



ORIGINAL RESEARCH ARTICLE

In situ measurement of light transmission into wine bottles and calculation of shelf life

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ABSTRACT

Wine exposed to UV-A and violet light (320–440 nm) can develop a light-struck flavour which deteriorates its quality. In a short introduction, we briefly describe the two most important photochemical pathways of this process and discuss conditions that facilitate the formation of undesired compounds. The main thrust of the study is to use a novel method to determine light transmission into real wine bottles, along with optical and photochemical modelling of wine damage. Calculation of light transmission into bottles and resulting photochemical damage is described in detail and compared to in situ measurements of transmitted light inside bottles. The effect of different light sources (sunlight, fluorescent bulbs and LED bulbs) is also discussed. It is found in in situ measurements that clear (flint) bottles transmit up to 35 % and bluish-green bottles up to 8 % of the harmful UV–violet light, while light amber bottles only up to 1.2 % and dark amber ones up to 0.2 %. Results of simple tasting experiments support the validity of shelf life calculations described here. According to these findings, white wine in flint (clear) and bluish-green bottles can survive only for a few days while displayed on shelves; light amber bottles conserve the quality for about a month and dark amber bottles for more than a year. Recommendations concerning the protection of shelf-displayed bottled wine from light exposure are also given.

KEYWORDS: light-struck wine, in situ light measurement, light transmission calculation, shelf life, light protection, organoleptic analysis

INTRODUCTION

Wine is kept in containers not accessible for light during processing and ageing or—even during maturing in bottles—in dark cellars. However, a large share of bottles is sent to retail and placed on shelves in wine stores or supermarkets for display before consumers would buy them, exposed to ambient lighting, sometimes also during the night. Substantial irradiation of bottles with UV-A and the low, violet edge of the visible light spectrum (320 to 440 nm) can degrade the sensorial quality of wines, a phenomenon first reported in 1978 (Maujean *et al.*, 1978). The nature and extent of this damage is highly dependent on the sort of the wine itself and the material and shape of the bottle. (Carlin *et al.*, 2022) In this introduction, we briefly describe the most important photochemical pathways of the development of the so-called ‘light-struck flavour’ and summarise current knowledge of the photophysical properties of different wine bottles.

The main part of the paper reports a spectral study of the emission of practically relevant light sources, along with the light transmittance of some common glass materials used to manufacture wine bottles. To determine the transmitted flux, *in situ* measurement of light transmitted into different bottles was performed, and the results are compared to optical modelling of this transmission. Based on the measured transmission, the calculation of possible damage to wine quality contained in bottles on illuminated shelves is also given, taking into account relevant photochemical data. These results are also compared to sensorial tasting.

1. Photochemical processes degrading wine quality

Wine is a ‘living material’ continuously changing its quality due to biochemical and chemical processes, the latter also taking place in bottles. Most important from the point of quality is maturing of the bottled wine, but there are also chemical reactions that can degrade wine quality. Here we are interested in photochemical reactions that happen following the interaction of wine with light transmitted into the bottle. Recent review articles (Fracassetti *et al.*, 2021; Grant-Preece *et al.*, 2017) give a detailed overview of related wine photochemistry in bottles. Two gross processes are mentioned to be mostly responsible for wine photodegradation: oxidation based on singlet oxygen and related species with the help of iron(III) complexes; and photocatalytic decomposition of sulfur-containing compounds.

Though direct photoexcitation of triplet oxygen can generate singlet oxygen (Blázquez-Castro, 2017), the efficiency of this process is very low compared to other photochemical pathways. An effective photooxidative process is based on the reduction of oxygen catalysed by the iron(III)/iron(II) redox system, leading to the highly oxidative hydroxyl radical and subsequently also to the oxidation of ethanol to acetaldehyde (Grant-Preece *et al.*, 2017). Another efficient iron-catalysed process can turn diphenols into quinones, which can also lead to the browning of white wine.

Concerning the taste of wine, the most important photoproducts are organic sulfur compounds acting as low-threshold off-aromas, providing the typical light-struck flavour. Sulfur-containing amino acids methionine and cysteine are the target of riboflavin (vitamin B₂), and pantothenic acid (vitamin B₅) catalysed photodegradation. (Furet *et al.*, 2022) The mechanism of the riboflavin-sensitised reaction has been thoroughly studied. It has been found that riboflavin can be excited to its first singlet excited state, irradiating it with UV to visible light up to *cca.* 500 nm. The singlet state undergoes very fast vibrational relaxation and intersystem crossing (within 5 ns) to form a triplet manifold, which survives up to 15 μ s and acts as an efficient agent to degrade sulfur-containing amino acids. The main volatile off-flavour compounds to form are H₂S, dimethyl sulfide (DMS) and dimethyl disulfide (DMDS), as a sensory study (Dozon and Noble, 1989) also confirmed. As these two amino acids and the two vitamins are inevitably present in wine, the formation of these compounds always occurs when wine is exposed to harmful light for longer time, as detailed analytic studies also found (Marchand *et al.*, 2000; Pripis-Nicolau *et al.*, 2000). As a recent study confirmed, the degradation of varietal aroma compounds also occurs as a consequence of illumination. (Carlin *et al.*, 2022) Due to the absorption of light by anthocyanins and polymeric pigment in red wine and the subsequent dissipation of the energy of absorbed photons, this damage is more pronounced in white wine.

2. Photophysical properties of wine bottles

There are plenty of different bottles wine is stored in, varying from one wine region to the other and having different historical origins. Apart from bottle types restricted to smaller areas (e.g., Chianti, Tokaji or Port bottles), there are a few types widely used all over the world. Such are the three types we deal with in detail in this study: Bordeaux, Burgundy and Alsace bottles. Though they could be produced from glass material of different colours, the traditional colour is dark green. However, recent trends have led to clear bottles, especially in Bordeaux shape, to sell white and rosé wines to display their colour better. In addition, more sophisticated colours also emerged—among them the one we also study here: bluish-green bottles. Recent studies have shown that amber bottles have advantageous properties from the point of view of lowering the transmission of harmful light into bottles; thus, amber bottles are more frequent nowadays in the commercial packaging of wines (Hartley, 2008)

As it is already stated in the previous section, the harmful wavelength range for the quality of wine is between 320 and 440 nm. There are a few studies reporting on the transmission spectra of UV-A–visible light of different colour glasses used to produce wine bottles, measuring the transparency of glass sections cut from the bottles or a window on the bottles’ body cut in half. (Clark *et al.*, 2011; Dias *et al.*, 2010; Hartley, 2008). Thus, they focus on the transmission of the glass material of bottles and find that flint (clear) glasses transmit up to 90 %, green glasses close to 70 %, while amber glasses only a few percent of incident light in this critical wavelength range.

