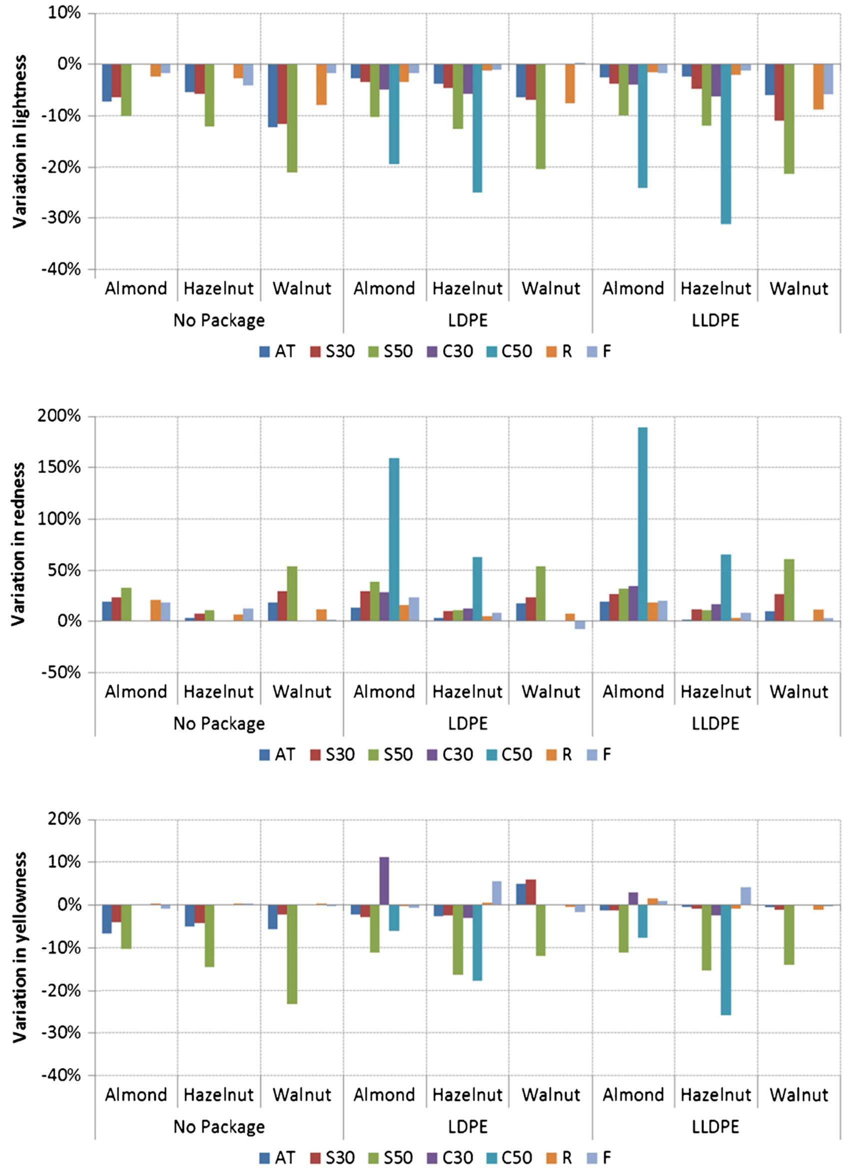



**Fig. 6** Average variation of the colour coordinates during storage: lightness (L*), redness (a*) and yellowness (b*; storage: *AT* ambient temperature, *S30* storage at high temperature 30 °C, *S50* storage at high temperature 50 °C, *C30* chamber at high temperature 30 °C and 90 % RH, *C50* chamber at high temperature 50 °C and 90 % RH, *R* refrigerated, *F* frozen)

observed some change in colour of the hazelnuts stored under $CO_2$ and in air, where there was an evident browning of the pericarp. However, these variations were observed in the colour coordinates hue and chroma, since brightness remained the same for all of the treatments. Furthermore, they registered a higher level of browning for the storage at 10 °C. Ledbetter and Palmquist (2006) studied the degradation of almond pellicle colour coordinates at different storage temperatures (2, 22 and 32 °C). Their results showed that the seed coat of almond kernels was subject to darkening during the long-term storage (11 months). Regression analysis revealed significant differences in degradation rates of pellicle luminosity and chroma at all three storage temperatures, but significant differences in pellicle hue angle degradation were only evident at the lowest storage temperature.

