

The Color Space of Foods: Virgin Olive Oil

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The CIE 1976 (L^* , a^* , and b^*) color space for virgin olive oil was determined. Such a space encompasses any acceptable sample of this type of oil irrespective of the agronomic treatment that the olives have undergone because its color is due to two types of pigments, a systematic combination of which provides the whole range of theoretically possible colors. Color is quantified from the visible spectra for pure samples. Therefore, the pigment spectra, which were the averages of those for several samples, were determined in a medium highly similar to the source oil following application of a photochemical method. A combination of the pigment spectra provided 651 spectra for virtual samples, the colors of which spanned the entire color space for virgin olive oil. The positions of more than 100 Spanish olive oil samples of diverse origins in the color space were also determined, and the results were examined in relation to oil type and quality. Similar color spaces can be obtained for other foods, which can thus be characterized in terms of an additional physical property.

KEYWORDS: Virgin olive oil; reconstructed spectra; chlorophyll spectrum of olive oil; carotenoid spectrum of olive oil; CIE- $L^*a^*b^*$ color coordinates; virgin olive oil color space

INTRODUCTION

Color is a sensory property with a strong influence on food acceptance as it contributes decisively to the initial perception that one can acquire of the condition, ripeness, degree of processing, and other characteristics of foods (1). This has led to the establishment of a maturity index based on measurements of color and other physicochemical properties for some foods (2–4). Similarly, a color index has been developed as a formula based on color coordinates (5). The maturity index—and also the shelf life in some cases (6)—varies among foods and even among food varieties as a result of differences in color (7). Such is the case with many fruits and their juices. One case in point is virgin olive oil (8), which can range from pale yellow to deep green in color and still strictly meet current European Union regulations on oil quality (9). In fact, as with other fruits, the color of olive oil depends not only on fruit ripeness but also on fruit variety, cultivation area, harvest time, and the particular processing methods used (10, 11).

Color differences in many foods are due to differences in their proportions of pigments, which are mostly carotenoids (12–15), anthocyanins (16–19), and, to a lesser extent, flavonoids (20–22), betanins (23), and chlorophylls (24). These pigments and other major components of virgin olive oil (25, 26) including polyphenols (19, 27–29) are determined by using various analytical techniques [especially chromatographies (30–33) but also visible spectroscopy (34–40)]. In fact, visible transmittance or absorbance measurements allow color to be quantified by calculating the color coordinates of the sample concerned in a CIE-1976 color system such as CIE- $L^*a^*b^*$ (41–43); this

entails using as large a number of spectral data as possible (i.e., measuring the absorbance or transmittance at short wavelength intervals) in order to minimize errors (44).

If the visible spectra for each variety of a natural food fit for consumption were available, they could be used to quantify their acceptable hue, saturation, and brightness or their color coordinate ranges in some color system. Such ranges in turn could be used to determine the color space spanned by acceptable samples of the food, which would thus constitute an additional useful physical property for characterization purposes.

Even if the visible spectrum for every possible variety of a given food is unavailable, it can be generated or simulated by using a reconstruction procedure. This entails combining, in appropriate proportions, the visible spectra for all types of pigments present in the food. Such proportions are bound by the maximum and minimum pigment contents in the body of available samples of the food. The spectral data for each type of pigment should preferably be obtained from solutions in a medium resembling the natural environment of the food. This is not easy because the natural medium for pigments usually comprises a wide variety of substances and thus differs markedly from the typical spectroscopic solutions, which usually contain a single pigment or a group of pigments of the same type in as transparent as possible a solvent. Olive oil is amenable to selective separation of one of the two types of pigments it contains in a medium that virtually coincides with the original environment. This is a result of the large differences in photodegradation time between chlorophyll and carotenoid pigments (45), which allows the visible spectrum for each pigment type to be obtained as if it were the sole type present in the oil. From these visible spectra, one can easily reconstruct that for any sample of virgin olive oil.

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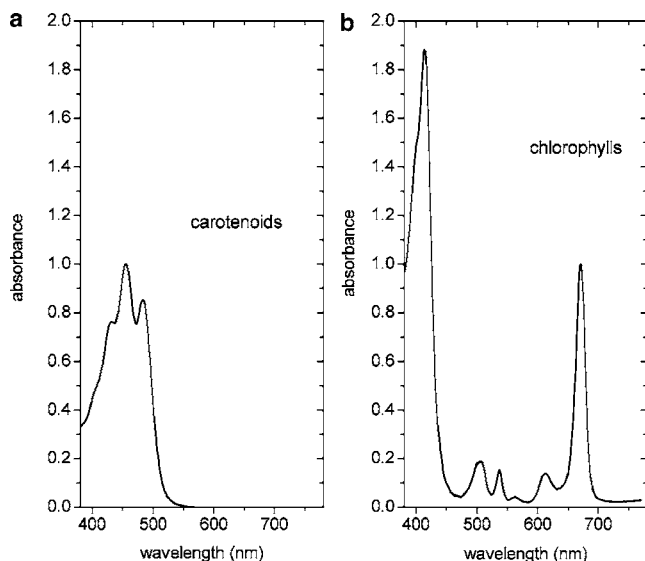


Figure 1. Averaged, normalized spectra for carotenoid pigments (a) and chlorophyll pigments (b) in virgin and extra virgin olive oil.

