Precision and Accuracy in the Color Specification of Virgin Olive Oils from the Bromthymol Blue Method

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ABSTRACT: Twenty experienced observers with nondefective color vision judged 27 virgin olive oil samples within an acceptable color range, using the bromthymol blue (BTB) method, under controlled observation conditions (daylight source with a correlated color temperature of 6500 K, and standard gray background). On the average, 44.8% of the observers agreed in their selections of the BTB standard solution matching a given oil sample, and this percentage increased to 88.2% considering ± one step in the two dimensions (pH and concentration) of the BTB scale. On the average, the lowest color difference between oil samples and available BTB solutions was 6.6 Commission Internationale de l'Éclairage 1976-(L*a*b*) (CIELAB) units, but this color difference was approximately two times greater for the color difference between oil samples and BTB solutions selected by our observers. The colors of the BTB standard solutions in the CIELAB space are not uniformly distributed, and thus one step in pH or concentration is equivalent to CIELAB color differences varying in a wide range (1.7–13.5 and 1.7–26.3 CIELAB units, respectively). From these values, indicating low precision, accuracy, and uniformity, some suggestions are made for future improvements of the current BTB method.

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KEY WORDS: Bromthymol blue method, CIELAB, color differences, oil color, virgin olive oils, visual judgments.

The bromthymol blue method (BTB method) (1) is a procedure for specifying the color of olive and seed oils on the basis of a visual comparison between the oil sample and a given set of standard solutions, arranged as a bidimensional color scale of $6 \times 10 = 60$ samples. The standard solution most closely matching the color of the oil sample indicates its BTB color index which is given as two integer numbers, the first designated as pH and the second as concentration; these are in the ranges, respectively of $2 \le pH \le 7$ and $1 \le$ concentration ≤ 10. Currently, the BTB method is the official method adopted in Spain by researchers and the oil industry (2).

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We have addressed some drawbacks of the BTB method (3), suggesting the use of Commission Internationale de l'Éclairage 1976-(L*a*b*) (CIELAB) color space (4) as a more appropriate way for color specification and proposing numerical relationships between the BTB index and the CIELAB coordinates. As pointed out by Judd and Wyszecki (5), in using color scales, it is rare that a perfect color match can be achieved by observers, who are faced with a difficult and sometimes impossible task. In addition, it should be borne in mind that the Commission Internationale de l'Éclairage (CIE) achieved an important goal in 1976 when two approximately uniform color spaces [designated as Commission Internationale de l'Éclairage 1976-(L*u*v*) (CIELUV), and CIELAB] were proposed, on the basis of numerous theoretical and practical considerations, in an attempt to promote uniformity of practice between users (6). Since this time, the use of CIELAB has been widely accepted (7), mainly for industrial applications involving object colors. In summary, it seems advisable to use the CIELAB system in place of any other color specification, such as the one provided by the BTB method.

In any case, the use of a color scale, such as the one proposed by the BTB method, is a valid and simple tool for color specification, in practical applications, as with other well-known color scales [e.g., the Munsell Soil Color Charts (8)]. As stated also by Judd and Wyszecki (5), a good color scale is a useful timesaver in spite of its drawbacks, as long as it is not used to provide a one-dimensional solution to what is essentially a multidimensional problem.

Thus, a reliable study on the main characteristics of the color scale provided by the BTB method (1) would be desirable. In the present article, we report quantitative data concerning the precision and accuracy achievable by the BTB method. Precision and accuracy are two independent concepts related to the error associated with any physical measurement. Fairchild and Reniff (9) have appropriately illustrated these concepts using the simple scheme shown in Figure 1. When accuracy is high, the average measurement falls on the bull's eye (true value), but when accuracy is low, the average measurement misses the bull's eye. When precision is high, repeated measurements fall very near each other, and when precision is low, repeated measurements do not fall near each

