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Deep quantification of a refined adulterant blended into pure avocado oil

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A B S T R A C T

In this work, a new method has been developed to detect adulterations in avocado oil by combining optical images and their treatment with deep learning algorithms. For this purpose, samples of avocado oil adulterated with refined olive oil at concentrations from 1 % to 15 % (v/v) were prepared. Two groups of images of the different samples were obtained, one in conditions considered as bright and the other as dark, obtaining a total of 1,800 photographs. To obtain these images under both conditions, the exposure or shutter speed of the camera was modified (1/30 s for light conditions and 1/500 s for dark conditions). A residual neural network (ResNet34) was used to process and classify the images obtained. A different model was developed for each condition, and during blind validation of the models, ~95 % of the images were correctly classified.

1. Introduction

Nutrition is a fundamental aspect of life, as biological functions cannot be performed properly without it (Reeves, 2009; Juneja, Wilczynska, Singh, Takahashi, Pella, Chibisov, Abramova, Hristova, Fedacko, Pella, & Wilson, 2019). Nevertheless, regardless of its importance, nutrition indirectly, and food products directly, are vulnerable to unlawful practices, which can potentially lead to critical health concerns and hazards. There are instances when food frauds are intentional, including adulteration, which consists of the addition of a substance that hinders the original quality and purity of the food for an economic benefit. Among the main types of adulterations there are substitutions, dilutions, or concealment of processes and/or components (Morin & Lees, 2018).

Food adulteration has been occurring for a long time. Already during Ancient Roman times, fraudulent processes were carried out in which food was mixed with other substances in order to cheapen the product (Shears, 2010; Morin & Lees, 2018). These practices were sanctioned, but not regulated under laws (Shears, 2010).

During the Industrial Revolution (1760–1840), food fraud flourished. During that period, movements against it took place. For example, the chemist Friedrich Accum publicly denounced such acts in several works he published, but he did not achieve the implementation of adequate legislation to sanction this problem (Gutiérrez Rodilla, 2018; Dayan & Dayan, 2021). During the nineteenth century, various food adulterations that were carried out were collected to develop dictionaries to help control these practices. Thanks to these directories, laws were introduced to regulate the food frauds that had been taking place (Gutiérrez Rodilla, 2018; Lehotay, 2018). Currently, regulations have continued to be established according to food developments, and food surveillance is tightly implemented (Spink & Moyer, 2011; Banerjee, Chowdhary, Chakraborty, & Bhattacharyya, 2016).

A product that has been seeing a great increase in popularity recently is avocado oil. This is due to its benefits in the field of cosmetics and nutrition. Specifically, in the latter case, it has been attributed benefits for human health, such as the prevention of cardiovascular diseases by reducing the absorption of low-density lipoproteins or due to its anti-inflammatory effects (Tapiero, Townsend, & Tew, 2003). The price of this oil exceeds the rest of the oils, reaching prices of 10 to 15 € per 250 mL. This is due to the price of the fruit and that the process of obtaining this oil is based on costly procedures such as centrifugation or aqueous separation processes, by pressure or solvent extraction. Through the application of complementary processes, such as enzymatic, thermal, or mechanical pre-treatments, it is possible to obtain a higher fat yield. All this is the reasoning for this product being subjected to different illegal adulterations (Quiñones-Islas, Meza-Márquez, Osorio-Revilla, & Gallardo-Velazquez, 2013; Qin & Zhong, 2016). Mainly the adulteration processes are carried out by the addition of lower quality and cheaper oils (Green & Wang, 2020). Therefore, the development and implementation of techniques capable of detecting such frauds is necessary.

The techniques and/or equipment used to detect adulterations in avocado oil are typically expensive and require specialized personnel. These methodologies are based on chromatographic tools (gas chromatography (GC) or high-performance liquid chromatography (HPLC)) (Obeidat et al., 2009), nuclear magnetic resonance (Tang, Green, Wang, & Hatzakis, 2021), spectrofluorimetric methods (Green & Wang, 2020),

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or Fourier transform infrared spectroscopy (FTIR) coupled to multivariate analysis (Quiñones-Islas et al., 2013), among others. The treatment of the data obtained is usually carried out by linear algorithms.