On the basis of the foregoing, we addressed the work with the following purposes: (i) to determine the color space of virgin olive oil; (ii) to check such space against a wide variety of commercially available samples of this type of oil; and (iii) to draw useful conclusions on the location in such a space of commercially available virgin olive oils, which are the most widely used.

MATERIALS AND METHODS

Samples. A total of 107 samples of Spanish olive oil were used, five of which were pomace oil, 12 that were olive oil, and 90 that were either virgin or extra virgin olive oil; all samples were purchased at randomly selected outlets in accordance with no special plan or sequence. Some samples were obtained from a single olive variety and others from a combination of several; also, some samples only differed in olive harvest fruits and others in the best before date as stated on the product label. The olive varieties used to obtain the oils were as follows, in decreasing frequency: Picual, Arbequina, Hojiblanca, Cornicabra, Picuda, Manzanilla, Lechín, Verdial, Zarzaleña, and Alameña. Three oil samples had to be centrifuged on a Selecta Centromix S-549 apparatus at 4200 rpm for 1 h because they were sold unfiltered; this allowed the spectral background to coincide with the baseline and spurious variations in color coordinates to be avoided.

Spectra. All spectra for the oil samples were recorded on an ATI Unicam UV4 spectrophotometer interfaced to a PC. Samples were used in pure form and withdrawn from freshly opened containers; each was used to additionally fill five topaz flasks of which three were frozen for subsequent use as references and other tests; the other two were refrigerated for immediate checking tests. The spectroscopic cells used were largely of the Dispolab Kartell 1937 disposable polystyrene type and 10 mm thick. However, at least one spectrum per sample was recorded in the UV region, using Suprasil cells of identical thickness. All spectra were obtained with an empty cell in the reference beam—the same cell for all samples—that was also used to record baselines. With such a convenient reference, the spectra exhibited a negative absorbance of less than -0.03 AU in the zone from 750 to 770 nm; therefore, the absorbances obtained with it were all corrected in order to avoid negative values in the visible range. The spectrum for each sample was recorded at least 3–4 times in order to minimize the effect of operational errors and changes in sample condition. The highest noise level encountered was 1×10^{-3} AU, and the mean square noise level was less than 0.5×10^{-3} AU.

Figure 1 shows the average normalized spectrum for each pigment group as obtained in a medium similar to the natural environment for virgin olive oil (45). These spectra were normalized for unity absorbance

at 455 and 670 nm as reconstructing spectra for virgin olive oil samples simply requires measuring the absorbances at these two wavelengths in order to quantify the two types of pigments. Normalization substantially increased the band at 414 nm, which is usually weak except in deep green-colored oils. The ordinates of the normalized spectrum for each pigment as measured at 1 nm intervals were multiplied by the respective experimental absorbance (that at 455 nm for carotenoids and that at 670 nm for chlorophylls). The combination of these two results at each wavelength provided the reconstructed spectrum for the sample. The content in chlorophyll pigments was best determined from the band at 670 nm, which was subject to no overlap; by contrast, the band at 414 nm was stronger but was within the broad band for carotenoids and was ill-defined in the spectrum—even for samples with a high chlorophyll content. The spectral data for carotenoid pigments should be managed carefully since, as can be seen from **Figure 1**, the spectrum touches the baseline at ca. 560 nm and slight baseline variations can alter the values of some properties derived from reconstructed spectra (e.g., color coordinates).

Processing of Spectral Data. Spectral noise and statistics were calculated by using a spreadsheet. The goodness with which a spectrum was reconstructed was measured in terms of (i) closeness (via the coefficient of determination, R_{xy}^2) and (ii) distances (via the root-mean-square deviations, $\text{rmsd} = \{[\sum(A_i - A_{ri})^2]/n\}^{1/2}$, or standard error of prediction, SEP). In our case, A_i was the absorbance of the sample concerned and A_{ri} was that of its reconstructed spectrum, $n = 196$ being the number of absorbance values measured at 2 nm intervals from 380 to 770 nm.

Color. The color of pure liquid samples was determined from spectral data and characterized in terms of CIE $L^*a^*b^*$ color coordinates by using illuminant C with the 2° standard observer and illuminant D65 with the 10° supplementary observer. These two illuminants were defined by CIE in 1931 and 1964, respectively, with a view to representing natural diurnal light. D65 is somewhat more efficient in the relative distribution of spectral energy in the UV region, so it is especially indicated for characterizing color in places under diurnal light similar to that in northern Europe. Computations were done by using software developed by the authors, which processes spectral data in ASCII format. Least-squares polynomial fitting was done by using the commercial software Origin v. 6 from Microcal Software, Inc. (Northampton, MA).

RESULTS AND DISCUSSION

Evaluation of the Procedure Used To Reconstruct the Visible Spectrum of Virgin Olive Oil. The procedure was applied to the 90 samples of virgin olive oil studied. Consistency between the real and the reconstructed spectra was checked in four different ways for each sample, namely, (i) by visual inspection of each pair of spectra, which were displayed superimposed on the PC screen; (ii) by assessing the goodness of reconstruction via rmsd (a total of 87 samples exhibited $\text{rmsd} < 0.006$, so their spectra were very reliably reconstructed; the other three were virgin olive oil but had lost quality relative to the previous one, so they were discarded); (iii) by linear fitting, using least-squares regression, of the absorbance values at each wavelength as measured at 2 nm intervals in the real and reconstructed spectra [the previous 87 samples exhibiting an acceptable coefficient of determination ($R^2 > 0.995$)]; and (iv) by comparing the CIE- $L^*a^*b^*$ color coordinates obtained from each real spectrum and its reconstructed counterpart. A comparison of such coordinates exposed slight differences between the spectra; however, the differences in each color coordinate for the body of samples were acceptable. This is apparent from **Figure 2**, which shows pairs of values of the color coordinate b^* obtained with illuminant D65 and the 10° supplementary observer. As can be seen, most points fell on the regression line, so the correlation coefficient was very close to 1.000.