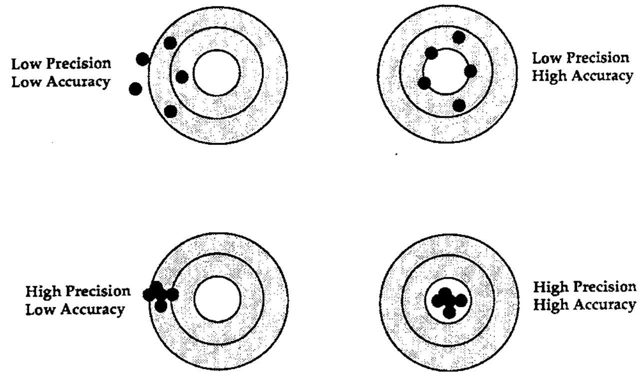

FIG. 1. Basic scheme from Fairchild and Reniff (9), illustrating the concepts of precision and accuracy in physical measurements. The bull's eye center corresponds to the true value.

other. In practice, accuracy and precision should be associated with systematic and random errors, respectively. In particular, the accuracy and precision achieved using the BTB method indicate the reliability of this method, and serve as a reference for future improved methods.

The uniformity of the scales provided by the BTB method is also analyzed in the present article. It would be desirable that one step in any of the two scales (pH and concentration) of the BTB solutions would correspond consistently to the same color difference. The CIELAB space, assumed by the CIE to be an approximately uniform color space, has been used to compute these color differences. Recently the CIE has proposed (10) a CIELAB-based color-difference formula, designated as CIE94, in an effort to improve the correlation between perceived and measured color differences in industrial applications. Unfortunately, CIE94 will not be used here because the color differences found in our current work are far greater than 5.0 CIELAB units (the upper limit recommended for the use of CIE94).

From the results achieved here with respect to precision, accuracy, and uniformity of the BTB method, complemented by those found in our previous study (3), we feel that some guidelines toward a future improved method for the color measurement of oil samples should be established.

EXPERIMENTAL PROCEDURES

Following the standard procedure (1), we prepared a set of BTB solutions. Each of the 60 BTB solutions consisted of a 45-mL solution in a cylindrical bottle (4.0 cm diameter × 7.6 cm height, approximately). From diverse olive varieties collected at the most representative production zones in Andalucía, Spain, 134 virgin olive oil samples were selected and placed in the same type of cylindrical bottles as the BTB solutions. All these oil samples were extracted in the laboratory by the Abencor® method (11), thus reproducing the industrial procedure. To cover the color range represented by the 134 natural virgin olive oil samples, we selected 27 samples for the visual experiment performed here.

Visual comparisons were made by a panel of 20 observers, who were asked which of the 60 BTB standard solutions most closely matched the color of each one of the 27 oil samples. Our observers were 12 men and 8 women, with an average age of 36, who were tested for normal color vision before the experiment, using the Ishihara test (12). From the 20 observers, 14 had previously performed color-matching experiments like the one proposed here, and the remaining 6 were familiar with oil-sample management through their work in oil laboratories. Thus, our visual experiment was performed by experienced observers.

The visual comparisons were performed using a Verivide color assessment cabinet CAC 120 (Leslie Hubble Limited, UK), having a daylight source with a correlated color temperature of about 6500 K (D65) within the tolerances prescribed by British Standard 950. On the right side of the Verivide cabinet the 60 BTB solutions were presented for selection to the observer, and, on the left side, the observer juxtaposed each oil sample with the selected BTB solution, in order to check the color match. Because the two solutions compared were transparent, we placed a gray mask (from the Munsell Book of Color) behind them, acting as a fixed standard background in our experiment.

Instrumental color measurements were performed using a SpectraScan PR-704 spectroradiometer (Photo Research Inc., Chatsworth, CA) equipped with appropriate software (13). This instrument was mounted on a standard tripod, using a circular aperture field of 1° and one cycle integration time. For color measurements of BTB solutions and of oil samples, the optical head of the spectroradiometer was focused on the center of the nearest surface of the bottles, which were placed inside the Verivide cabinet in front of the Munsell gray mask. In this way, the instrumental color measurements as well as the visual comparisons were made under the same experimental conditions. The CIE 1964 Standard Observer (4) was assumed for our color measurements, because the angle subtended by the juxtaposed samples in the visual comparisons was greater than 4°. A pressed bariumsulfate-powder plaque, provided with the spectroradiometer and measured under the same experimental conditions as the samples, served as the reference white necessary to calculate color coordinates in the CIELAB space. The CIELAB coordinates of our gray background were $a^* = 1.1$; $b^* = 3.4$; $L^* = 93.9$.