In recent years, nonlinear algorithms have been used as mathematical alternatives to serve as tools for adulteration detection and quantification. In particular, artificial intelligence and machine learning have been widely used in a multitude of fields thanks to the excellent results they achieve in data processing tasks (Torrecilla, Mena, Yanez-Sedeno, & Garcia, 2007; Abiodun et al., 2018). There are many authors who have used artificial neural networks to analyze the data they have obtained, achieving successful outcomes. Within the food field, researchers such as Medus and co-workers have employed convolutional neural networks (CNNs), a type of neural network that excels in image analysis and classification (Pradana-López et al., 2021a), to analyze hyperspectral images during real-time food packaging control (Medus, Saban, Francés-Víllora, Bataller-Mompeán, & Rosado-Muñoz, 2021). Al-Sarayreh and collaborators employed this same technique to detect adulterations in red meat (Al-Sarayreh, Reis, Yan, & Klette, 2018). Also, using the combination of near-infrared hyperspectral imaging and CNNs, Zhu et al. were able to classify different soybean varieties (Zhu et al., 2019). Meanwhile, Pradana-López et al. used photographic images to detect adulterations in coffee samples (Pradana-López et al., 2021b).

It should be noted that for many cases the analytical techniques used to evaluate avocado oil are destructive and require sample conditioning. A suitable alternative to detect fraudulent activities would be to carry out inexpensive and straightforward processes to analyze the oil samples, during different phases of production and/or distribution. In this line, the objective of this work is to detect fraudulent practices in avocado oil by processing its photographs with CNNs based on transfer learning (Pradana-López et al., 2021a). On the other hand, it is intended, in the future, to facilitate the use, both by companies and users, of techniques capable of detecting possible food frauds in a simple, fast, and non-destructive way, even for in situ quality assessments.

2. Materials and methods

This section presents the oils used (avocado and refined olive oils), the equipment used to obtain the images, and the deep learning models used to classify the images.

2.1. Avocado oil and refined olive oil

Avocado oil is a product obtained from the fruit of the avocado (*Persea americana*). This fruit has a high calory content, and is mainly composed of unsaturated fatty acids, vitamins C, E, and B6, carotenes, and potassium. (Ozdemir & Topuz, 2004; Ranade & Thiagarajan, 2015). Avocado oil is contained in the idioblast cells, evenly distributed throughout the mesocarp of the fruit (Platt & Thomson, 1992), and its composition includes a large quantity of oleic acid, palmitic acid, and β-sitosterol (Salgado et al., 2008; Ranade & Thiagarajan, 2015), as well as omega acids, phytosterols, tocopherols, and squalene (Tapiero et al., 2003; Dos Santos, Alicieo, Pereira, Ramis-Ramos, & Mendonça, 2014). The adulterant agent used in this work will be refined olive oil, which is a product that has undergone a significant processing phase to mask/neutralize potential defects in the original olive oil (Torrecilla, Rojo, Domínguez, & Rodríguez, 2010; Cancilla et al., 2014).

The oils used were purchased in supermarkets in Madrid (Spain). All of them were analyzed within the best-before date and from the time of acquisition until the measurements were taken, they were stored in a dry and dark cabinet. Specifically, avocado oil and refined olive oil from the brands Ethnos and Carbonell, respectively, were used.

Seven binary mixtures with different concentrations were studied. The study range was established between 1 % and 15 % by volume of refined olive oil (1.0, 2.5, 5.0, 7.5, 10.0, 12.5, and 15.0 %) mixed into pure avocado oil.

The mixtures have been carried out by vigorous stirring for not less than five minutes. Likewise, the concentrations of each of the adulterated samples have been chosen to respond to the fraudulent activities that are taking place in the avocado oil sector and also to be able to verify the potential of the prototype that has been designed.

2.2. Camera and image database

The device with which the photographs of the pure and adulterated oil samples were taken was a NIKON camera, model D5100 with an AF-s NIKKOR 18–105 mm 1:35–5.6G ED lens. All photographs were taken under the same conditions. These conditions were set at an aperture of F5.6, an ISO sensitivity of 800, and a focal length of 18 mm. The only camera parameter that was varied was the shutter speed, in order to obtain images of the samples in light conditions (1/30 s) and, in addition, in dark conditions (1/500 s). This opens up the range of application of the prototype to different extreme light conditions.