On the basis of the previous results, the reconstructed spectrum for a virgin olive oil can be highly reliably used instead

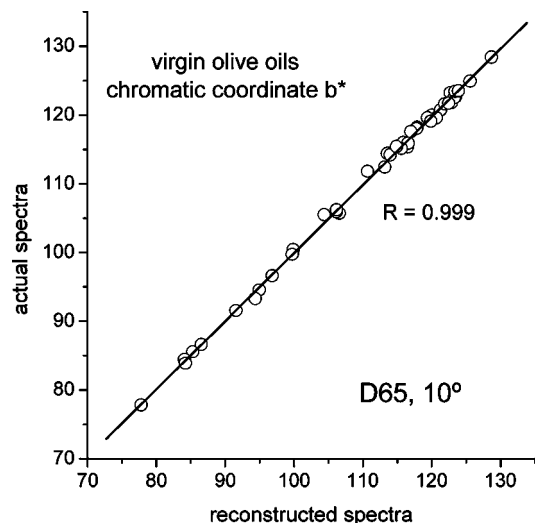


Figure 2. Least-squares fitting of the b^* coordinates obtained from actual spectra to those from reconstructed spectra for virgin and extra virgin olive oil.

of its real spectrum as the two differ very little. Thus, even the color coordinates calculated from reconstructed spectra are appropriate replacements for the values provided by real spectra.

These results can be of great help with a view to establishing the color space of virgin olive oil as this involves using the spectra for a large number and variety of samples, and the reconstruction procedure allows a collection of visible spectra for virtual samples obtained by systematically combining those for the two types of pigments in virgin olive oil to be compiled. With this purpose, sample spectra were deemed different if they departed by at least 0.1 AU from each other in the absorbance at 455 nm (A_{455}) or that at 670 nm (A_{670}). The lower limit for the absorbance was taken to be that for a sample containing no pigments, even though it represented a highly unlikely case for virgin olive oil. On the other hand, the upper limit was defined by an oil with a high pigment content exhibiting $A_{455} = 3.0$ AU and $A_{670} = 2.0$ AU; these values were dictated by the authors' experience in the analysis of virgin and extra virgin olive oil. The upper limit, which is occasionally reached by pure samples when using a 10 mm thick cell, can be measured by modern spectrophotometers with adequate photometric sensitivity in the region of interest. In any case, a potentially nonlinear photometric response has no effect on the location of a sample in the color space but can be a source of error when determining color-related parameters in real samples.

With the previous absorbance limits, the CIE $L^*a^*b^*$ color space for virgin olive oil was bounded by the color coordinates established from 104 reconstructed spectra for samples of the following four types: (i) No-chlorophyll group, which consisted of 31 samples containing no chlorophyll pigments ($A_{670} = 0.0$) and variable amounts of carotenoid pigments giving A_{455} values over the range 0.0–3.0 AU. (ii) High-chlorophyll group, which consisted of 31 samples with high contents in chlorophyll pigments ($A_{670} = 2.0$) and variable amounts of carotenoid pigments giving A_{455} values over the range 0.0–3.0 AU. (iii) No-carotenoid group, which comprised 21 samples containing no carotenoid pigments ($A_{455} = 0.0$) and variable amounts of chlorophyll pigments giving A_{670} values up to 2.0 AU. (iv) High-carotenoid group, which consisted of 21 samples with high contents in carotenoid pigments ($A_{455} = 3.0$) and variable amounts of chlorophyll pigments giving A_{670} values over the range 0.0–2.0 AU.

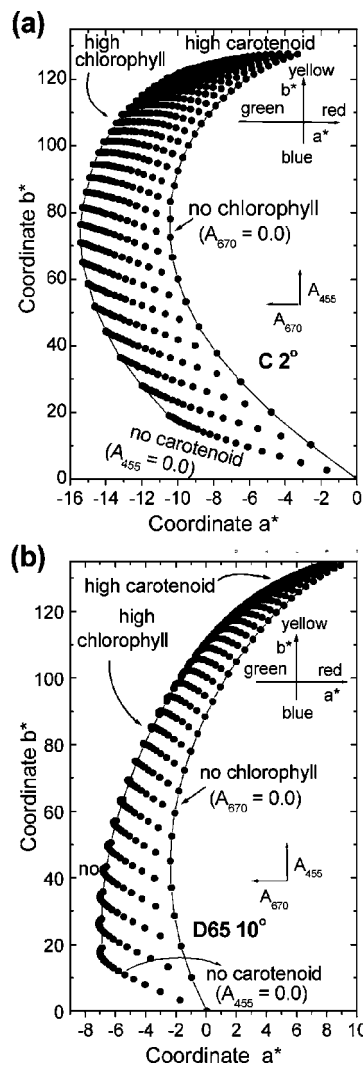


Figure 3. b^* vs a^* color space for virgin olive oil virtual samples as obtained with the illuminants C 2° and D65 10° .