However, the authors of the present study were unable to find reports of in situ measurements within wine bottles. A US patent (Hidalgo and Gobbo, 1996; Hidalgo and Gobbo, 2000) describes transmission measurements inside a beer bottle but includes only the transmittance of a small window on the body of the bottle.

In the present study, transmission spectra of the glass materials were recorded first, and then we focused on measuring transmitted light within the bottles in situ, using different light sources typical in stores selling wine. We have also made calculations concerning the transmission of light into different bottles and photon fluxes within the harmful wavelength range. These calculations also enabled us to get a reliable estimate of white-wine shelf life.

MATERIALS AND METHODS

The main goal of this study at the experimental level was to determine light transmission inside different types of bottles manufactured from different glass materials. We have examined four bottles: flint (clear) Bordeaux, bluish-green slender-shape Alsace, light amber Burgundy and a somewhat bulky dark amber Alsace. All four bottles are in use by the winery Laposa Birtok Kft., Badacsonytomaj, Hungary, to bottle their various sorts of wine (Clear Bordeaux for rosé, the other three for white wines).

1. Transmission of wine bottle glass materials and that of white wine

To get the wavelength-dependent transmission of the glass material, we have prepared glass fragments of a few square centimetres from the middle part of the bottle body and

measured their transmittance using a spectrophotometer. We have used an AvaSpec-2048-USB2 versatile fibre-optic spectrometer, whose detector is connected to a linear CCD array via optical fibre, having a sensitivity range of 300 to 1100 nm wavelength. However, for our purpose, we could not use its light source, which is connected by an optical fibre to the detector itself. Instead, we have recorded transmission spectra of the glass fragments in two steps: using an incandescent lamp first and then a high-pressure mercury lamp. Transmitted light intensity through the fragments of perpendicular incidence was divided by the direct incident light intensity of the lamps to obtain transmission spectra. Spectra recorded using a mercury lamp below, and those of an incandescent lamp above 555 nm were joined together and scaled to match exactly at 555 nm. The combined spectra were completely smooth at the junction. However, to check the validity of this operation, we also registered all transmission spectra using an Agilent 8452 diode array spectrophotometer, fixing the glass fragment at the position of the test cuvette against no object in the reference cuvette position. Scaling the two spectra exactly to the same amplitude resulted in an almost perfect overlap. To measure the transmittance of the bottles in situ, we needed the detector of the fibre-optic spectrometer and external light sources; this is the reason to use it also for the simpler task of measuring the transmission of glass fragments. Results of the measurements using the AvaSpec-2048-USB2 versatile fibre-optic spectrometer are shown in Figure 5, with all transmission data corrected to the same 3 mm glass thickness. The thickness of individual glass fragments varied for an average thickness of bodies as follows: clear Bordeaux 3.15 ± 0.10 mm, bluish-green Alsace 3.19 ± 0.45 mm, light amber Burgundy 2.97 ± 0.27 mm and dark amber Alsace 2.41 ± 0.18 mm.



FIGURE 1. Wine bottles studied experimentally and modelled for light damage of white wine bottled in them. From left to right: clear (flint), bluish-green, light amber and dark amber bottle.

Note that the flint bottle (leftmost) is filled with rosé wine to show its shape better.

We only recorded one spectrum of a piece of glass of each bottle and measured its actual thickness. Corrections to 3 mm thickness were made according to the Lambert law of light attenuation. To have an idea of the uncertainty of transmission of the bottle material, we supposed that the quality of the glass is homogeneous, but the actual thickness of the bottle wall varies. Thus, we have measured the thickness of several fragments from the same bottle type and considered the difference between the smallest and the greatest measured thickness as a robust estimate of the uncertainty of wall thickness. These estimates are given above after the \pm sign.

Transmission of wine has been measured using the Agilent 8452 diode array spectrophotometer the standard way: wine in a 1 cm quartz cuvette and a reference of distilled water also in a 1 cm quartz cuvette.

2. Transmitted light measured within bottles

To measure the light transmission of real bottles, we have used the experimental setup shown in Figure 2. The bottom of the bottles had been cut off, and the detector of the fibre-optic spectrometer was introduced vertically up to a position of 20 cm from the closure of the vertically aligned bottle. The detector has a collector lens and measures the total intensity of light within an 11° conical field of vision. Cork, capsule and label(s) were at their original place.

Artificial light sources were placed 1 m above the bottle so that incident light was parallel to the axis of the bottle. This arrangement resulted in maximum transmitted light intensity in the bottle. Changing the angles of the light source moved to the side, we always measured less transmitted intensity.

In the case of sunlight from the clear sky, the arrangement of the bottle and detector was the same, but the axis of the bottle and that of the detector was bent to 45° with respect to the perpendicular direction in a shaded place so that the experimental arrangement was illuminated not by the direct sunshine, but it was facing the clear sky on a sunny summer day, at the side of a building which hid the sun. The main reason for this arrangement is that a clear sky is an ideal homogeneous light source in contrast to our point-like artificial light sources; thus, it provides transmission data for eventual homogeneous lighting, which might be the case—though rarely—also in a wine store. Another reason was that wine is rarely exposed to direct sunshine; a shaded place has far less danger both for light strike and warming of the wine; thus, this arrangement is also more lifelike.

Transmission spectra were derived as described above: transmitted light intensity measured inside the bottle was divided by the direct incident light intensity in the same arrangement but with the bottle removed. Thus, we also recorded the emission spectra of the light sources.



FIGURE 2. Experimental arrangement to measure light intensity entering wine bottles in situ.

The bottom of the bottle is cut off; the light detector is introduced in a vertical position at the centre, 20 cm from the closure. The light source is in a central perpendicular position, 1 m above the bottle closure.

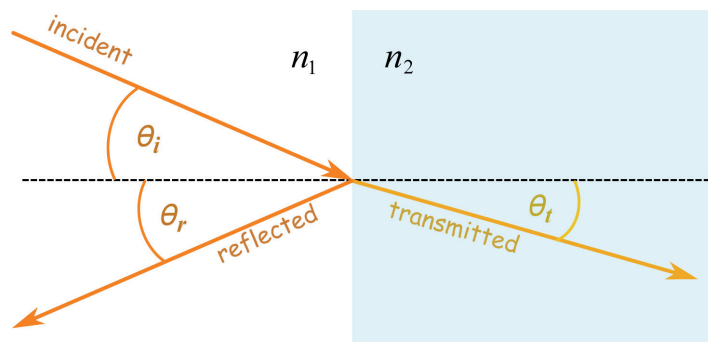


FIGURE 3. Notation for transmission calculations through an interface.