Figure 2 shows the CIELAB coordinates that were measured for the 60 BTB solutions, the 134 oil samples, and the 27 oil samples finally selected for our visual experiment. Although the 60 BTB solutions were prepared about 4 mon before the visual experiment, their colors together with those of the oil samples were measured just before the experiment. The BTB samples were spread out in the CIELAB space (Fig. 2), following a different pattern from the usual triangular shape shown before (see, for example, Fig. 2, Ref. 3). This result is not surprising, because we wished to perform the current measurements under the same experimental conditions as those used for the visual experiment, where the pathlength of the samples was approximately 4.0 cm. Previous measure-

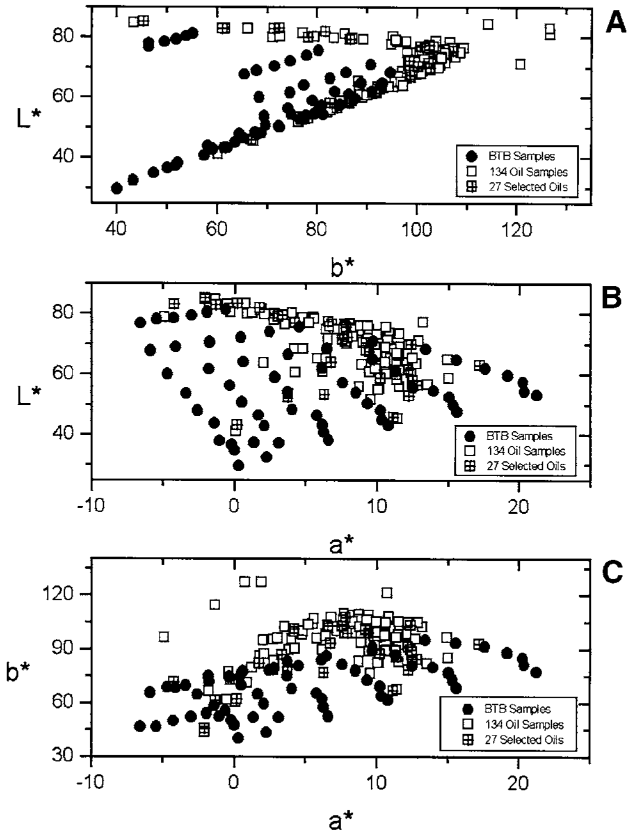

FIG. 2. Color coordinates measured for the 60 bromthymol blue (BTB) solutions, 134 virgin olive oil samples, and 27 oil samples selected for our current visual experiment, referred to the planes b*L* (A), a*L* (B), and a*b* (C), of the Commission Internationale de l'Éclairage (CIELAB) color space.

ments were performed by transmission, using a spectrophotometer and quartz cells of a lower pathlength (0.5 cm in Ref. 3). It would be futile to perform color measurements using a conventional spectrophotometer with cells of a given pathlength, and then to use a different pathlength for the visual comparisons prescribed by the BTB method. Although it should be assumed from Grassman's laws that a color match for a given pathlength stands up for other pathlengths, in a rigorous procedure of visual comparison, the BTB and oil samples should be placed in bottles with the same pathlengths, as indicated (1,2).

In Figure 2, the BTB solutions are not evenly spread in the CIELAB space and thus do not constitute a uniform color scale. In addition, the coordinates of most of the 134 virgin olive oil samples appear to be far away from the color range covered by the BTB samples, as also happened in our previous study using 502 virgin olive oils (3). These two observations are undesirable characteristics of the current BTB method (1) and will be more thoroughly analyzed below.

RESULTS AND DISCUSSION

In the next two subsections, the precision and accuracy of the visual experiment described above will be analyzed. The third subsection focuses on the uniformity of the BTB scales, which is also related to the precision and accuracy found. Finally, in the fourth subsection, some conclusions and guidelines toward an improved BTB method have been provided.

Precision. Unfortunately, as might be expected, for a given oil sample different BTB solutions were selected by different observers who were trying to achieve the best visual color match. In other words, there was a lack of precision using the BTB method even for experienced observers working under well-controlled experimental conditions.