In addition to the 104 samples defining the boundaries of the color space, we examined the variation of the color coordinates for virtual samples falling within the space. To this end, we reconstructed another 547 spectra for virtual samples the color coordinates of which fell in such a space. The body of 651 virtual samples ranged from a colorless oil containing no pigments to a deep yellow oil containing abundant pigments and encompassed oils with variable amounts of chlorophylls but no carotenoids (a result, for example, of prolonged storage in the dark) and oils containing virtually no chlorophylls but variable amounts of carotenoids (e.g., oils obtained from olives harvested by the end of the season or exposed to light over long periods).

Figures 3 and **4** show the 651 coordinates used. Each figure contains two diagrams; one was obtained with illuminant C and the 2° standard observer, and the other was obtained with illuminant D65 and the 10° supplementary observer. In **Figure 3a,b**, b^* is plotted against a^* as in the CIE 1976 chromaticity diagram; in **Figure 4a,b**, coordinate L^* is plotted against chroma (C). These figures expose the effect of using a different illuminant and observer, the effect being more marked in a^* than in b^* , L^* , or C . The boundaries of the color enclosure are labeled no chlorophyll, high chlorophyll, no carotenoid, and high carotenoid in the figures. **Figure 3a,b** (top right corner) includes a color graph showing the variation of color with the color

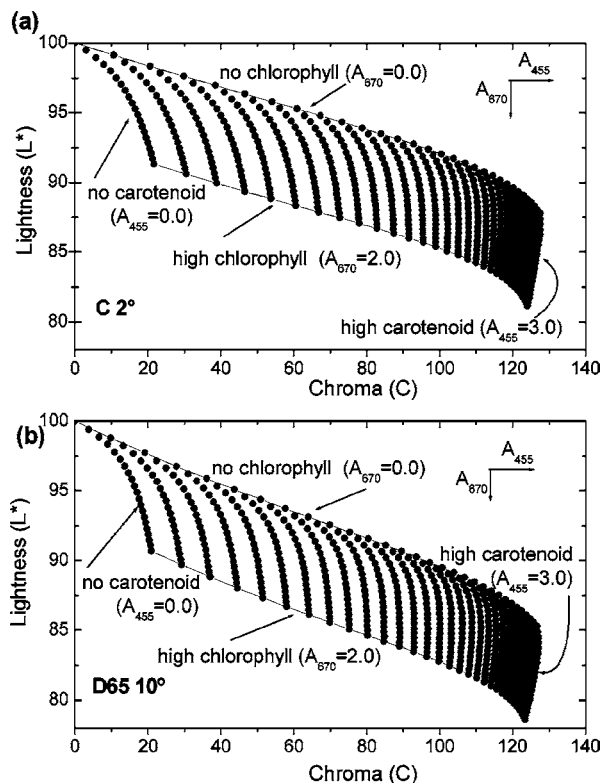


Figure 4. L^* vs C color space for virgin olive oil virtual samples as obtained with the illuminants $C\ 2^\circ$ and $D65\ 10^\circ$.

coordinates of the samples, with coordinate a^* (green-red) on the x -axis and coordinate b^* (yellow-blue) on the y -axis.

To facilitate interpretation of the relationship between the absorbance at the maximum of the spectral bands and the variation of the color coordinates, each pair of graphs in **Figures 3** and **4** shows the direction of the intensity change in the two most characteristic bands in the spectrum for virgin olive oil. As can be seen, the color of this oil is strongly influenced by the absorbance of the carotenoid band at 455 nm. In fact, as can be seen from **Figure 3a,b**, coordinate b^* increases with increasing absorbance—and so does chroma as a result since $C = (a^2 + b^2)^{1/2}$ (see **Figure 4a,b**). This is a consequence of carotenoids absorbing in the blue region and an increase in band intensity, thereby leading to a more yellow color in the oil.

The no chlorophyll and high chlorophyll lines in **Figure 3a,b** are curved by effect of the variation of A_{455} also depending on a^* , which initially becomes increasingly negative with an increase in A_{455} and then increases after a minimum value. The increase in the band at 455 nm is accompanied by an increased width, which results in increased absorption in the green region and hence in an increase in a^* . Such is the case with oils that are perceived as orange rather than green in color, which is especially apparent under lighting conditions similar to those of illuminant D65 (see **Figure 3b**). The points in a line parallel to the no carotenoid line in both graphs of **Figure 3** have an identical carotenoid content ($A_{455} = \text{const}$) but a different chlorophyll content that increases by 0.1 AU from one point to the previous one; this results in an increased green hue and a decreased value of a^* . These dotted lines exhibit two salient features, namely: (i) None is parallel to the x -axis as the color of oil is influenced by the absorption of chlorophyll pigments in the blue region (see **Figure 1**). Thus, an increase in the band at 414 nm increases the surrounding spectral profile by effect

of the broad carotenoid band, the outcome being a slight increase in yellow hues and also in coordinate b^* .

(ii) As can be seen at the left end of each line with $A_{455} = \text{const}$, gradually increasing the chlorophyll content in the oil results in a^* going by a minimum that falls slightly outside the high chlorophyll boundary line and then bends and returns to the boundary (see **Figure 3b**). The line curves upward when the carotenoid band is weak. This is a result of an increase in chlorophyll pigment content increasing not only the band at 670 nm but also the stronger one at 414—which is broadened as a result and invades the green zone of its underlying profile, the outcome being that coordinate a^* becomes slightly less negative even if b^* continues to increase. Above $A_{455} = 0.90$ AU, however, the line curves downward as the previous effect is less substantial by virtue of the increase in the carotenoid band—by 0.1 AU from one dotted line to the next; this leads to an increase in yellow hues and also in b^* .