## Texture

Figure 7 presents the texture properties evaluated (hardness and friability) of the almonds and hazelnuts from the different origins. Hardness, or firmness, measures the mechanical strength of the product to being crushed. It is important to assure the physical integrity of the product and on the other hand it is one of the most important organoleptic attributes of foods. The results in Fig. 7 show that the almonds from the USA with skin were the harder, 50.23 N, whereas the almonds from Spain were softer, 21.14 N. Regarding the hazelnuts, the hardest sample was from Portugal (34.70 N) and the softer was from Turkey (19.70 N). Ozdemir and Akinci (2004) evaluated the rupture force of hazelnut kernels from 4 cultivars and their values varied from 50 to 64 N, thus corresponding to harder fruits than those hazelnuts evaluated in this study.

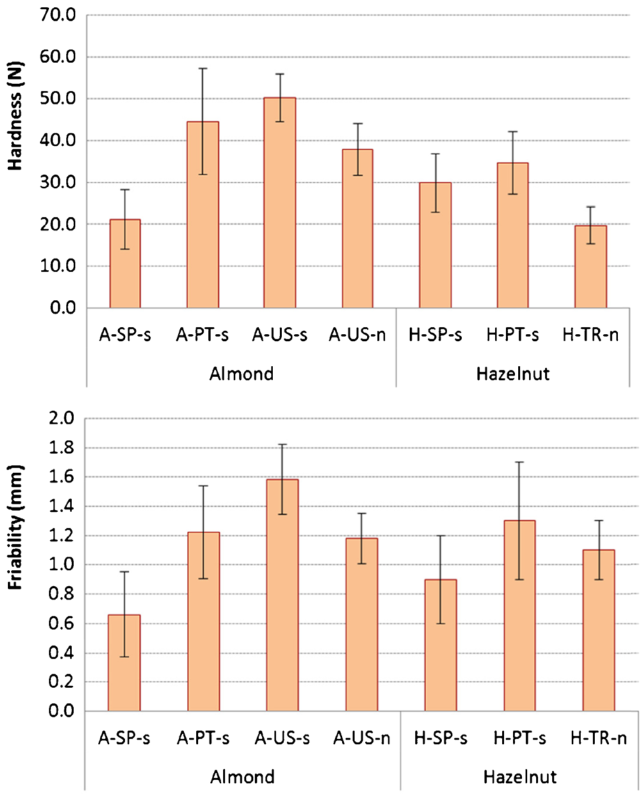


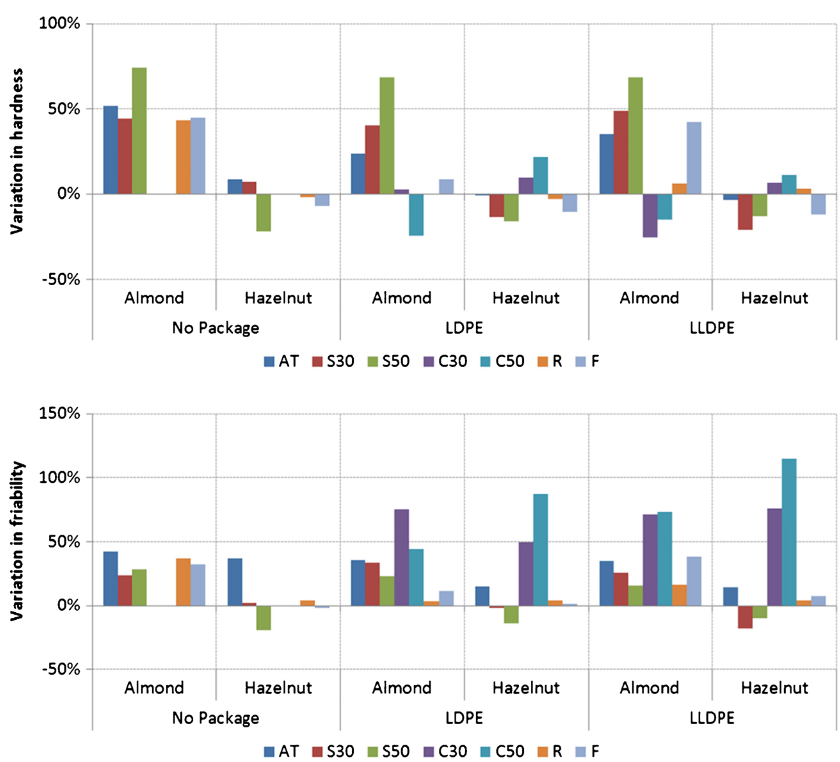

**Fig. 7** Texture properties of the nuts studied: hardness and friability (*A* almonds, *H* hazelnuts, *W* walnuts; country codes: *SP* Spain, *PT* Portugal, *US* United States, *TR* Turkey, *CH* Chile, *RO* Romania; *s* with internal skin, *n* without internal skin)

Ghirardello et al. (2013) reported a value of the rupture force for hazelnut kernels of 91.83 N.

The friability indicates the tendency of the product to fracture, and it is important to minimize product loss during handling and transportation. The results for friability in Fig. 7 show that among the almond sample the most fragile was the A-US-s (with 1.59 mm) and among the hazelnuts samples the most fragile was H-PT-s (with 1.30 mm).

Figure 8 presents the influence of the different storage conditions tested on the textural properties of almonds and hazelnuts. In general an increase in hardness was observed with the different storage conditions, although some decrease was also observed particularly for the hazelnuts. Storage in the stove at 50 °C produced the most intense changes, followed by the stove at 30 °C. However, these are general trends because there is a great variability in the results obtained for the variations in hardness. While for the almonds, it is difficult to indicate the best possible storage method; for the hazelnuts, there is a general trend that indicates refrigeration and freezing are the best methods to cause less change