Table 1 illustrates the precision achieved with the BTB method in our experiment. For a given oil sample, the BTB solution most frequently selected by the 20 observers has been designated as "main" in Table 1 (in the case of three oil samples, where two different BTB solutions were selected by the same number of observers, we arbitrarily selected one of them as "main"). The numbers in Table 1 indicate the percentage of observers selecting the main and neighboring BTB solutions (average ± standard deviation for the 27 oil samples assessed). Roughly half (44.8% ± 14.0%) of our observers agreed on the BTB solution nearest to a given oil. This percentage increased to 88.2% of the observers considering ±1 step in pH and concentration around the main sample. Thus, this margin represents the precision of our experiment at approximately a 90% confidence level.

Table 1 reveals a similar spread of the visual assessments through the pH and concentration scales (i.e., as desired, the

TABLE 1
Results Corresponding to Precision Achieved in Reported Experiment in Terms of pH and Concentration Scales[a]

	pH						
Concentration	−3	−2	−1	Main	+1	+2	+3
+3				0.2 ± 1.0	0.7 ± 2.2		
+2			0.6 ± 2.1	3.3 ± 5.9			
+1			0.6 ± 2.8	10.0 ± 11.5	3.5 ± 7.2	0.6 ± 2.1	
Main		1.5 ± 6.8	5.2 ± 10.5	**44.8 – 14.0**	7.8 ± 12.0	0.4 ± 1.3	
−1		0.2 ± 1.3	5.0 ± 7.5	10.4 ± 9.2	0.9 ± 2.4		
−2			1.1 ± 2.8	2.0 ± 4.4	0.6 ± 2.1		
−3			0.2 ± 0.9	0.2 ± 1.0	0.2 ± 0.9		

[a]The bromthymol blue (BTB) solution most frequently selected by the group of observers is designated as "main." The numbers indicate the percentage of observers selecting each BTB solution (average ± standard deviation for the 27 oil samples assessed). Boldface: central value.

units of both scales are comparable, from a perceptual standpoint). However, strictly speaking, with respect to the "main" solution, 59.7% of the assessments were spread through the pH scale and 70.9% spread through the concentration one. Perhaps this slightly greater value for the concentration scale should be attributed to the larger number of samples available on this scale (10 samples for each pH, against 6 samples for each concentration). In addition, we also note from Table 1 that solutions with simultaneous variations of pH and concentration, with respect to the most frequently selected one (main), were rarely selected by the observers (14.2% of the assessments).

Accuracy. As might be expected, there is usually a nonzero color difference between a given oil sample and its nearest BTB standard solution. This color difference should be interpreted as the best accuracy achievable from the BTB method. However, as discussed above, the nearest BTB sample to a given oil sample is not always unanimously selected by the observers, thus leading to even poorer accuracy.

Table 2 shows the accuracy achieved for each one of the 27 oil samples used in our experiment. The BTB solution nearest to each oil sample (based on instrumental determinations) and its corresponding color difference (in CIELAB units) are given in columns 2 and 3, respectively. Column 4 shows the BTB solution most frequently selected by our observers' group (i.e., the sample designated as "main" in Table 1), and column 5 shows the color difference between each oil sample and the BTB solutions selected by the 20 observers (average ± standard deviation).

From Table 2, by using the average of our 27 oil samples, the best accuracy achievable by the BTB method was 6.58 CIELAB units, whereas the accuracy achieved by our experienced observers was 2.32 times greater (12.27 CIELAB units). The standard deviation of the color differences between oil samples and BTB solutions selected by the observers (3.67 CIELAB units) should also be considered another measurement of the precision achieved in our experiment. This measurement of the precision as a standard deviation is compatible with the one previously indicated (±1 step in pH and concentration), which is roughly equivalent to 7–8 CIELAB units, and includes approximately 90% of the visual judgments made by the group of observers.