In order to maintain the lighting conditions required for the study, and to avoid environmental interference, the photographs were taken in a dark chamber illuminated only by two spotlights (Fig. 1). This device has a circular opening at the top, the diameter of which coincides with the size of the lens of the camera used; it also has two LEDs to maintain constant and natural light conditions (5 V, 1.2 W). Since the purpose of the LEDs included in the equipment is for simple and stable illumination of the samples, this is done with white light. Finally, a lateral opening in the device allows manipulating the samples.

A total of 1,800 images were taken, 900 with the shutter speed of 1/30 s and 900 with the shutter speed of 1/500 s. The images were collected in JPEG format with a size of 4928 × 3264 pixels.

2.3. Deep learning algorithms trained

Deep learning has enabled the development of multilayer computational models which are capable of performing recognition and classification of images, voice, or video systems among others, by recognizing patterns from a set of data (LeCun, Bengio, & Hinton, 2015; Rusk, 2015).

To detect avocado oil adulterations, algorithms based on convolutional neural networks (CNNs) were used. Such models are responsible for classifying the images of the different samples of pure avocado oil and of those adulterated with refined olive oil obtained in the experimentation phase (Albawi, Mohammed, & Al-Zawi, 2017).

CNNs are composed of several types of layers (Rahadian & Suyanto,

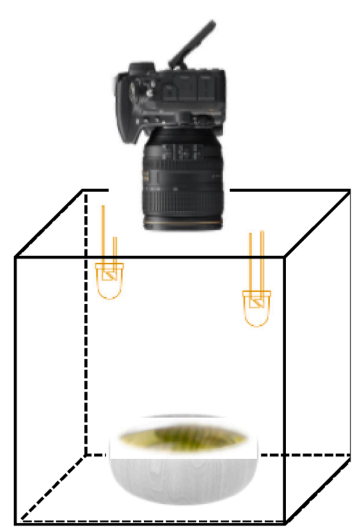

Fig. 1. Diagram of the device developed to maintain constant lighting conditions during imaging. Camera is positioned on an opening at the top, and images are taken of samples while being illuminated by two LEDs.

2019), differentiated into input, convolution, pooling or clustering, fully connected, and output layers. Among all of them, convolutional, pooling, and fully connected layers stand out (O'Shea & Nash, 2015). Convolutional and pooling layers alternate forming pairs (Pradana-López et al., 2021a), and their total number will depend on the study being performed and its complexity (Rusk, 2015). Because each layer relates to a specific function (Islam, Hossan, & Jang, 2018), it allows the next pair of layers to learn about a feature of the analyzed data (Rusk, 2015).

The collected images are fed into the network, and first analyzed by the convolution/clustering layers where the most characteristic features are selected from them, finding local patterns that can be expressed as edges or lines (Ting, Tan, & Sim, 2019). In order to perform this task, it is necessary that these layers possess filters based on fixed-dimensional matrices (Izquierdo et al., 2020). They are mainly applied to decrease the size of the images that are introduced to the neural network, which leads to an increase of the matrices per image (Pradana-López, Pérez-Calabuig, Cancilla, Otero, & Torrecilla, 2022). In the convolution layers it is necessary to apply the stride parameter, which is responsible for indicating the pixel displacement of the 3 × 3 filter on the image (Izquierdo et al., 2020). The pooling layers are applied after the convolution layers. They can be differentiated into two types, depending on the pooling function applied: Max pooling, which selects the maximum values of the matrices, and Average global pooling, which averages the values of the matrices (Izquierdo et al., 2020; Pradana-López et al., 2022). Finally, the activation function, which lies between the convolutional and pooling layers, needs to be applied. The most commonly employed function, and used in this work, is the rectified linear unit function, ReLu (Morandi et al., 2012): $f(x) = \max(0, x)$. The last layer to be applied is the fully connected layer. In it the information provided by the previous convolution/clustering layers is collected and classified into groups (Islam et al., 2018), where the number of outputs is equal to the number of classes of the classification to be carried out (Pradana-López et al., 2021b).