The no chlorophyll and high chlorophyll boundary lines in **Figure 4a,b** are nearly straight except at very high chroma levels. As in **Figure 3**, the color coordinates for the virtual samples within the color graph form dotted lines that are quite parallel. A comparison of the two diagrams in **Figure 4** reveals that using a different illuminant has little effect on lightness (L^*) or chroma (C). As expected, an increase in pigment content results in decreased lightness and also in increased chroma, which is strongly influenced by the increased magnitude of b^* relative to a^* and, hence, as noted earlier, by the carotenoid content. The accumulation of points on the right side of **Figure 4a,b** is a result of the maximum absorbance limits used. Such limits are appropriate as the color coordinates for a virgin olive oil with $A_{455} > 3.0$ AU and/or $A_{670} > 2.0$ AU should fall within the dotted boundaries in the figures; however, an extremely green, highly unusual oil containing a vast amount of chlorophyll pigments could exhibit an L^* value falling below the high chlorophyll line in **Figure 4a,b**.

The color coordinates of any virgin olive oil sample must necessarily fall within this color space. However, for a sample to be deemed virgin olive oil, it must also comply with the applicable regulation (9). Once the color space for virgin olive oil was established, we checked it against commercially available samples. **Figures 5** and **6** show the boundaries of the color space and the color coordinates for the virgin, extra virgin, olive, and pomace oils examined. Although the samples spanned a variety of origins and fruit types, the color coordinates for the virgin and extra virgin olive oils and some olive and pomace oils fell within the boundaries of the color space. Such a space lies near the middle of the yellow region in the CIE color space, which is slightly shifted to orange hues if illuminant D65 and a viewing angle of 10° are used.

The mean values and standard errors of the color coordinates for the virgin and extra virgin olive oil samples were as follows: $a^* = -10.5 \pm 1.5$, $b^* = 104 \pm 15$, $C = 104 \pm 15$, and $L^* = 89.8 \pm 2.6$ with $C\ 2^\circ$ and $a^* = 0.5 \pm 2.5$, $b^* = 103 \pm 16$, $C = 103 \pm 16$, and $L^* = 88.6 \pm 2.8$ with $D65\ 10^\circ$. These mean values provide an indication of the usual color coordinates for virgin and extra virgin olive oils complying with their specific regulation (9); note, however, that the gamut of potential colors includes every point in their color space. Olive oils provided the following mean values and standard errors: $a^* = -10.7 \pm 2.2$, $b^* = 65 \pm 32$, $C = 66 \pm 32$, and $L^* = 94.1 \pm 3.5$ with $C\ 2^\circ$ and $a^* = -3.0 \pm 1.6$, $b^* = 68 \pm 29$, $C = 68 \pm 30$, and $L^* = 92.3 \pm 3.9$ with $D65\ 10^\circ$. Note that coordinate b^* was much smaller and so was C as a result of the much lower pigment content leading to an increased

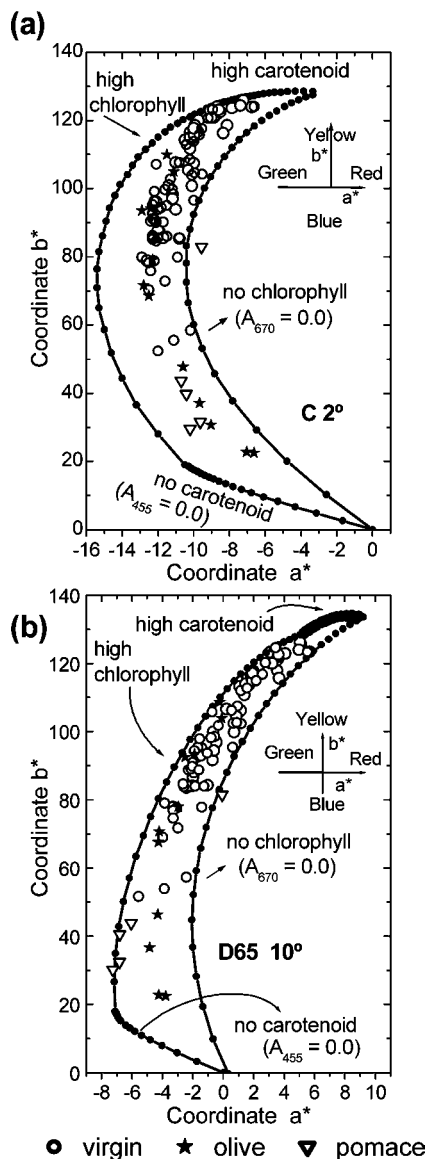


Figure 5. Boundaries of the b^* vs a^* color space and the coordinates for the virgin, extra virgin, olive, and pomace oil samples as obtained with the illuminants C 2° and D65 10°.

lightness. The standard errors obtained were very large as a result of olive oils spanning a wide range of virgin oil contents used in their production. The pomace oil samples provided the following mean values and standard errors: $a^* = -10.1 \pm 0.5$, $b^* = 45 \pm 22$, $C = 47 \pm 21$, and $L^* = 95.5 \pm 2.6$ with C 2° and $a^* = -5.4 \pm 3.0$, $b^* = 46 \pm 21$, $C = 46 \pm 20$, and $L^* = 94.4 \pm 3.0$ with D65 10°. Consistent with their production method, these oils exhibited decreased b^* and C values and increased L^* values, relative to the olive oils.