in hardness. Furthermore, this trend is maintained regardless of the type of package used. Ghirardello et al. (2013) studied the effect of storage under different conditions on the texture of hazelnut kernels. Their results revealed that after 12 months storage the in-shell hazelnuts stored at ambient temperature increased rupture force by 16 %, whereas the shelled hazelnuts stored under $N_2$ and refrigerated showed lowest variations

**Fig. 8** Average variation of the texture properties during storage: hardness and friability (storage: *AT* ambient temperature, *S30* storage at high temperature 30 °C, *S50* storage at high temperature 50 °C, *C30* chamber at high temperature 30 °C and 90 % RH, *C50* chamber at high temperature 50 °C and 90 % RH, *R* refrigerated, *F* frozen)



(−4 % and +8 %, respectively). Moscetti et al. (2012) evaluated the firmness of unshelled hazelnuts during storage under modified atmospheres. They concluded that the use of modified atmosphere was effective in controlling the increase in firmness of the kernel over time, while the worst maintenance of texture was observed in the fruit stored in air. Furthermore, the only temperature that maintained the firmness unaltered under both $N_2$ and $CO_2$ was 4 °C. Conversely, the firmness tended to increase at 10 °C.

The results in Fig. 8 also indicated that friability increased, in general, for all storage conditions. The increase was more intense for the storage in the chamber at 30 or 50 °C and 90 % RH, and these results did not differ much among the two types of plastic tested (LDPE or LLDPE). As previously observed for hardness, also for friability the storage under refrigeration and freezing were the methods originating in general smaller changes.

### Artificial Neural Network Results

Table 4 shows results obtained for function approximation using the neural networks. Due to the randomness characteristics of these neural networks, the results shown in the table were captured from selected experiments, thus, showing the best performances obtained, valid for the sensitivity analysis of the input weights presented in Section 3.5. Columns 2 to 4 show the *R* value for the linear regression between the ANN predicted values and the experimental results, for the whole dataset, as well as for the train and test subsets. The last column shows the root mean square error.