It should be known that the process that demands the most computational time during the training phase (weight optimization) of a CNN falls on the convolutional layers. This can be significantly lowered by using transfer learning, which is based on employing most of the optimized weights of the first general layers of a model that is previously trained for other purposes, and fine-tuning (training) the final weights to become suitable for a new task, thus only requiring a fraction of time to train. In other words, the first layers are frozen as they are in charge of detecting more general aspects of the images to be classified (they can work properly for many models, so tuning them is generally not a vital requirement). The final layers are modified based on the unique specifications of the model to be trained, and these are in charge of more specific aspects of the system to be analyzed (Rahadian & Suyanto, 2019; Pradana-López et al., 2021b). The artificial neural networks that operate as mentioned are residual neural networks, among which ResNetX (where "X" is the number of weighted layers present in the network) are noteworthy (Rahadian & Suyanto, 2019). In this work, a ResNet34 has been optimized (Rahadian & Suyanto, 2019) to be classify the 1,800 images of avocado and refined olive oil binary mixtures prepared. To perform this classification, it is necessary to modify the range of the learning coefficient of the residual network, which is chosen using a learning curve where the loss of information is related to the learning coefficient. After that, a number of epochs or training cycles are performed where the network is trained and validated. In addition, a hyperparameter known as batch size is used in this process, which is defined as the number of training samples to work with before optimizing the model with the selected learning coefficient range. When the batch size is larger than one sample but smaller than the size of the training dataset, it is known as mini-batch.

The 1,800 images were divided into two groups: 900 photographs that were taken at a shutter speed of 1/30 s and 900 images obtained at 1/500 s. Each group was then randomly divided into two. The first group is composed of the images that make up the training and verification datasets of the model (~90 %), and the remaining images are to be used for blind validation. The algorithms have been developed using a free software, Python 3.x.x in a Linux Ubuntu 21.10 environment.

3. Results and discussion

In this section, the results of the image acquisition are presented, as well as the application of the residual neural networks used for the classification of avocado oil samples in terms of refined olive oil content.

3.1. Oil image acquisition

Pure samples of avocado oil and refined olive oil, as well as samples of avocado oil adulterated with refined olive oil, were prepared and photographed, collecting a total of 1,800 images. Before processing and using the JPEG images of the samples, they were reduced from a size of 4928 × 3264 to 224 × 224 pixels, compatible with the CNN used.

Fig. 2 shows an example of the pure avocado oil and refined olive oil used for the study for both shutter speeds evaluated It can be seen with the naked eye that there is a difference in color between avocado oil and refined olive oil, even in dark conditions (shutter speed 1/500 s). While avocado oil presents a deep green color, the color of refined olive oil tends to be more yellowish, allowing the two oils to be distinguished easily. However, after adulterating pure avocado oil with refined olive oil in different concentrations, from 1 to 15 % by volume, it is observed that it is not possible to discern or classify these images with the naked eye (Fig. 3), even using the most favorable light conditions (shutter speed 1/30 s). Therefore, the use of an algorithm capable of analyzing and classifying the images qualitatively and quantitatively is necessary.

3.2. Convolutional neural network performance

The CNN used to perform the image classification is a residual neural network, specifically a ResNet34. It is a pre-trained network composed of 34 layers, designed to classify images using deep learning (He at al., 2016). The network has been trained to detect and classify images of pure avocado oil and adulterated samples with different concentrations of refined olive oil.

The 1,800 images taken are divided into two groups according to the shutter speed (1/30 s and 1/500 s). Then, each of these two 900-image datasets are randomly divided into two, the learning (~90 %) and blind testing datasets (~10 %). Considering the 1/30 s group, 97 images were initially removed to form the blind validation sample. Of the remaining 803 images, 643 were used to train the CNN (80.07 %), and 160 for its internal validation (19.93 %). On the other hand, for the 1/500 s shutter speed dataset, 816 images were used for training (653 images; 80.02 %) and internal validation (163 images; 19.98 %). The remaining 84 images (9.33 %) were used for blind validation. It is important to note that the images in the blind validation groups were never seen by the networks during their training process. These samples were only used during the final performance evaluation of each model.