Figure 5a,b warrants several interesting conclusions, namely: (i) Although the virgin and extra virgin olive oil samples studied fell preferentially in a specific zone of the color space, some samples can have pigment contents resulting in their coordinates falling in another zone.

(ii) The three samples that were seemingly virgin olive oil but were rejected in the spectral reconstruction process had color coordinates falling within the color space as their color was acceptable for virgin olive oil. However, their ordinates were 52.4, 55.5, and 58.4, respectively, for b^* (C 2°) and 51.6, 53.9, and 57.2, respectively, for b^* (D65 10°). On the basis of their chromaticity diagram, locations, and the mean values for the

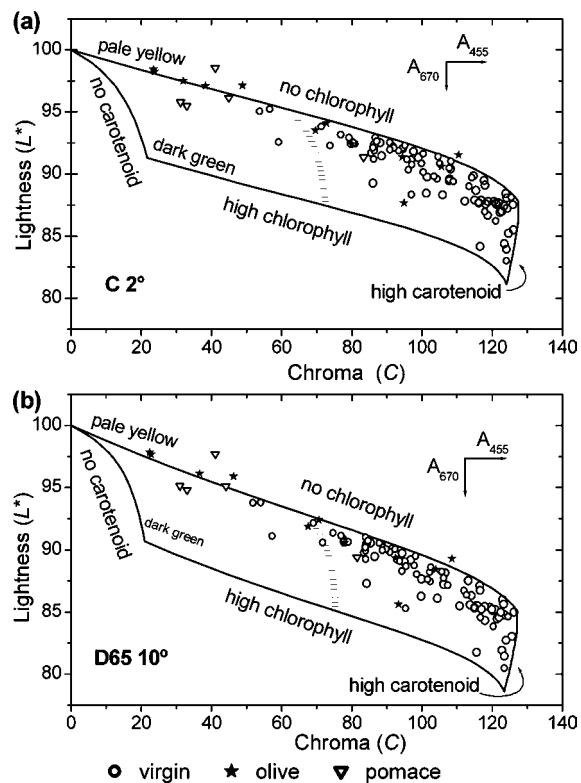


Figure 6. Boundaries of the L^* vs C color space and the coordinates for the virgin, extra virgin, olive, and pomace oil samples as obtained with the illuminants C 2° and D65 10°.

other samples, these might have lost some pigments. Such a relationship between pigment losses and the decrease in b^* was previously observed by other authors (3). The three samples obviously exhibited other signs of degradation [e.g., a K_{270} value exceeding the regulated threshold (9) for virgin olive oil].

(iii) The pomace oils and some olive oils also had low b^* values by effect of their pigment contents, which were high enough in some olive oils to give coordinates falling within the zone for most virgin or extra virgin oils. One of the pomace oil samples had a relatively high b^* value (near 82 with both illuminants) and fell to the right of the no chlorophyll line. This was a result of the blue region in the visible spectrum for pomace oil exhibiting a band peaking in the UV region and this absorption substantially strengthening that of the small amount of carotenoid pigments from virgin olive oil that is added to commercial pomace oil.

(iv) Because the color space in the C 2° diagram was larger than that in the D65 10° diagram, the samples exhibited more zones containing no experimental points. Most samples fell at the top of the enclosure and some overlapped with the high chlorophyll or high carotenoid lines. This had a two-fold origin. One was that many virgin olive oils were more green than yellow; the other was that the color enclosure was defined by using absorbance limits for A_{670} and, especially, A_{455} that exceeded the indications of use for many current instruments, so the loss of linearity in the response of some detectors to very strong bands altered the color coordinates that we would have obtained with thinner cells. However, thicker cells provide a more realistic perception of sample color.

Figure 6a,b shows the color enclosure for the olive oils as plots of lightness (L^*) against chroma (C), as well as the values of both coordinates for the body of samples. The two figures are very similar; if only, the range spanned by L^* was slightly narrower with illuminant C 2° and the decrease in lightness with

Table 1. Equations of the Lines Defining the b^* vs a^* Color Space for Virgin Olive Oil as Obtained with Illuminant C and the Standard Observer (See **Figures 3a** and **5a**)

boundary line and range spanned	C 2° (b^* , a^*)	R^2	SD
no carotenoid $-10.50 \leq a^* \leq 0.00$	$b^*_{\text{sample}} \geq -0.07198 - 1.78636 \times a^* - 0.08648(a^*)^2 - 0.00845(a^*)^3$	0.9997	0.106
high carotenoid $-8.64 \leq a^* \leq -3.33$	$b^*_{\text{sample}} \leq 124.27976 - 1.72463 \times a^* - 0.23964(a^*)^2 - 0.00395(a^*)^3$	0.9995	0.032
high chlorophyll $18.98 \leq b^* \leq 123.8$	$a^*_{\text{sample}} \geq -3.53108 - 0.50532 \times b^* + 0.00918(b^*)^2 - 8.55255 \times 10^{-5}(b^*)^3 + 3.34573 \times 10^{-7}(b^*)^4$	0.9939	0.182
no chlorophyll $0.00 \leq b^* \leq 127.5$	$a^*_{\text{sample}} \leq 0.16883 - 0.32179 \times b^* + 0.00392(b^*)^2 - 3.08834 \times 10^{-5}(b^*)^3 + 1.41716 \times 10^{-7}(b^*)^4$	0.9972	0.157