As the table shows, MC, $a_w$ and TCD are learnt by the neural networks with very high accuracy. The correlation is above 0.87 for the whole dataset. Variables *H* and *F* are much more difficult to model, as already pointed earlier. There are two main reasons that cause uncertainty in the model: outliers due to human errors and variability of the fruits themselves, even among samples of the same origin. The original dataset, being composed of experimental data, is subject to the presence of outliers due to human errors or other source of spurious results. Analysing the errors between the expected value and the predicted value, it was possible to identify some candidates to outliers and improve the expected quality of the dataset by removing those from the training process. This procedure was unnecessary for MC, $a_w$ and TCD. However, for *H* and *F* the 40 worst results (1.5 %) were removed although the improvements obtained were still negligible. For *H* and *F*, the data in general is skewed and with a large standard deviation. The average *F* value is 1.249 with a standard deviation of 0.612 for the whole dataset. In 20 samples of almonds coming from Spain, unpacked, with skin and stored at ambient temperature, the average *F* is 0.784 and the standard deviation 0.500. Thus, there is a large diversity even among samples of the same subset. Considering the difficulty of the dataset, and the poor improvement obtained by removing some of the worst samples, it seems reasonable to assume that the impact of outliers has been ruled out and the quality of the dataset is adequate. For *H* and *F*, the neural network showed *R* for the whole dataset and the test set greater than for the train dataset, which is an indication that the model abstracted is actually very good for the quality of the data and difficulty of the problem. The learning process was stopped at a point where there is no over-fitting to the training examples. Instead, the neural network generalized very well from the training examples to the unknown examples it had not seen during the training.

**Table 4** Correlation between the experimental value measured and the value predicted by the neural network

| Characteristic | Whole dataset | Train dataset | Test dataset | RMSE[1] |
|---|---|---|---|---|
| MC (moisture content) | 0.963 | 0.962 | 0.960 | 0.477 |
| $a_w$ (water activity) | 0.948 | 0.951 | 0.949 | 0.035 |
| TCD (total color difference) | 0.870 | 0.872 | 0.861 | 3.578 |
| H (hardness) | 0.575 | 0.569 | 0.577 | 15.076 |
| F (friability) | 0.483 | 0.475 | 0.502 | 0.556 |

[1] RMSE = Root mean square error measured after trainning.

### Neuron Weights Analysis

One interesting characteristic of neural networks is that a wealth of information about the data can be discovered by analysis of the weights of each neuron inputs. The network learns by adjusting those weights, so they reflect the contribution of the inputs to predict the output. The analysis of the input weights of trained neural networks is one common method to discover information in data mining.

Table 5 presents a summary of the weights of each input for the single neuron in the hidden layer of the neural networks used. Each neuron input is weighted and also given a bias, but the bias, being a constant, is not relevant to the present analysis.

Table 5 shows clearly that the moisture content was highly influenced by the storage temperature for the fruits stored in the stove. Temperature increase in the stove, where the relative humidity was low, caused a huge loss of water, and thus, S50 is the most important predictor captured by the neural network for MC. Variable C50 is not such a good predictor for MC, despite the fact that the temperature was also high—the relative humidity in the chamber prevented the fruits from losing moisture, and the neural network did not capture that variable as an important predictor for the moisture content—on the

**Table 5** Summary of the hidden neurons' input weights for the neural network for each variable

| | | MC[1] | $a_W$[1] | TCD[1] | H[1] | F[1] |
|---|---|---|---|---|---|---|
| Type of nut | Almond | 0.49 | −0.24 | 1.00 | −0.47 | 0.06 |
| | Hazelnut | 0.34 | −0.19 | 0.91 | −0.13 | −0.14 |
| | Walnut | 0.33 | −0.16 | 0.81 | – | – |
| | Skin | 0.07 | −0.02 | 0.12 | −0.19 | 0.15 |
| Origin[2] | SP | 0.60 | −0.06 | 1.43 | −1.79 | −2.54 |
| | PT | 0.55 | −0.03 | 1.42 | −1.97 | −2.15 |
| | USA | 0.53 | 0.00 | 1.44 | −1.92 | −2.20 |
| | TR | 0.44 | 0.01 | 1.53 | 0.92 | −2.11 |
| | CH | 0.60 | −0.07 | 1.48 | – | – |
| | RO | 0.65 | −0.14 | 1.39 | – | – |
| Storage[3] | AT | −0.14 | 0.32 | −2.01 | −0.14 | 0.20 |
| | S30 | −0.22 | 0.31 | −2.02 | −0.15 | 0.02 |
| | S50 | −2.45 | 1.74 | −2.22 | −0.20 | −0.08 |
| | C30 | 0.08 | −0.22 | −2.07 | 0.04 | 0.65 |
| | C50 | 0.24 | −1.48 | −2.55 | 0.19 | 8.62 |
| | R | −0.09 | 0.18 | −1.98 | −0.06 | 0.12 |
| | F | −0.09 | 0.20 | −1.98 | −0.10 | 0.05 |
| Packing[4] | LDPE | 0.07 | −0.14 | 0.02 | 0.04 | −0.02 |
| | LLDPE | 0.11 | −0.12 | −0.01 | 0.01 | 0.01 |