3.2.1. CNN training and internal validation

Two ResNet34 models were trained and optimized for the 1/30 s and 1/500 s datasets. Since these networks are optimized by transfer learning, the computation time used in the optimization is significantly lower than other architecture comparable CNNs (He at al., 2016). The weights of the convolution/clustering layers at the end of the algorithm are optimized so that the model can be used specifically for the classification of avocado oil images based on refined olive oil content (in volume (%): 0.0, 1.0, 2.5, 5.0, 7.5, 10.0, 12.5, and 15.0). The main parameters used to train and validate the network are shown in Table 1.

The range of learning coefficient values for each network were decided based on the area of the learning curve that included a negative slope, meaning that the network uses an increasing amount information from the image to perform its training and, therefore, less loss. On the

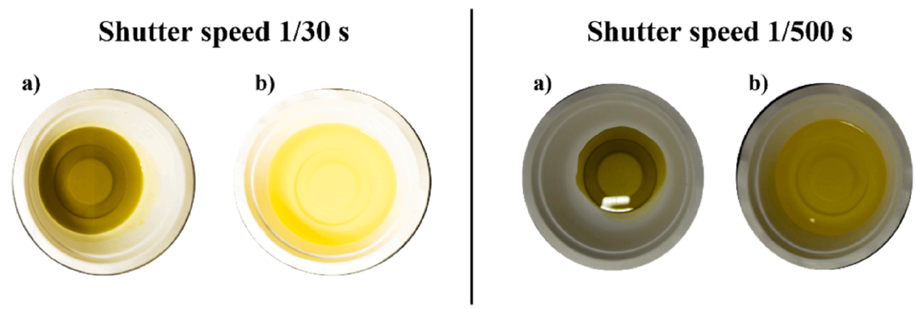

Fig. 2. Examples of the pure avocado (a) and refined olive (b) oils used in the study at the two shutter speeds (1/30 s left and 1/500 s right).