The (colourless) left medium is air, and the (blue) right one is the glass material of the bottle. The dashed black line is the direction of perpendicular incidence to the air/glass interface. θ_i is the angle of incidence, i.e., the angle of the incident beam; θ_r that of the reflected beam and θ_t that of the transmitted beam, with respect to perpendicular incidence. n_1 is the refractive index of air, and n_2 is that of the glass material.

3. Transmission and photon flux calculations

We can calculate the reflection when a light beam hits a denser medium coming from a less dense one using the Fresnel equations (Hecht, 2002; Clarke, 2010). According to these equations, the amplitude reflection coefficient for polarised light perpendicular to the plane of incidence (the plane of Figure 3) is given by

$$r_{\perp} = -\frac{\sin(\theta_i - \theta_t)}{\sin(\theta_i + \theta_t)}, \quad (1)$$

and that of the polarised light parallel to the plane of incidence by

$$r_{\parallel} = \frac{\tan(\theta_i - \theta_t)}{\tan(\theta_i + \theta_t)}. \quad (2)$$

According to the Snell–Descartes law, the relative refractive index n_2/n_1 is the ratio of the sine of the angle of incidence to that of the sine of the angle of transmittance: $n_2/n_1 = \sin \theta_i / \sin \theta_t$; thus, we can eliminate θ_t from the

equations by considering n_1 of the air as unity (instead of ≈ 1.0003) and substituting $\theta_t = \arcsin(\sin \theta_i / n_2)$:

$$r_{\perp} = -\frac{\sin(\theta_i - \arcsin(\sin \theta_i / n_2))}{\sin(\theta_i + \arcsin(\sin \theta_i / n_2))}; \quad r_{\parallel} = \frac{\tan(\theta_i - \arcsin(\sin \theta_i / n_2))}{\tan(\theta_i + \arcsin(\sin \theta_i / n_2))}. \quad (3)$$

The reflection coefficient (reflectance) of the polarised light can be given as the square of the amplitude reflection coefficient, and the reflectance of nonpolarised light is the average of the reflectances of perpendicular and parallel polarised lights; thus:

$$R = \frac{r_{\perp}^2 + r_{\parallel}^2}{2}. \quad (4)$$

Finally, based on the relation that the sum of the reflected and transmitted intensities is the same as the incident intensity, the transmittance (at the surface of the incidence) can be given as

$$T = 1 - \frac{r_{\perp}^2 + r_{\parallel}^2}{2}. \quad (5)$$

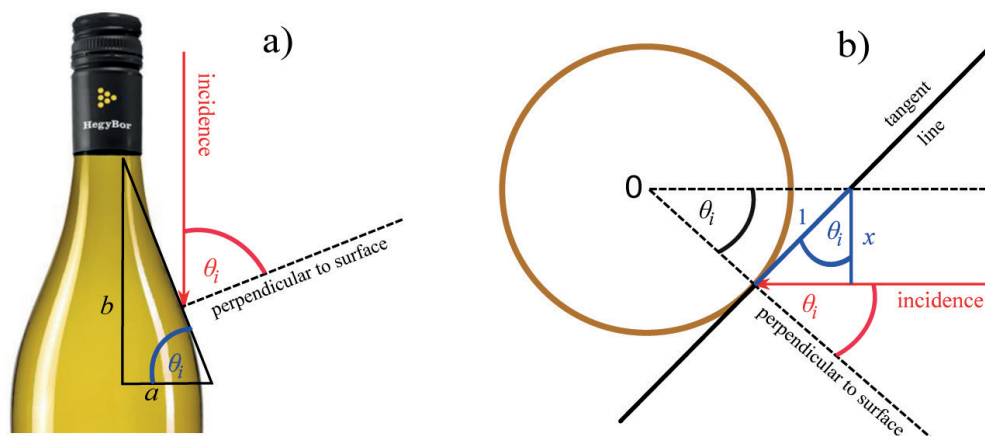


FIGURE 4. Notation for transmission calculations a) for vertical and b) for horizontal (side) incidence of the parallel light beam based on conical and cylindrical shapes of the relevant part of the bottle.

In the case of vertical parallel illumination of the bottle, the reflectivity can be calculated with fair precision using the incident angle θ_i based on the conical shape of the upper part of the bottle. In case of side illumination of the entire cylindrical body, the weighted average reflectivity between $\theta_i = 0^\circ$ and $\theta_i = 90^\circ$ should be used, the weight being proportional to $1/\cos \theta_i$. In this latter case, the figure only shows the circular section of the cylindrical shape, with the origin of the circle denoted by O .

Summing up: to calculate the transmittance of an air/glass interface, we need to know the refractive index of the glass n_2 and the angle of incidence θ_i .

For parallel vertical incidence on the top of the bottle, the angle of incidence θ_i can be taken equal to the angle between the radius and the generatrix of a cone that fits the upper part of the bottle around the neck. It can be seen from Figure 4a that $\theta_i = \arctan(b/a)$. For parallel, perpendicular side incidence, it can be calculated based on the cylindrical shape of the bottle body. As it can be seen in Figure 4b, showing a circular section of the cylinder, the angle of incidence, in this case, varies from 0° to 90° in each direction from the perpendicular incidence (shown in the figure as a horizontal dashed line). Thus, the overall reflectance in the case of a homogeneous parallel side illumination of the entire cylindrical body is the weighted average of the calculated reflectance as a function of the angle of incidence θ_i between 0° and 90° . The weight takes into account the increase in the length of the arc seen by the unit aperture of the incident beam with increasing θ_i . At a small enough increase in the angle θ_i , the length of the arc is well approximated by the length of the segment. According to the blue orthogonal triangle in Figure 4b, the weight is proportional to $1/\cos \theta_i$, as $\frac{1}{x} = \cos \theta_i$, thus, $x = \frac{1}{\cos \theta_i}$.

Calculation of the incident photon flux entering the bottle is more complex; to do this, we should have photometric data of the light source in the entire wavelength range we are interested in. Let us consider first the radiative power of direct sunshine; these data—the solar radiation flux density as a function of wavelength—are easily accessible on the NREL website (NREL, 2003; Gueymard *et al.*, 2002). For incident direct sunshine on Earth's surface, the closest conditions available are tilted pyranometers facing the sun and measuring direct incidence plus a 2.5 degrees circumsolar component. These solar radiation flux density data are typically tabulated by 1 nm wavelength increments in $\text{W} \cdot \text{m}^{-2} \cdot \text{nm}^{-1}$ units. This is the power P^λ of the radiation on 1 m² surface, per 1 nm spectral width at the wavelength λ . If the wavelength increment is different from 1 nm, it should be taken into account properly when performing numerical integration.