[1] MC= moisture content, $a_w$ = water activity, TCD = total colour difference, H = hardness, F = Friability.

[2] SP = Spain, PT = Portugal, USA = United States, TR = Turkey, CH = Chile, RO = Romania.

[3] B = before storage, AT 0 ambient temperature, S30 = stove at 30 °C, S50 = stove at 50 °C, C30 = chamber at 30 °C and 90 % RH, C50 = chamber at 50 °C and 90 % RH, R = refrigerated, F = frozen.

[4] LDPE = low density polyethylene, LLDPE = linear low density polyethylene.

contrary, storage in the chamber at 50 °C slightly increased the amount of moisture in the fruits. The weights learnt to the origin of the nuts are also relatively high, which is according to previous observations that nuts from different origins contain different amounts of moisture. Actually, the origin has been captured as a better predictor for the amount of moisture than the type of nut itself. The presence or absence of pellicle skin had very little influence on the amount of moisture, though the weights also show that the skin helps keeping some moisture content, about as much as packing in LDPE plastic. Packing, using LLDPE bags, contributes more to keeping the moisture contents than LDPE.

As for water activity, weights show that it is almost indifferent to the origin of the nuts, though samples from Romania were the most susceptible. Water activity was different for each type of nut, as the weights captured for the type of nut show it is a good predictor. But, above all, $a_w$ was dependent on storage conditions. Storage at 50 °C was the best of all predictors, and the relative humidity defines whether $a_w$ will be impacted either positively or negatively.

Colour was also very affected by storage conditions, even more so for higher temperatures and humidity, as the weights captured for S50 and C50 show they are the best predictors for TCD. On the other extreme, TCD was almost unaffected by the type of package used, and the presence or absence of pellicle skin is also a negligible predictor for TCD. The type of nut and origin are very important predictors, and they will have an impact contrary to that of the storage conditions.

As stated earlier, the confidence of results for $H$ and $F$ was not as high as for the other output variables, due to the high variance of the data. Results for walnuts, as well as for samples from Chile and Romania, were not available, so the total number of input variables for $H$ and $F$ was reduced to 16. Nonetheless, the results clearly show that $H$ and $F$ are very dependent on the origin of the nuts. The origin is by far the best class of predictors, both for $H$ and $F$. Additionally, it can be observed that storage in the chamber at 50 °C had a very huge impact on friability, since the weight of C50 is much higher than the weight of any other predictor.

## Conclusions

The results of the present work allowed concluding that, in general, the moisture content and water activity varied according to the geographical origin of the nuts at study. The almonds from Spain and Romania had $a_w$ greater than 0.6, which could compromise their stability; whereas all the other samples presented values of $a_w$ lower than 0.6. Furthermore, both moisture and water activity were better preserved when the nuts were stored under refrigeration or freezing, regardless of the type of package. On the other hand, storage at high temperature had the greatest impact on the nuts properties.

As to colour, the results indicated that lightness varied from 40 to 49 in the fresh samples but decreased during storage, due to darkening, and it was also possible to conclude that refrigeration and freezing allowed better preservation of the colour of all the nuts at study.

Regarding texture, differences were encountered among the nuts from different origins in both hardness and friability. In general an increase in hardness and friability was observed during storage with the different conditions. However, while refrigeration and freezing had negligible impacts in the textural properties, storage at high temperature and humidity had a huge impact on friability.

Neuron weight analysis has shown that the origin was a good predictor for moisture content and texture; whereas, the storage condition was a good predictor for $a_w$ and colour.

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