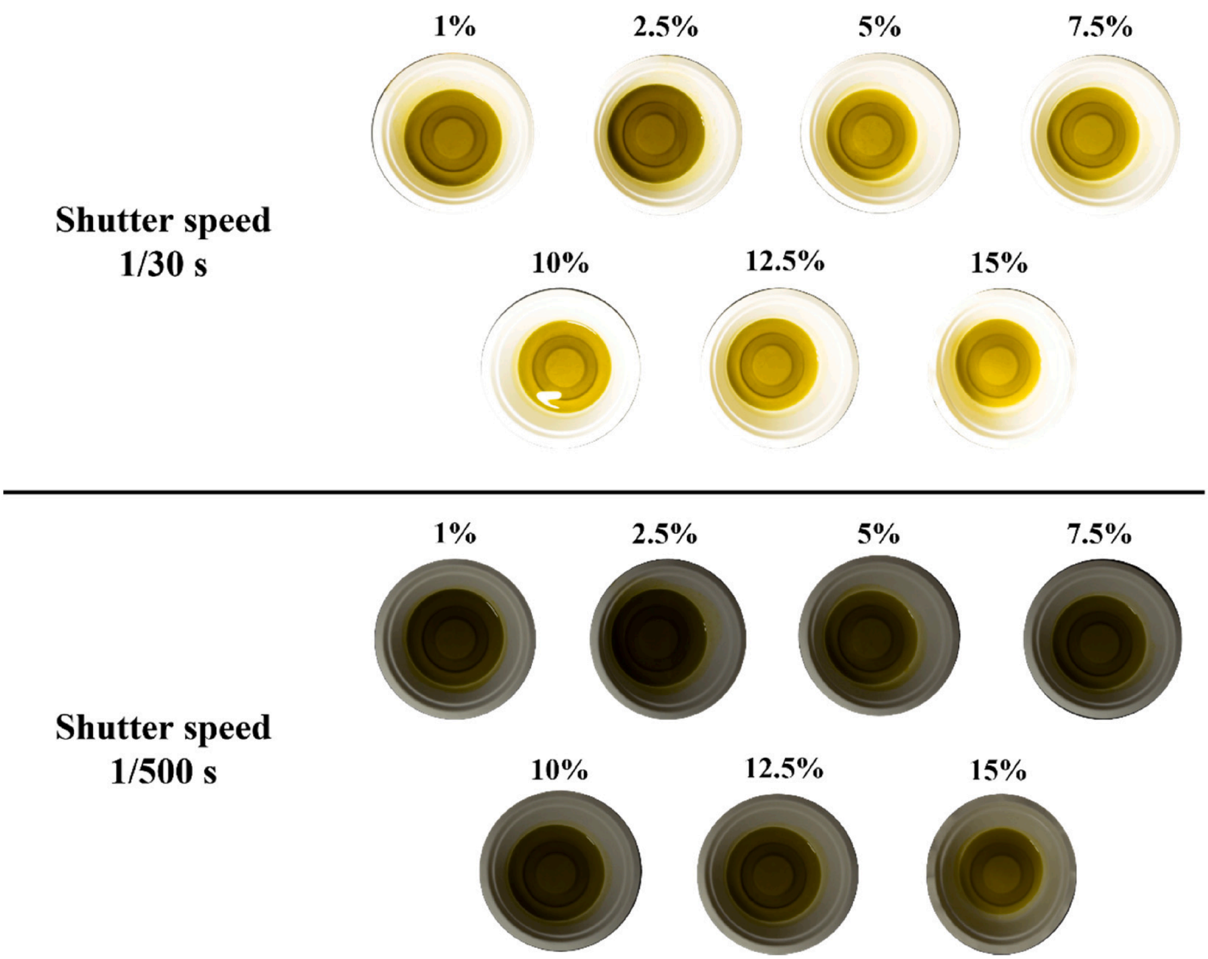

Fig. 3. Images of avocado oil adulterated between 1% and 15% (v/v) with refined olive oil prepared at the two shutter speeds selected for the study.

Table 1
Parameters set to optimize the ResNet34 models to classify avocado oil images in terms of refined olive oil content.

Parameters	ResNet34 1/30 s	ResNet34 1/500 s
Range of learning coefficient	$8\cdot10^{-6}$ – $1\cdot10^{-4}$	$1\cdot10^{-6}$ – $1\cdot10^{-4}$
Size of inputted images (vertical pixels × horizontal pixels × color channels (RGB))	224 × 224 × 3	
Stride	1	
Number of epochs	5	
Mini-batch size	64	

other hand, a number of epochs of five was chosen for both cases considering previous values of information loss and misclassification rate. Finally, the mini-batch size was chosen according to the ResNetX used, being 64 for the one with 34 layers (Cancilla et al., 2022). Once the networks were optimized, the validation of the models was carried out. For the internal validation of each model, confusion matrices were obtained and are presented in the **Supplementary Information**.

In the model developed for the images captured with a shutter speed of 1/30 s, out of the 160 images used for internal validation, 11 were misclassified (Table 2), resulting in a misclassification rate of 6.9 %. In Table 2 it can be seen that nine of the 11 errors committed are images that have been classified by the network as samples from the group of concentrations immediately above or below the real concentration of the sample. It is important to note that only one image belonging to a

Table 2
Incorrectly classified images during internal validation of ResNet34 for shutter speed 1/30 s.

Real label (adulteration %)	Number of misclassifications	Predicted class (adulteration %)
0 % (pure avocado oil)	1	2.5 %
1 %	2	2.5 %
1 %	1	5 %
2.5 %	3	1 %
2.5 %	1	5 %
10 %	1	7.5 %
10 %	1	12.5 %
12.5 %	1	10 %

pure avocado oil sample was classified as adulterated, which demonstrates the robustness of the model developed for this work.

In the case of the model developed for the faster shutter speed (1/500 s; darker images), 163 images were part of the internal validation set, of which 17 were misclassified (Table 3), so the error rate was 10.43 %. As can be seen in Table 3, all the misclassified images belong to adulterated samples, and in almost all cases, the classification made by the model corresponds to an adulterant class adjacent to the real one (15 out of 17).

3.2.2. Testing with blinded images

To evaluate the performance of the trained models, blind validations were carried out. For this purpose, ~10 % of the images initially and randomly separated (*vide supra*), and never shown to the neural network under testing, were used.

Of these images, the 1/30 s model managed to correctly classify 92 of the 97 photographs, obtaining an accuracy of 94.8 %. In Fig. 4**,** the misclassified images are shown, whereas the confusion matrix obtained from this process is presented in the **Supplementary Information**. As can be seen, four of the five misclassifications correspond to predictions of the class immediately adjacent to the real class, indicating the quantitative nature of the trained algorithm. Furthermore, none of the adulterated samples were classified as pure avocado oil, further enhancing the applicability of this tool regarding food safety. Also, only one out of the 97 samples would be incorrectly discarded, as a single pure avocado oil sample has been classified as adulterated.