As 1 W = 1 J/s and the energy of a photon is $E = hv = hc/\lambda$, the number of photons per second at the wavelength λ is given by $N^\lambda = \frac{P^\lambda \lambda}{hc}$, where h is Planck's constant and c is the velocity of light. In fact, N^λ is a differential quantity; it should be integrated over the entire wavelength range we are interested in. In the special case, if tabulated data are given by 1 nm increments, this integration is equivalent to the sum of all tabulated data in a given wavelength range:

$$N = \sum_{\text{all wavelengths}} \frac{P^\lambda \lambda}{hc} \quad (6)$$

is the total number of photons hitting 1 m² surface in 1s time.

$$S^2[F(\vartheta_1, \vartheta_2 \dots \vartheta_p)] = \sum_{i=1}^p \left(\frac{\partial F}{\partial \vartheta_i} \right)_{t_i}^2 S^2(T_i) + 2 \sum_{j=1}^p \sum_{i < j} \left(\frac{\partial F}{\partial \vartheta_i} \right)_{t_i} \left(\frac{\partial F}{\partial \vartheta_j} \right)_{t_j} \text{Cov}(T_i, T_j), \quad (\text{Equation 7})$$

Considering a wine bottle having roughly 1 dm² free surface facing the sun, N should be divided by 1000 and multiplied by the overall transmittance of the bottle, taking into account its geometry facing the sun.

In the case of artificial light sources, this calculation can also be done if we know the radiation flux density of the light source used. As the measurement with a reliable spectroradiometer is quite complicated, we did not measure data for this case. However, we have found data in the literature (Eadie *et al.*, 2009) that are almost identical to the radiation spectrum of the compact fluorescent lamp (CFL) we have used. Thus, we have scaled our relative spectral intensity data to a characteristic phosphor peak at 487 nm to have the same radiation flux as the one in Eadie *et al.* (2009) then calculated the emitted number of photons from these values. Our data are not measured at unit wavelength increments; thus, we have also taken this into account when integrating the number of photons in the entire wavelength range of interest.

As different commercial CFLs have quite similar radiation spectra in the wavelength range of 320 to 440 nm, we can base our estimations on the illuminances (luminous fluxes incident on a surface in units of $\text{lux} = \text{lumen/m}^2$). Though luminous flux also includes weighting the radiative power with the sensitivity of the human eye—which for photopic vision has nonzero values above 400 nm wavelength (Smith, 2000)—a fairly good guess of the radiation power can be obtained supposing its proportionality in the wavelength range of 320 to 440 nm to luminosity—which is always given in case of current lighting devices.

The relevant quantity from the point of view of developing light-struck flavour is the *absorbed* photon flux by the wine within the bottle which can be calculated from known and measured parameters. If the incident radiative flux is denoted by I_o , in situ transmission by T , absorption of wine by A , and the quantum efficiency of the formation of harmful products by Φ , then the reaction rate can be calculated as the product $I_o \cdot T \cdot A \cdot \Phi$.

4. Error calculations

According to standard statistics, the sample variance S^2 of a function $F(\vartheta_1, \vartheta_2, \dots, \vartheta_p)$ of p parameters ϑ_1 to ϑ_p measured with a random error can be given the following way:

(see equation 7 below ▼), where T_i is the estimator of ϑ_i , $\left(\frac{\partial F}{\partial \vartheta_i} \right)_{t_i}$,

is the value of the partial derivative of the function F with respect to its variable ϑ_i at the realisation t_i of the estimator T_i (similarly for j), $S^2(T_i)$ is the sample variance of the estimator T_i and $\text{Cov}(T_i, T_j)$ is the sample covariance of the estimators T_i and T_j . In case the random errors are independent, the second term containing the covariances can be dropped. In a univariate case (i.e., when F is only the function of a single variable ϑ), there is no covariance term, and the relevant derivative is a simple one.

5. Organoleptic analysis of illuminated wine

Tasting experiments aimed to detect an approximate critical time until the onset of the light-struck flavour. We have followed a simplified version of the triangle protocol (Meilgaard and Carr, 2007), with two tasters performing the evaluation. Wine samples from two bottles were poured into three coded glasses; tasters should decide which is the odd one if any. A difference between the duplicate and the odd samples has been concluded only if the two taster's opinions coincided. In case a difference was found, the irradiated (or longer-time irradiated) wine was evaluated for the characteristics of the light-struck flavour by agreement of the two tasters on the aroma notes used to characterise wine faults.

For the first session of testing, four series involved illuminated wine duplicates and a non-illuminated odd from the dark cellar. Bottles were either bluish-green Alsace or clear (flint) Bordeaux type, with cork and label. For other series (and for the second tasting session), we have combined different bottle types not illuminated, illuminated for the same, or for different times. We have applied three different illumination times: 15, 75 and 150 hours, with the same 20 W/1250 lm/2700 K ('soft white') commercial CFL lighting device in a cellar of 12 °C temperature, with no other light source, switched on. The light source was placed at 40 cm from the bottle body, irradiating its central part horizontally where a label of 60 × 85 mm was facing the irradiating CFL.

Wine in all the bottles were identical at the start of the experiments: a "Badacsony Olaszrizling" (better known as Welschriesling) of the 2019 harvest from Part Pince, Badacsony, Hungary. Characteristics of the wine are the following: 15.35 v/v% alcohol; < 1 g/L sugar; 4.6 g/L acid of which 0.3 g/L volatile acidity; 15 mg/L free SO₂ and pH = 3.61.

RESULTS

1. Transmission spectra of wine bottle glass materials and absorption of white wine

As expected, the transmission spectra shown in Figure 5a are in accordance with previous findings (Dias *et al.*, 2010; Hartley, 2008). Clear glass has a transmittance of well above 80 % between 360 and 440 nm; bluish-green glass slightly above 60 % between 360 and 395 nm; light amber glass around 2 % between 340 and 390 nm; while the transmission of dark amber glass is below 0.3 % in the entire 320–440 nm range of the harmful light. Though we could formulate already some recommendations based on the above results, we are more interested in the practically relevant situation: what is the actual amount of light transmitted from common lighting devices and daylight into a bottle on a shelf, having a cork, a capsule, and label(s) on its body surface.

To see the light-sensitivity of white wine in the harmful wavelength range, Figure 5b shows the absorption spectrum of the wine we used for organoleptic tasting experiments. The spectrum was recorded in a 1 cm quartz cuvette, using distilled water as a reference.

2. Transmission spectra measured in situ in bottles

In situ measurements have been made using sunlight and two kinds of artificial light sources: a compact fluorescent lamp and a LED bulb. As explained above, the transmission of light into the bottles has been derived from the light intensity measured inside the bottles divided by the measured intensity of the incident light. The latter data were, in fact, the emission spectra of the light sources. In the case of sunlight measurements, light arrived at the bottles from a large radiating surface (the clear sky), while in the case of artificial light, it arrived from a point-like source. It turned out that this difference also resulted in considerably different transmission into the bottles.