Figure 3 shows a relatively good correlation (r = 0.837) between the accuracy achieved by the observers and the best

TABLE 2
Results Corresponding to the Best Accuracy Obtainable in our Experiments, and Accuracy Achieved in Practice by 20 Experienced Observers

Oil	Nearest instrumental sample		Nearest visual selected samples (average ± SD)	
sample	BTB index	ΔE^*_{ab} [a]	BTB index[b]	ΔE^*_{ab} [c]
1	2-6	1.83	2-7	7.20 ± 2.64
2	2-3	13.77	2-3	16.52 ± 2.89
3	3-7	2.76	3-9	8.16 ± 3.26
4	3-6	3.92	3-8	10.65 ± 5.21
5	4-8	3.26	4-10	5.65 ± 0.82
6	2-4	15.65	3-4	20.63 ± 2.02
7	2-4	7.28	3-5	17.54 ± 5.00
8	3-5	3.20	3-8	15.16 ± 5.39
9	2-2	7.44	2-2	9.76 ± 2.24
10	6-7	3.12	5-9	8.85 ± 4.18
11	3-4	4.67	3-7	13.55 ± 3.68
12	2-3	12.92	3-3	18.67 ± 2.24
13	4-2	12.22	4-2	13.48 ± 2.22
14	2-4	5.66	3-5	15.58 ± 5.19
15	2-2	5.11	2-2	9.38 ± 4.58
16	2-4	10.21	3-4	19.98 ± 6.30
17	3-6	3.39	3-8	9.34 ± 4.75
18	4-6	2.58	4-8	9.18 ± 3.89
19	5-5	2.74	5-7	10.50 ± 3.67
20	6-1	7.94	4-1	9.74 ± 0.86
21	2-4	8.30	2-4	12.21 ± 6.18
22	3-2	11.03	3-2	12.59 ± 2.77
23	2-4	7.82	3-4	12.65 ± 2.67
24	2-2	6.94	3-2	8.20 ± 1.80
25	3-4	5.21	4-6	17.08 ± 5.28
26	3-5	2.61	3-7	10.67 ± 3.10
27	2-1	6.16	2-1	8.49 ± 6.25
Average	—	6.58	—	12.27 ± 3.67

[a]Color differences (oil sample – nearest BTB sample) in CIELAB units.
[b]BTB sample most frequently selected by the observers' group (designed as "main" in Table 1).
[c]Color differences (oil sample – BTB samples selected by the observers) in CIELAB units.

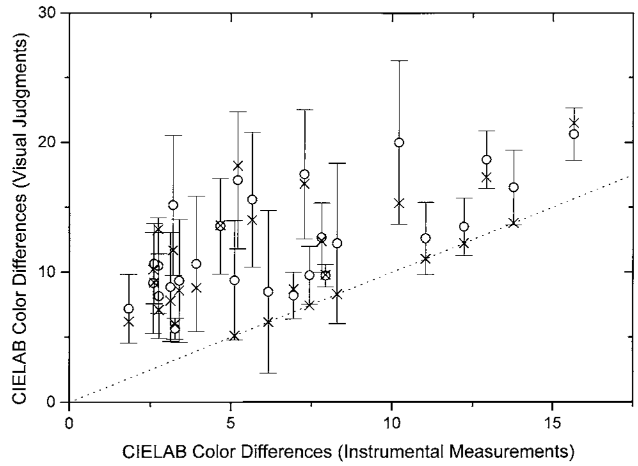

FIG. 3. Relationship between the (CIELAB) color differences [(oil sample – nearest neighbor BTB solution) (x axis) and (oil sample – BTB solutions selected by our observers' group (y axis)]. The error bars indicate the standard deviation corresponding to the different BTB solutions selected by the observers. The crosses (×) indicate the color difference between each oil sample and the BTB solution most frequently selected by the group of observers. For abbreviations see Figure 2.

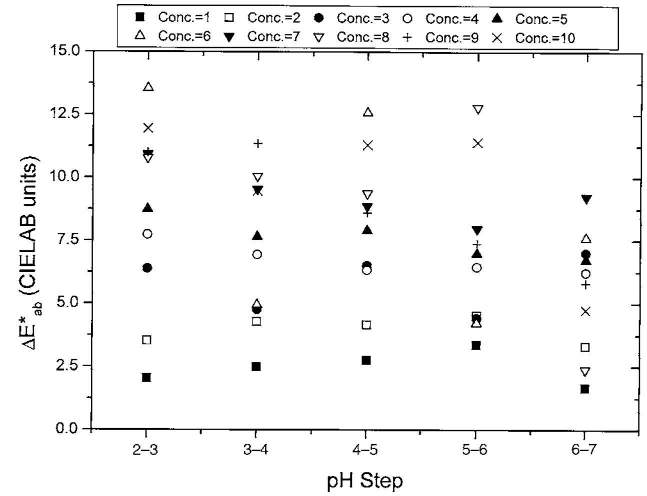

FIG. 4. Color differences in CIELAB units corresponding to one step in pH. For abbreviation see Figure 2. ΔE^*_{ab} is the usual symbol for color differences in CIELAB units.