The same procedure was carried out for the images obtained with a shutter speed of 1/500 s. In this case, the set of blind images consisted of 84 photographs, of which 80 were correctly classified, leading to a correct classification rate of 95.2 %. The misclassified images are shown in Fig. 4, while the confusion matrix is in the **Supplementary Information**. It is worth mentioning that one of the four errors implies the misclassification of an adulterated sample as pure avocado oil, suggesting that the overall safety of the 1/30 s model may be better. It is hard to pinpoint a reason for this misclassification, but as new quality data is collected, these errors will lower for further iterations of the algorithm.

Table 3
Images misclassified in the internal validation of ResNet34 for shutter speed 1/500 s.

Real label (adulteration %)	Number of misclassifications	Predicted class (adulteration %)
1 %	1	2.5 %
2.5 %	3	1 %
2.5 %	1	5 %
5 %	1	2.5 %
5 %	1	10 %
7.5 %	1	5 %
7.5 %	1	10 %
7.5 %	1	12.5 %
10 %	1	7.5 %
12.5 %	6	10 %

The purpose of taking pictures at two different shutter speeds is to compare the results of the two models as they can represent unique scenarios. In this sense, the images taken at 1/30 s are intended, for instance, for the retailer to verify the nature and quality of their product. On the other hand, those taken at 1/500 s are designed to create models that can be implemented directly during production or early distribution where lighting conditions may not be optimal for imaging, as well as requiring real-time monitoring. On the other hand, it's likely that low lighting environments are intended to preserve the properties of some delicate products, like avocado oils which are susceptible to oxidation from light.

Previous studies have already tested techniques to locate adulterated avocado oil. Green and Wang were pioneers in detecting avocado oils labeled as virgin or extra virgin adulterated with lower quality oils in the market using ultra-high-performance chromatography with charged aerosol detector (Green & Wang, 2020). On the other hand, Jimenez-Sotelo *et al.* employed attenuated total reflectance with Fourier transform infrared spectroscopy to detect up to 2 % of seed oils within avocado oils without sample pretreatment (Jiménez-Sotelo et al., 2016). Comparing these works with the one developed in this research, it is observed that, although both studies achieve promising results, the methods used require skilled personnel and expensive equipment. The technique presented here, on the other hand, has not only allowed to reliably identify adulterations in this oil with lower concentrations than previously mentioned works (Jiménez-Sotelo et al., 2016), but it is also a more cost-effective and easier-to-use method. Based on these results, it can be concluded that the model developed for the detection of refined olive oil in avocado oil is valid for the quantification of low amounts of this adulteration.

4. Conclusions

In this work, ResNet34s have been trained to carry out the detection and quantification of adulterations present in avocado oil at low concentrations. In order to carry out this research, it was only necessary to acquire photographs of the samples, obtained by means of a digital camera, and a viable computer to train the CNNs, which makes this technique more affordable than other procedures. The networks were trained to detect and classify the adulteration of avocado oil with refined olive oil for two different shutter speeds (1/30 s as light conditions and 1/500 s as dark conditions). The accuracies of the trained models for the blind validations were 94.85 % for the 1/30 s and 95.24 % for the 1/500 s.

The developed tools signify great progress in the detection of adulterations of this type due to the simplicity, speed, and straightforwardness of this method with respect to more traditional alternatives, carried out in a laboratory and with more complex equipment. In this case, via transfer learning and CNNs, the tool can be implemented for in situ and real-time analysis at any phase of the production or distribution chain serving as an effective quality and health control.

CRediT authorship contribution statement

Ana M. Pérez-Calabuig: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Supervision, Data curation, Visualization, Writing - review & editing. **Sandra Pradana-López:** Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Supervision, Data curation, Visualization, Writing - review & editing. **Andrea Ramayo-Muñoz:** Investigation, Visualization, Writing - original draft. **John C. Cancilla:** Conceptualization, Validation, Investigation, Supervision, Methodology, Software, Visualization, Writing - review & editing. **José S. Torrecilla:** Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Supervision, Funding acquisition, Data curation, Project administration, Resources, Visualization, Writing - review & editing.