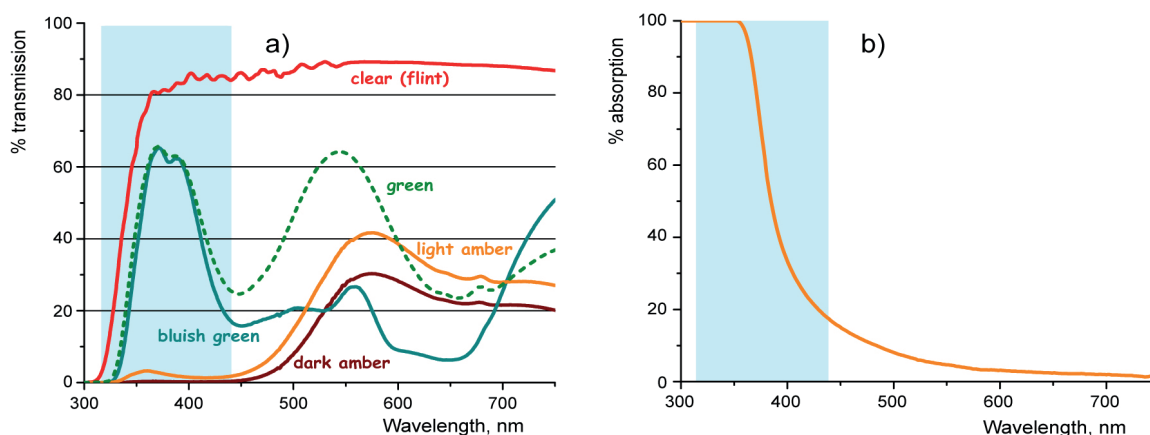


FIGURE 5. a) Transmission spectra of different colour glass materials of 3 mm thickness. b) Absorption spectrum of the tasted white wine for comparison.

Clear, bluish-green, light- and dark amber glasses (solid curves) were measured in this study; green glass (dashed curve) data have been taken from Hartley (2008) and scaled to match the bluish-green spectrum between 320 and 440 nm. The blue window indicates the photochemically harmful wavelength range of 320–440 nm.

2.1. Emission spectra of different light sources

The measured intensity of emitted light from different light sources is shown in Figure 6. As we can see, all sources have important radiation intensity in the harmful spectral range. However, sunlight and fluorescent light have by far more emission in this region than either the (nowadays outdated) incandescent lamp or the (recently developed and increasingly in use) LED light sources. Concerning LEDs, lower colour temperature ('warm' or 'soft') light bulbs obviously emit less in the upper edge of the harmful spectrum range.

There is a wide variety of indoor fluorescent light tubes available with different photoluminescent pigments ('phosphors'), but all of them are based on mercury vapour; thus, the characteristic visible spikes of mercury emission are always present in their emission spectra. We have measured the emission spectrum of a 20 W "soft white" compact fluorescent lamp (CFL) with a total emission of 1250 lm and a colour temperature of 2700 K. As can be seen in Figure 6, mercury spikes at 405 and 436 nm have an important contribution to the emitted light even of this low colour temperature CFL.

2.2. Measured in situ transmission spectra of different wine bottles

Results of the measurements using the method described in Section 2 of Materials and Methods are shown in Figure 7. The most striking feature we can see is the difference in transmissions between a homogeneously distributed incident light (sunlight from the sky) and a point-like source. The background of this difference is the dependence of reflection (and transmission) of the glass surface on the angle of incidence, as described in Section 3.

Homogeneously distributed sunlight from all directions of the sky enters the whole surface of the bottle not covered by the cork or the labels. As a result, there is a relatively high transmitted intensity to measure inside the bottles, whose spectral distribution is quite similar to that of the glass material (with a perpendicular incidence). However, this is not the typical situation where wines are exposed to light; on the shelves in a store, illumination is provided by point-like artificial light sources. Thus, we have also made measurements, as illustrated in Figure 2, using an 8 W LED bulb emitting 720 lm with a colour temperature of 4000 K. As there is practically no emission from this bulb below 400 nm (Figure 6B), we have extrapolated measured in situ spectra by downscaling the glass material's transmittance (Figure 5) so that the downscaled continuation value at 430 nm exactly matched the measured value of the in situ transmission using the LED bulb. As in situ transmission should be proportional to the transmission of glass material, this procedure provides reliable values of transmission below 430 nm with the same geometry of illumination. However, it is no more the *observable* transmission of the LED bulb but potential transmission if there is some important emission intensity of a different point-like source in this range.

Differences between the two cases can be understood based on the fact that sunlight can enter from a wide range of angles of incidence, while artificial light sources irradiate bottles from one single direction only. Thus, we can conclude that daylight illumination results in almost twice as much transmission into the bottles than artificial lighting—at least if the light sources are point-like and artificial light does not come from a large surface in a diffuse manner—which is rarely the case. In the case of homogeneously diffuse artificial lighting, transmission should be the same as detected for the daylight illumination from the clear sky.

Comparing in situ transmittivities of the bottles in the 320–440 nm range to those of the glass material, the clear bottle's transmittance is below 40 % exposed to sunshine and well below 30 % in artificial light. Coloured bottles have even more reduced transmittance; bluish-green below 8 % and both light and dark amber below 2 %, even in case of illumination with sunlight. Note that the clear bottle studied had a Bordeaux shape, while the coloured bottles had Burgundy (light amber) and Alsace (dark amber and bluish-green) shapes.

Measured in situ transmissions given in Table 1 also have their uncertainties indicated. To calculate these uncertainties, we considered that in the case of homogeneous glass material, its origin is in the nonhomogeneous thickness distribution of the bottle wall. Thus, we calculated in situ bottle transmission error using the propagation of error rule explained in Section 4 of Materials and methods. Transmission as a function of thickness can be explained as $T = e^{-\alpha \ell}$, from which the absorption coefficient α can be calculated for a known optical path length (in this case, average thickness) ℓ . Once the coefficient α is obtained, we can apply the univariate version of the error propagation rule in the following form:

$$S^2[T(\ell)] = \alpha^2 e^{-2\alpha \ell} S^2(\ell) \quad (8)$$

We performed the calculations in the middle of the harmful wavelength range at 365 nm. In the case of three bottle types, calculated errors were practically identical. However, for the bluish-green Alsace bottle, the uncertainty in the case of transmission of light from the clear sky was ± 3.0 %, while in the case of a point-like artificial light source, only ± 2.0 %. The difference is due to enhanced differences in the respective absorption coefficients in these two cases.