Table 2. Equations of the Lines Defining the L^* vs C Color Space for Virgin Olive Oil as Obtained with Illuminant C and the Standard Observer (See **Figures 4a** and **6a**)

boundary line and range spanned	C 2° (L^* , C)	R^2	SD
no carotenoid $91.32 \leq L^* \leq 100.00$	$C_{\text{sample}} \geq 24672.26321 - 794.44131 \times L^* + 8.54646(L^*)^2 - 0.03069(L^*)^3$	0.9998	0.104
high carotenoid $81.13 \leq L^* \leq 87.78$	$C_{\text{sample}} \leq -142.82381 + 5.77018 \times L^* - 0.03057(L^*)^2$	0.9993	0.031
high chlorophyll $21.7 \leq C \leq 124.1$	$L^*_{\text{sample}} \geq 90.45497 + 0.13531 \times C - 0.00597 \times C^2 + 6.80984 \times 10^{-5} \times C^3 - 2.69471 \times 10^{-7} \times C^4$	0.9981	0.135
no chlorophyll $0.00 \leq C \leq 127.6$	$L^*_{\text{sample}} \leq 99.91014 - 0.05534 \times C - 0.00117 \times C^2 + 1.89831 \times 10^{-5} \times C^3 - 9.5387 \times 10^{-8} \times C^4$	0.9993	0.099

increase in pigment content was somewhat more marked with illuminant D65. Near the center of the enclosures in both graphs is a dashed line bounding the zone where most virgin and extra virgin olive oil samples fell. The three rejected, low-quality samples of virgin olive oil fell to the left as a result of their markedly decreased C values and slightly increased lightness. Their coordinate values were as follows: 53.75, 56.66, and 59.20 for C and 96.06, 96.23, and 92.60 for L^* with C 2° and 51.89, 54.05, and 57.28 for C and 94.78, 94.81, and 91.12 for L^* with D65 10°. As can be seen, one sample of pomace oil and several of olive oil fell above the no chlorophyll line. Translucency and lightness in nonvirgin olive oil samples increase with a decreasing amount of virgin olive oil added. Also, as noted in discussing **Figure 5a,b** above, their color cannot be justified in terms of pigment content alone. Thus, the color space for virgin olive oil also contains nonvirgin oil samples with a high pigment content. As can also be seen in **Figure 6a,b**, some virgin olive oil samples fell very close to the no chlorophyll line, partly as a result of the influence of baseline fitting on L^* . In addition, commercial samples usually exhibit slightly increased lightness some time after processing, freshly processed samples echoing the values for the virtual samples used to define the boundaries of the color space.

On the basis of the foregoing, the color coordinates for virgin or extra virgin olive oil must fall within the theoretical color space provided this is established from spectra for pure samples recorded with 1 cm thick cells. All acceptable colors for virgin olive oil fall within this color space. In any case, most samples will have color coordinates differing little from the mean values for the 87 samples studied in this work and probably falling within the same ranges. Such ranges were as follows: $-12.90 \leq a^* \leq -6.60$, $70.22 \leq b^* \leq 125.90$, $71.33 \leq C \leq 126.10$, and $83.03 \leq L^* \leq 94.07$ with C 2° and $-3.96 \leq a^* \leq 5.68$, $68.93 \leq b^* \leq 126.20$, $69.04 \leq C \leq 126.30$, and $80.47 \leq L^* \leq 92.16$ with D65 10°. Therefore, b^* and C usually exceed 70 and L^* 80, with both C 2° and D65 10° as illuminants. On the other hand, the range spanned by coordinate a^* was more strongly influenced by the particular illuminant. The pigment spectra used to establish the color space were obtained by averaging the results for a variety of oil samples, so the

likelihood of a virgin or extra virgin olive oil with color coordinates falling outside the color space is very low. Even if that were not the case, one should check for quality loss in the sample concerned (9); in fact, commercial oil usually loses some quality upon exposure to light, whether natural or artificial (46–48), or as a result of its best before date being exceeded. A degraded or adulterated sample can exhibit a carotenoid profile departing from that of **Figure 1a**; hence, b^* and C values are outside the color space of **Figures 5** and **6**.

To facilitate the use of the color space, each boundary line was approximated to a polynomial function by using least-squares regression. The equations thus obtained are shown in **Tables 1–4**. The first column in each lists the name of each boundary line as shown in **Figures 3–6** and the range spanned by the independent variable (viz. a^* , b^* , L^* , or C). The second column lists the polynomial equations for the boundary lines, the subscript sample denoting that the value of the color coordinate concerned in a test sample should be greater than or equal to (\geq) or less than or equal to (\leq) the value obtained by applying the equation following the symbol and using the experimental value for the test sample as that for the independent variable. One can expect any virgin or extra virgin olive oil to obey the inequalities listed in the tables and hence its color coordinates to fall within the color space. The right-most columns in each table list the statistics for the least-squares fitting (viz. the coefficients of determination and the standard deviation of the fit, which can be used to include a given sample in the color space). The order of each polynomial was chosen by favoring small standard errors in their coefficients over those in R^2 unless the difference between successive coefficients of determination obtained with polynomials of a different order altered the second decimal place.