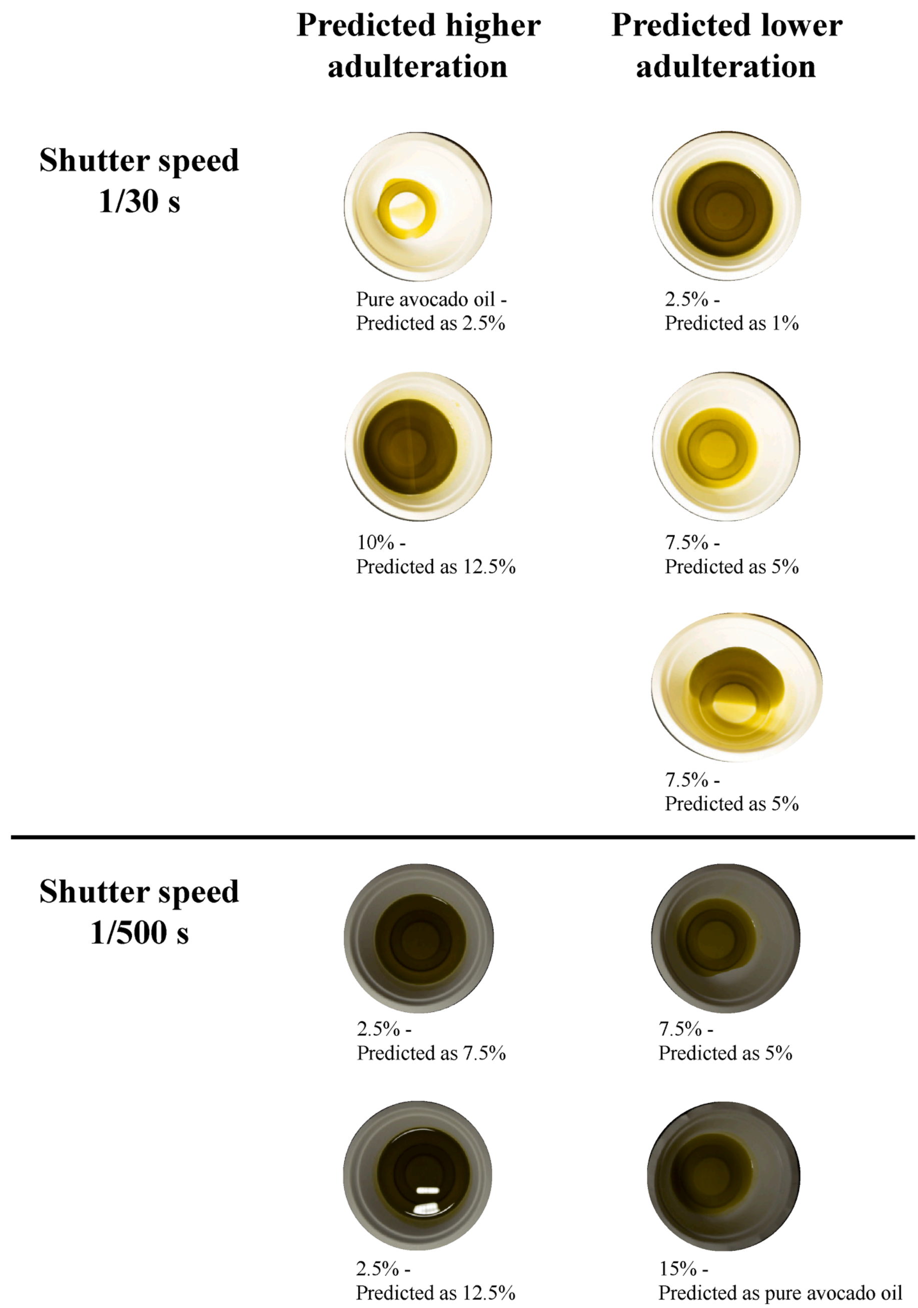

Fig. 4. Blind images incorrectly classified by the ResNet34s trained (1/30 s and 1/500 s). For each image, the real label is shown followed by the prediction done by the model.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2022.134474.

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