3. Calculated in situ transmittance of bottles of different colours and shapes

It is interesting to check how calculated transmissions based on optical and geometrical modelling compare to experimentally determined transmittance results. Calculated¹ transmissions for the more bulky Bordeaux and Burgundy bottles overestimate measured ones by a factor of 2.9 and 2, respectively, while for the slim Alsace bottles, this overestimation is considerably higher: somewhat more than three times for the dark amber and more than eight times for the bluish-green bottle.

¹ Details of the calculations can be found in the Supplementary Material S2.

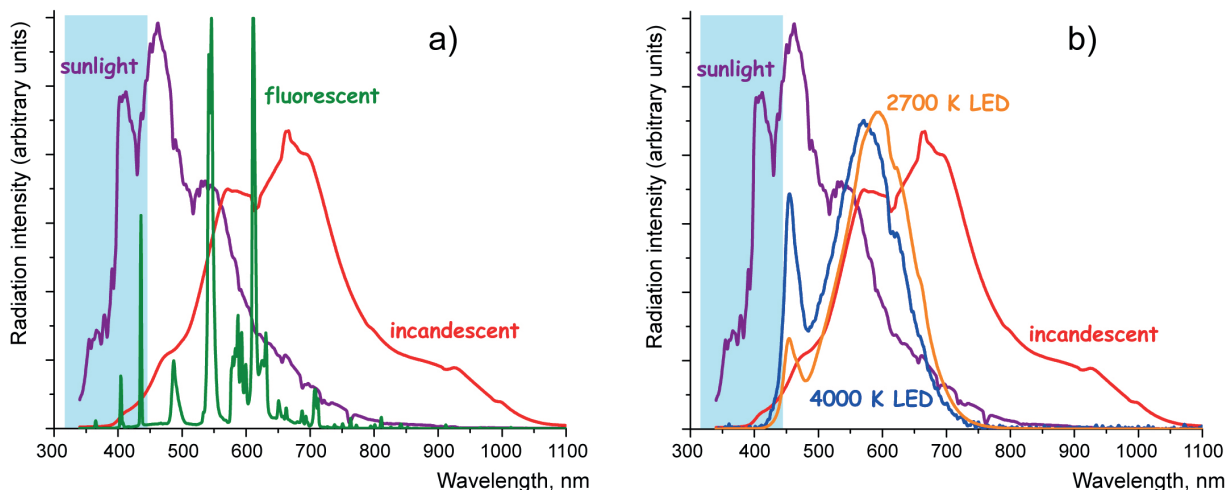


FIGURE 6. Emission spectra of a) fluorescent and b) LED light sources compared to sunlight and incandescent lamp.

To avoid crowded pictures, two diagrams are shown. Amplitudes are scaled so that they fit into the same diagram. The blue window indicates the photochemically harmful wavelength range of 320–440 nm.

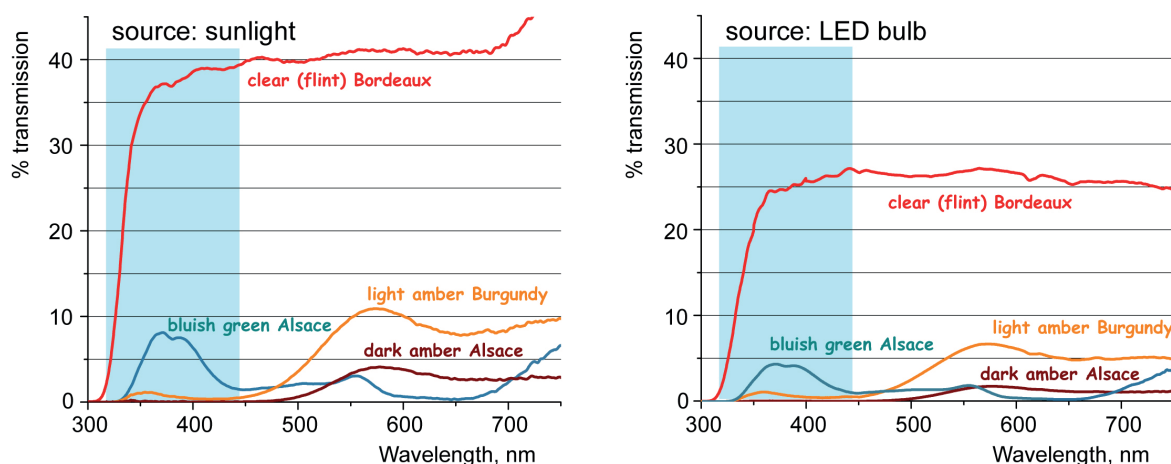


FIGURE 7. In situ transmission of wine bottles of different colours and shapes.

The left panel shows the percentage of incident indirect sunlight entering wine bottles. The right panel shows the percentage of incident light from a LED bulb entering wine bottles. Blue windows indicate the photochemically harmful wavelength range of 320–440 nm.

TABLE 1. Calculated and measured in situ transmission of wine bottles compared.

bottle colour and type	calculated	measured	calculated/measured
Clear Bordeaux	57.2 %	19.7 ± 1.2 %	2.9
Bluish-green Alsace	20.2 %	2.4 ± 2.0 %	8.3*
Light amber Burgundy	1.82 %	0.89 ± 0.4 %	2.0
Dark amber Alsace	0.13 %	0.04 ± 0.03 %	3.4

Values following ± signs indicate uncertainties of the measured in situ transmissions.

*Note that studied bluish-green Alsace bottles have extremely large labels. Uncertainty of the measured transmission, in this case, is different for sunlight from the sky and a point-like artificial light source. For sunlight, it is ± 3.0 percent, while for a point-like source, it is the value given in the table: 2.0 %.

One reason for the large overestimation might be the placement and size of the labels. Bluish-green Alsace bottles used in this study have much larger labels than the other three—see pictures of bottles in Figure 1. Another reason might be the worse fit of the cone to the bottle shoulder in the case of Alsace bottles used in calculations.

Though the considerably simplistic calculation we performed overestimates the measured *in situ* transmission, it can be used to get a robust estimate in favour of the necessary light protection of wines. In case we do not have measured data for light transmission into bottles in the harmful wavelength range of 320–440 nm, simple calculations shown here can help to devise efficient protection as they always overestimate the transmitted light dose.

3.1. Calculation of reaction rate of harmful photochemical processes

Once we have reliable data for the transmission of light into bottles and the radiation flux density of the light source used in a wine store, we can also calculate the approximate photon flux entering the bottles and, from this, the approximate reaction rate of wine degradation; thus, also the expected shelf life of the wine before discernible off-aroma formation could occur.