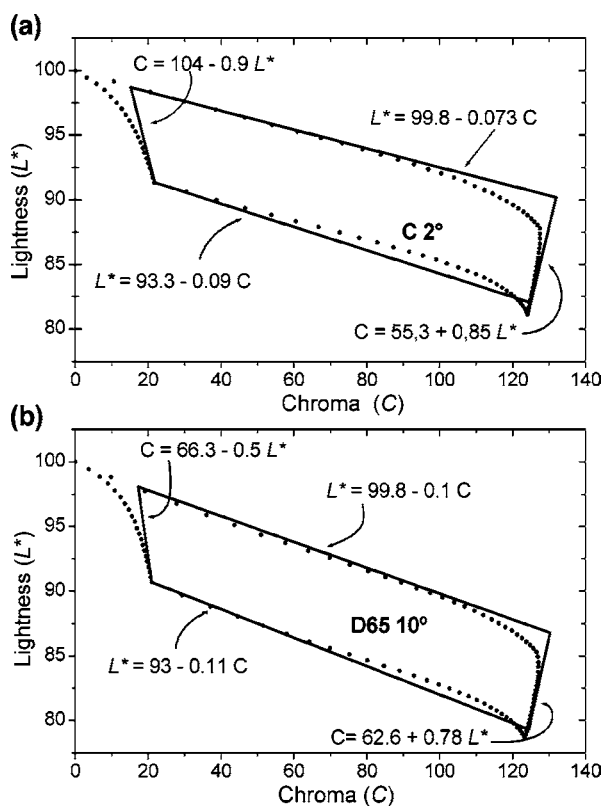
The color spaces of **Figure 4a,b** (or **Figure 6a,b**) can be compared with a quadrilateral (see **Figure 7**, which also shows the equations for the lines forming the quadrilateral). Such simple equations are highly acceptable replacements for those in **Tables 2** and **4** as the space that they enclose is very similar to the L^* – C color space. On the other hand, the b^* – a^* color spaces of **Figure 3a,b** (and those of **Figure 5a,b**) cannot be reduced to a simple geometric figure, so the polynomial

Table 3. Equations of the Lines Defining the b^* vs a^* Color Space for Virgin Olive Oil as Obtained with Illuminant D65 and the Supplementary Observer (See Figures 3b and 5b)

boundary line and range spanned	D65 10° (b^* , a^*)	R^2	SD
no carotenoid $-7.02 \leq a^* \leq 0.00$	$b^*_{\text{sample}} \geq 0.03825 - 0.87736 \times a^* + 0.85351(a^*)^2 + 0.21763(a^*)^3 + 0.01812(a^*)^4$	0.9983	0.266
high carotenoid $4.809 \leq a^* \leq 8.918$	$b^*_{\text{sample}} \leq 96.73604 + 8.63649 \times a^* - 0.46997(a^*)^2 - 0.0036(a^*)^3$	0.9999	0.027
high chlorophyll $18.99 \leq b^* \leq 126.6$	$a^*_{\text{sample}} \geq -4.28429 - 0.22917 \times b^* + 0.00617(b^*)^2 - 5.9196 \times 10^{-5}(b^*)^3 + 2.2898 \times 10^{-7}(b^*)^4$	0.9991	0.118
no chlorophyll $0.00 \leq b^* \leq 133.8$	$a^*_{\text{sample}} \leq 0.13 - 0.13949 \times b^* + 0.00261(b^*)^2 - 1.96031 \times 10^{-5}(b^*)^3 + 8.54306 \times 10^{-8}(b^*)^4$	0.9994	0.095

Table 4. Equations of the Lines Defining the L vs C^* Color Space for Virgin Olive Oil as Obtained with Illuminant D65 and the Supplementary Observer (See Figures 4b and 6b)

boundary line and range spanned	D65 10° (L^* , C)	R^2	SD
no carotenoid $90.69 \leq L^* \leq 100.0$	$C_{\text{sample}} \geq 26854.44981 - 866.76773 L^* + 9.34089 L^{*2} - 0.03359 L^{*3}$	0.9994	0.161
high carotenoid $78.59 \leq L^* \leq 85.3$	$C_{\text{sample}} \leq -189.16405 + 7.00644 L^* - 0.03854 L^{*2}$	0.9994	0.031
high chlorophyll $20.98 \leq C \leq 123.50$	$L^*_{\text{sample}} \geq 95.25116 - 0.25659 C + 0.00261 C^2 - 1.29143 \times 10^{-5} C^3$	0.9973	0.188
no chlorophyll $0.00 \leq C \leq 126.7$	$L^*_{\text{sample}} \leq 100.1781 - 0.15399 C + 0.00111 C^2 - 6.31506 \times 10^{-6} C^3$	0.9990	0.141

**Figure 7.** Boundaries of the L^* vs C color space for virgin and extra virgin olive oil. Initial color space, ---; assimilated color space and its equations, —.

equations of **Tables 1** and **3** are most probably the simplest possible functions for representing the boundaries of the color space of virgin olive oil in the CIE 1976 color diagram.

Establishing the color space occupied by every possible variety of virgin and extra virgin olive oil allows one to define the entire acceptable color range that they can span. Also, the equations for the lines bounding the color space allow one to check whether a sample falls within the space. Therefore, this

color space could be included among the characteristics of virgin olive oil established by the IOOC and regulated in the EC Commission Regulation (9). In fact, if the color coordinates for a given oil sample fall outside the color space—with provision for the standard errors in **Tables 1–4**—the sample should be rejected. Similar color spaces can be established for other foods fit for human consumption by using a procedure similar to that employed for virgin olive oil in this work.

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