accuracy obtainable. However, only 7 of the 27 BTB solutions most frequently selected by the group of observers (25.9%) corresponded to the nearest-neighbor BTB solution to the oil sample, in CIELAB space. Thus, visually perceived color differences and color differences measured in CIELAB are not as well correlated as might be desired. This result is not surprising, and, in the last few years, different color-difference formulas have been proposed to improve the performance of CIELAB (10,16).

Uniformity. We computed the CIELAB color differences corresponding to one step in both pH and concentration, and the results are shown in Figures 4 and 5, respectively. On average, one step in pH and concentration corresponds to rather similar values, although with very different standard deviations: 7.21 and 8.11 CIELAB units (with standard deviations of 3.13 and 6.14 CIELAB units), respectively. Thus, one step in both pH and concentration is equivalent to color differences ranging from 1.67 to 13.53 and 1.74 to 26.25 CIELAB units, respectively. From Figure 4 we see that one step in pH usually has a greater size in CIELAB units, when the concentration increases. Also in Figure 5, great color-difference values correspond to step 1–2 in concentration. That is, Figures 4 and 5 (as well as Fig. 2) illustrate that both scales of the BTB method (pH and concentration) have poor uniformity. This result is certainly not surprising, because the different BTB samples were easily prepared by changing the proportion of different solutions, and this does not necessarily imply a uniform sampling in CIELAB.

With regard to obtaining uniform color scales, the work carried out by the Optical Society of America Committee on Uniform Color Scales is relevant and has been described (14). It has led to an array where each color was surrounded by 12 nearest neighbors, all differing from the central color by the same perceptual amount. Among other advantages, this arrangement (designed as a regular rhombohedral or cuboctahedron lattice) is the basis on which to assemble a collection of color chips that will have the greatest possibility of including a near match for any color chosen at random (15). This property should be of particular interest for a visual comparison procedure, as the one proposed by the BTB method.

In using the 60 BTB samples and a color tolerance of 2.0 CIELAB units, only two of our 134 oil samples (i.e., 1.5%) should have been matched. Previously, we found a slightly better result (3), showing that, with a tolerance limit of 1.5 CIELAB units, 13.1% of 502 oil samples could be matched. In summary, real oil samples can be matched only with difficulty by using the samples provided by the BTB method, as a consequence of both the lack of uniformity of the BTB color scale and the displacement in color space of its center of grav-

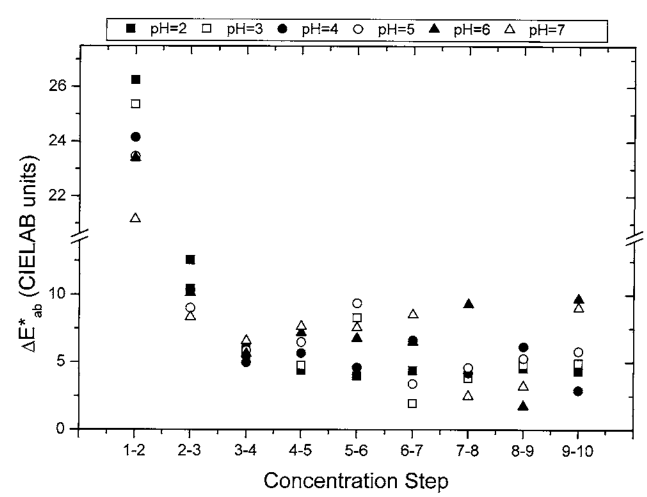

FIG. 5. Color differences in CIELAB units corresponding to one step in concentration. For abbreviation see Figure 2.