We have calculated the photon flux entering the bottles as described in Section 3 of Materials and Methods. Reaction rates are given in *moles* of (off-flavour) products per second; thus, the suitable unit for the number of photons per second is *moles* of photons per second, which is often called *einstein*. (However, *einstein* is not part of the SI system of units; we shall use here ‘mole photons’ instead.) As the perception threshold of light-struck products is in the range of nanomoles per litre, we shall also use the nmol/s unit for photon flux, as well as for the reaction rate. From the quantities in the formula to calculate the reaction rate as the product $I_0 \cdot T \cdot A \cdot \Phi$, incident photon flux I_0 is estimated from known data for different light sources and lighting situations, T is measured *in situ* transmission, quantum efficiency Φ is taken from literature, and we use a reliable guess for the absorption A of the wine within the bottle.

We have calculated the incident photon flux I_0 as described in Section 3 of Materials and Methods. Having taken the intensity of sunlight from an overcast sky to be 100 times inferior to that of the bright sunshine (i.e., approximately 1000 lux), the flux in the harmful wavelength range is 22.4 nmol/(dm²·s). Numerical integration of the scaled 1250 lumen ‘soft’ CFL radiative power in the same wavelength range at a distance of 20 cm resulted in 2.127 nmol/(dm²·s) photon flux. From this, we can estimate the fluxes for a bright illumination store of 500 lux to be 0.85 nmol/(dm²·s) and that of a dimmer illumination of 100 lux 0.17 nmol/(dm²·s), respectively.

The transmission of light into the bottles has been taken from Table 1.

Quantum efficiency for the formation of sulfur-containing off-flavour compounds is reported to be 0.26 between 365 and 490 nm in a model wine at pH = 4, independently of wavelength. (Furet *et al.*, 2021). To have an idea of the absorbed photon flux in wine, we have used the measured absorptivity of the tasted wine. Based on the data shown in Figure 5b, we have calculated an average absorption of 61.6 % along a 1 cm path length in the range of 320 to 440 nm; thus, absorption in the entire bottle could be close to 100 %. However, a rough measurement of the light intensity passing through the full bottles in case of side illumination, measured 20 cm behind the bottle, has shown that there is somewhat more than 30 % transmission for the clear Bordeaux bottle and more than 15 % for the bluish-green Alsace bottle. Based on these results, we have used 80 % absorption A by the wine in the entire bottle.

Using these quantities to calculate reaction rates for the formation of off-flavour compounds and taking into account the sensory threshold of these species, we can also calculate the shelf life defined as the formation of the off-flavour compounds up to an organoleptically discernible quantity. According to Fraccasetti *et al.* (2021), the perception threshold of MeSH is 10 µg/L, and that of DMDS is 45 µg/L, respectively, with a considerably high uncertainty of 38 %. From this, we took an average of these quantities resulting in a 300 ± 114 nmol/L threshold, which should be divided by the calculated reaction rate to get the critical time of off-flavour formation to a noticeable degree. The results of these calculations are summarised in Table 2. Details of the calculations can be found in Supplementary material S2. In Table 2, we have also given shelf lives for the 313 lux irradiation to compare calculated shelf lives with the results of organoleptic tasting analysis.

We have also estimated the uncertainty of the above model calculations. Shelf lives can be calculated using the above quantities according to the formula

$$t = \frac{c}{I_0 T A \Phi}, \quad (9)$$

where t is the shelf life, c is the average sensory threshold concentration of sulfur-containing off-flavour compounds, I_0 , T , A and Φ are defined above. Applying the error propagation rule, we get the following expression for the variance of the shelf life:

(see equation 10 below ▼)

In the above expression, we have dropped the additive term, $(T A \Phi)^2 S^2(I_0)$ in square brackets, as shelf lives considered here are related to exact (errorless) incident light intensities. However, if the light intensity is uncertain in an actual lighting and shelf arrangement, this term should also be added to the

$$S^2(t) = \left(\frac{1}{r}\right)^2 S^2(c) + \left(-\frac{c}{r^2}\right)^2 [(I_0 T A)^2 S^2(\Phi) + (I_0 T \Phi)^2 S^2(A) + (I_0 A \Phi)^2 S^2(T)], \quad (\text{Equation 10})$$

four terms within the square brackets. Taking into account that the reaction rate can be expressed as $r = I_0TA\Phi$, Eq. 10 can be simplified to the following compact form:

$$S^2(t) = \frac{1}{r^2} \left[S^2(c) + c^2 \left(\frac{S^2(\Phi)}{\Phi^2} + \frac{S^2(A)}{A^2} + \frac{S^2(T)}{T^2} \right) \right] \quad (11)$$

Concerning contributions of the remaining four variables c , Φ , A and T to the uncertainty of the calculated shelf life, we have used the following variances. For the *in-situ* bottle transmission, values given in Table 1 are used. For the average threshold concentration of sulfur-containing off-flavour compounds, the equivalent of a 38 % relative error was reported by Fracassetti *et al.* (2021), i.e., 114 nmol/L is used. Furet *et al.* (2021) also report a relative error of 15 % for their quantum efficiency of the formation of sulfur-containing off-flavour substances, which in our case, is equivalent to an absolute error of 0.039. The uncertainty of the estimated 80 % transmission of the wine within the bottles can only be ‘guessed’ based on the measurements described above. We adopted 10 % uncertainty which seems to be quite reasonable.

Uncertainties shown in Table 2 are calculated using these values. The largest contribution to the error comes from the large variance of the threshold concentration of sulfur-containing off-flavour compounds, which reflects considerable differences in organoleptic sensitivity among professional tasters as well as the large variability of different wine matrices. Another important contribution is associated with the sensitivity of the error to the calculated reaction rate r . As can be seen in Eq. 10, this contribution to the variance is proportional to the inverse fourth power of r .

As a consequence, the smaller the reaction rate, the greater the relative error of the calculated shelf life becomes.

4. Tasting experiments

The results of the organoleptic tasting are summarised in Table 3. We can conclude that the light-struck flavour has developed already within 15 hours in the clear Bordeaux bottle, while it was not detectable in the bluish-green Alsace bottle after 15 hours, only after 75 hours illumination. A comparison of different illumination times and different bottle types indicate that wine in bluish-green Alsace bottles deteriorates somewhat slower than in clear Bordeaux bottles. These results are in agreement with previous findings (Dozon and Noble, 1989), though they used green bottles; but as it can be seen in Figure 5, they are supposed to have the same transmission in the harmful wavelength range.

Tasters have also evaluated the flavour notes of deteriorated wines. In the case of the clear Bordeaux bottle—though there was no detectable change in taste after 15 hours illumination—light-struck notes (vegetal, rotten cabbage) were clearly present in the smell, along with a slight drains component. However, after two days re-tasting, also the taste has changed. After 75 hours illumination, apple aromas appeared with related oxidation notes and further light-struck notes like rotten egg and onion, along with a reduced acid taste. These smells and aromas became more intense after 150 hours illumination, along with the appearance of an asparagus aroma. Summing up the results, tasters did find off-aroma compounds related to sulfur-containing light-strike products and also oxidative notes but did not see any browning of the wine even after 150 hours illumination.

TABLE 2. Calculated shelf life of white wine in studied bottles with different illumination conditions.

Perpendicular incidence				
Bottle type	Illumination	Transmission at 350 nm	Shelf life hours	Shelf life days
Clear Bordeaux	sunlight	33.8 %	0.08 ± 0.024	0.01 ± 0.002
	CFL strong ¹	19.7 %	3.6 ± 1.28	0.3 ± 0.11
	CFL weak ²	19.7 %	17.8 ± 6.4	1.5 ± 0.53
Bluish-green Alsace	sunlight	4.3 %	0.63 ± 0.49	0.05 ± 0.041
	CFL strong	2.4 %	29.0 ± 23.6	2.4 ± 1.97
	CFL weak	2.4 %	144.9 ± 118	12.1 ± 9.8
Light amber Burgundy	sunlight	1.0 %	2.72 ± 0.14	0.23 ± 0.012
	CFL strong	0.9 %	79.1 ± 42.9	6.6 ± 3.6
	CFL weak	0.9 %	396 ± 215	33.0 ± 17.9
Dark amber Alsace	sunlight	0.06 %	45.3 ± 0.44	3.8 ± 0.036
	CFL strong	0.04 %	1761 ± 1259	146 ± 105
	CFL weak	0.04 %	8803 ± 6294	734 ± 525
Side incidence				
Clear Bordeaux	CFL 313 lux	19.7 %	5.7 ± 2.1	0.5 ± 0.17
Bluish-green Alsace	CFL 313 lux	2.4 %	46.3 ± 37.7	3.9 ± 3.14

Values following ± signs indicate uncertainties in the calculated shelf lives.

¹ ‘CFL strong’ is for a bright store illumination of 500 lux.

² ‘CFL weak’ is for a dimmer store illumination of 100 lux.

TABLE 3. Results of the tasting experiments. In each of the two tasting sessions on different days, eight “triangle” samples were tested, each containing the same white wine. Samples are identified with bA for bluish-green Alsace and cB for clear Bordeaux bottles. Numbers after the bottle identifier indicate the time of illumination in hours. No number indicates a non-irradiated bottle. ‘–’ sign means no perceptible difference between the three samples, ‘+’ sign indicates a relevant difference between the duplicates and the odd sample. The first tasting session was performed on the third and the second session on the sixth day after illumination started.

Series	Samples			Difference*
I.	bA 15	bA 15	bA	–
II.	cB 15	cB 15	cB	+
III.	bA 75	bA 75	bA	–
IV.	cB 75	cB 75	cB	+
V.	bA 15	bA 15	cB 15	+
VI.	cB 75	cB 75	bA 75	+
VII.	bA 15	bA 15	bA 75	+
VIII.	cB 75	cB 75	cB 15	+
I.	cB 75	cB 75	cB	+
II.	bA	bA	bA 75	+
III.	cB 150	cB 150	cB	+
IV.	bA	bA	bA 150	+
V.	cB 75	cB 75	bA 75	+
VI.	bA 150	bA 150	cB 150	+
VII.	bA 75	bA 75	bA 150	–
VIII.	cB 75	cB 75	cB 150	+

*Two tasters evaluated the samples; relevant difference means that both reported a difference.

In the case of the bluish-green Alsace bottle, there was no detectable change at all after 15 hours illumination. After 75 hours illumination, the same oxidative and light-struck aromas were present as in the clear bottle, along with some wet-wool flavours and reduced acid taste. However, there was no relevant difference between the samples after 75- and 150 hours illumination.

When tasted two days later, samples did not exhibit a further detectable organoleptic change in either bottle type.

Results of the organoleptic analysis thus support the reliability of the calculations based on in situ transmission measurements, literature data on the perception threshold of off-aroma compounds, and optical and photochemical modelling of light-strike phenomena as described here.

DISCUSSION

The study aimed to measure the transmission of light into wine bottles in situ with a detector inside the bottle having a cork, capsule and label(s) in their original place and calculate the reaction rate of light-strike phenomena based on optical and photochemical modelling to estimate the shelf life of white wine in different colour bottles studied. We have found that within real-life conditions, considerably less light is transmitted into bottles than the transmission of the glass material bottles are made of. Natural sunlight coming from the sky—an ideal mimicking of homogeneous diffuse illumination—has a higher transmission than artificial light

from point-like sources. Apart from the distance of lighting devices from the bottle and the direction of incidence, the colour of the glass and the shape of the bottle also have a great influence on the actual transmission into the bottle.

Clear (flint) bottles and bluish-green ones transmit a large part of the light in the range of 320 to 440 nm, which causes the development of light-struck flavour and degradation of characteristic aroma notes. Green bottles have the same transmittance in this wavelength range as bluish-green ones studied; thus, this applies to them as well. Amber bottles transmit substantially less light in the harmful wavelength range, especially dark amber ones. As a consequence, white wine in clear, green and bluish-green bottles survives light-strike only for a few days, while the shelf life in light amber bottles is about a month and in dark amber bottles about a year or more. Concerning clear and bluish-green bottles, tasting experiments confirm the results of these model calculations, which is also in accordance with the results of previous studies reported in the literature (Dozon and Noble, 1989).

In accordance with recent findings, our results support recommendations that flint glass and green (as well as bluish-green) bottles are better avoided from the bottle assortment wine is held in. However, there are methods to protect wine, even in these light-sensitive bottles. We do not consider the addition of different adjuvants or removal of riboflavin and pantothenic acid from the wine as suitable chemical protection (Fracassetti *et al.*, 2021). A better approach would be to completely avoid sunlight and use artificial lighting that

has as small a contribution in the harmful wavelength range of 320–440 nm as possible. Evidently, CFL sources are not suitable for this purpose as they all emit at the characteristic spikes of the mercury vapour (405 and 436 nm). According to our measurements concerning the emission spectra of light sources, ‘soft white’ (or ‘warm white’) LED lamps are more apt for this purpose as their emission is quite low in the critical wavelength range.

In cases when the wine in light-sensitive bottles is exposed to lighting with harmful wavelength components, some kind of sensors to record total irradiation doses in this wavelength range could help to detect the limit of shelf life of different wines in different bottles, as suggested by Arapitsas *et al.* (2020).

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Supplementary material concerning measured spectra (*S1-MeasuredSpectra.xlsx*), reflectance, photon flux, reaction rate and shelf life calculation (*S2-ReflectancePhotonFluxCalculation.xlsx*), results of the tasting experiments (*S3-TastingResults.xlsx*) and bottle technical sheets (*S4-BottleTechnicalSheets.pdf*) are available at the Journal website.

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