ity with respect to the coordinates of most of the virgin olive oils (Fig. 2). As an alternative, we tested the performance of a cuboctahedron lattice with 46 samples, having a color difference of 3.86 CIELAB units between nearest neighbors (which is the average value of the color difference between nearest neighbor BTB samples). The cuboctahedron lattice was developed starting from the center of gravity of our 134 oil samples in CIELAB; and we found that, with a tolerance limit of 2.0 CIELAB units, 48 of the 134 oil samples (35.8%) could be matched. Although this result should be even better bearing in mind the particular spread of our 134 samples (Fig. 2), we have considered that such an effort should be made on the basis of a larger and more complete set of oil samples. We feel that a more uniform and well-centered color scale replacing the current BTB scale should be considered.

Toward an improved color specification method. The previous results indicate a low precision and accuracy using the BTB method, even for experienced observers and under well-controlled observation conditions. Thus, the precision in our experiment was around ±1 step in pH and concentration, roughly equivalent to more than 7 CIELAB units, and the best accuracy was around this same value (or even greater if we consider the BTB solutions selected by a group of experienced observers). In this respect, it should be said that these color differences are very large, because a color difference of around 0.38 CIELAB units is just noticeable for normal observers, and a color difference around 1.75 CIELAB units is usually considered as a suprathreshold (17). Although the precision and accuracy reported here correspond to a given sample of 27 virgin olive oils, we feel that they can be assumed as representative of the ones achievable by the BTB method for virgin olive oils, but not in general for other oils. Thus, for example, oils with visually detectable reddish hues are explicitly excluded for the application of the BTB method (2), and use of these oils could lead to very poor results. A general procedure for the color specification of any oil would be desirable.

Another important shortcoming for the current BTB method is that, as also shown previously (3), the colors of the 60 BTB solutions are not uniformly spread over CIELAB color space. In addition, agreement between the color ranges covered by virgin olive oils and BTB solutions in CIELAB space is not good and, consequently, only a low percentage of oil samples can be matched using the current BTB scale.

We feel that in the near future, the use of the CIELAB system for color specification of oil samples will probably prove to be the most appropriate tool. Thus, visual comparisons such as the one proposed by the current BTB method (1) could be perhaps considered as an interim solution to the problem of color specification of oil samples. In our opinion, the subjectivity of the BTB method cannot be compared with the benefits of objective color measurements (with a remarkable precision and accuracy), which are easily made using current colorimetric instrumentation. With respect to the use of CIELAB space, we feel that presently this is the best option for oil color specification and, in particular, for some of the purposes of this article (e.g., study of the perceptual uniformity of the BTB scales). Specifically, we think that the use of CIELAB is based on well-established international recommendations (4), usual practice in most industries (7), potential software available in current instruments (13), and approximate uniformity. In this sense, it should also be recognized that while most people working with object colors use CIELAB, those working with light sources (e.g., self-luminous displays) often prefer CIELUV (which is also a valid option from current international recommendations). Mention should also be made of recent efforts toward new color spaces that are more uniform than CIELAB (18,19), which are currently being studied by the CIE Technical Committee 1-55.

In any case, to achieve a more rigorous color specification of oil samples from visual comparisons, we offer the following suggestions:

(i) The experimental observation conditions employed must be well defined. The current BTB method does not provide accurate information on the luminous source to be employed and the neutral background to be placed behind the samples. The previous use of a D65 luminous source and a Munsell gray mask should be considered in this respect.

(ii) A more uniform color scale than the one provided by the BTB method may be desirable. From a number and variety of oil samples greater than the 134 virgin olive oils considered in the current study, a cuboctahedron lattice, as proposed in the Uniform Color Scales of the Optical Society of America (14,15), should be designed, starting from the center of gravity of the oil samples. In this way, the low percentage of oil samples which can be matched using the current BTB solutions may be significantly improved.

(iii) As an alternative to the preparation of the solutions leading to the 60 BTB standards, a commercial light box with a D65 source (similar to those commonly used to analyze X-ray photographs) should be used, together with a set of slides with color samples designed as indicated in the previous paragraph. The pathlength of the bottles where the oil samples will be placed for visual comparisons with the slide samples should be previously fixed. The temporal degradation of color and the expenses of this procedure are probably not much greater than those from the current BTB method.

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