Angular Variations of Color in Turbid Media – the Influence of Bulk Scattering on Goniochromism in Paper

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Abstract

The angular variations of color of a set of paper samples are experimentally assessed using goniophotometric measurements. The corresponding simulations are done using a radiative transfer based simulation tool, thus considering only the contribution of bulk scattering to the reflectance. It is seen that measurements and simulations agree and display the same characteristics, with the lightness increasing and the chroma decreasing as the observation polar angle increases. The decrease in chroma is larger the more dye the paper contains. Based on previous results about anisotropic reflectance from turbid media these findings are explained. The relative reflectance in large polar angles of wavelengths with strong absorption is higher than that of wavelengths with low absorption. This leads to a loss of chroma and color information in these angles. The increase in lightness is a result of the anisotropy affecting all wavelengths equally, which is the case for transmitting media and obliquely incident illumination. The only case with no color variations of this kind is when a non-absorbing, non-transmitting medium is illuminated diffusely. The measured and simulated color differences are clearly large, and it is an open issue how angle resolved color should be handled in standard color calculations.

Introduction

The angular variation of color, or goniochromism, is well known within several fields of research. Metallic materials and paints containing metal-flake pigments display large angular variations in color [1, 2, 3, 4]. Pearlescent coatings display similar characteristics [5]. Angular variations of color are important also in the field of dentistry when matching the color appearance of a tooth restoration [6]. The present work concerns goniochromism of paper. To the authors' knowledge, previous work dealing explicitly with goniochromism of paper focuses exclusively on surface effects such as gloss [7]. In this work the focus is on the contribution of bulk scattering to goniochromism. As pointed out by Chirdon et al. [6] goniochromism is a property of most materials. Despite this, research is focused on studying the phenomenon in materials with complex diffraction and interference patterns, and little has been done to study goniochromism in bulk scattering materials. The paper samples studied in this work are prepared to minimize surface effects such as gloss, i.e. they have a rough surface. In this way the interaction between light and the samples is dominated by bulk scattering and absorption, and the samples can thus be considered as turbid media. Some attention has been given to angular variations of color in turbid media within the field of atmospheric physics [8].

The first purpose of this work is to measure the angle resolved color of a set of paper samples. Since the samples are dominated by bulk scattering and absorption, goniochromism of turbid media can in this way be experimentally assessed. The second purpose is to replicate the measured angular variations using an angle resolved light scattering model, DORT2002 (freely available from the authors). DORT2002 has been developed by Edström [9] and is adapted to light scattering simulations in paper and print. DORT2002 is based on radiative transfer theory [10] and implements a numerical solution of the equation of radiative transfer in plane-parallel turbid media.

By comparing measurements and simulations in this way it is possible to conclude if the goniochromism of the paper samples can be explained within the framework of radiative transfer theory. If measurements and simulations disagree this can be attributed to phenomena not included in the model, such as various surface effects. But if measurements and simulations agree the paper samples are indeed dominated by bulk scattering and absorption and can thus be adequately described by radiative transfer theory.

Neuman and Edström [11, 12] showed that angular variations, or anisotropy, of light reflected from turbid media are present in all situations encountered in practice, and that the variations depend on the relative contributions to the reflectance from different scattering depths in a medium. If near-surface bulk scattering dominates the reflectance, the light intensity is higher in large polar angles than in the normal direction of the medium. Near-surface scattering dominates in the case of strong absorption, high transmittance or obliquely incident illumination. This can be understood intuitively since the light is likely to be absorbed, transmitted or absorbed/scattered in these cases respectively when penetrating the medium further. It is the final purpose of this work to investigate if goniochromism of turbid media can be explained in a similar manner.

Method

Measurements

The angle resolved measurements are made with a spectral goniophotometer at Joensuu University, Finland. It has a halogen illumination of ceiling lighting type with an illuminated area much larger than what is viewed by the detector. The detector is a Hamamatsu PMA-11 C7473 fibre spectrophotometer with a spectral range of 380–780 nm and a spectral resolution of 2 nm. The measurement spot is a circle of 10 mm diameter at normal viewing angle.

The measurement device is spectrally calibrated using a matte white ceramic reference tile. The reference tile is measured with a standard spectrophotometer having 45/0 geometry [13], and the goniophotometer is calibrated by adjusting its readings for the same illumination and detection angles.

The samples are then measured in the goniophotometer with a directed illumination at 45° to the sample normal. The detector is moved from the normal direction in steps of 1° to 80° from the normal on the side opposite to the illumination. A thick black glossless paper is used as background in order to absorb transmitted light, thus minimizing boundary effects.

This procedure thus gives access to the angle resolved spectral reflectance factor.

Material

Five paper samples are used in this work. Paper sample 1 was produced on a small-scale experimental paper machine in order to resemble a wood-free commercial office paper, with fully comparable fiber composition, filler type, filler content, sizing and shading dye. Sample 1 has a grammage of approximately 90 g/m^2.

Paper samples 2-5 were prepared with a Formette Dynamique and vary in dye content. All samples were made from a mix of equal amounts of kraft and birch pulps, having Shopper/Riegler numbers 18° and 23° SR respectively. Sheets having approximate grammage 30 g/m^2 were prepared and all sheets contain 22 % filler. The amount of blue dye was changed from 0 to 1 % of the fiber weight in four steps, with sample 2 having the lowest dye content and sample 5 the highest. The dye used was Levacell Fast Blue KS-6GLL Liquid, manufactured by Lanxess. The dye used is a cationic direct dye and is assumed not to affect the structure of the paper. The sheets were dried in a cylinder dryer during 5 minutes at a temperature of 105° C and a pressure of 1.50 bar. Special care was taken when preparing the samples to minimize gloss. Gloss measurements were made with a Zehntner gloss meter to ensure low gloss levels, and the gloss was found to be low with an average of 1.0 for 20° and 2.9 for 60° and 75°. An overview of the paper samples is shown in Table 1.

Table 1. Paper samples

Sample	Dye content	Grammage(g/m^2)
1	zero	92.25
2	zero	31.80
3	low	31.69
4	medium	32.54
5	high	32.27

None of the samples used in this work contains fluorescent whitening agents (FWAs). This is essential since fluorescence would make spectral reflectance measurements and simulations far more complicated.

Simulations

The DORT2002 model developed by Edström [9] implements a numerical solution of the radiative transfer equation in plane-parallel turbid media. It has been used successfully in paper and print applications [14, 15, 16, 17] and is thoroughly evaluated [18, 19]. Radiative transfer theory describes the intensity of light at all positions and in all directions in a scattering and absorbing medium. The intensity is proportional to the reflectance factor [20]. The widely used Kubelka-Munk model [21] is a simplified case of general radiative transfer theory, and is unable to describe angular variations of light intensity. This has been investigated by Neuman and Edström in a previous publication [11].

The radiative transfer equation can be stated as

$$\frac{\mathrm{d}I(s,\theta,\varphi)}{\mathrm{d}s} = \sigma_e\left[-I(s,\theta,\varphi)+S\right], \tag{1}$$

where $I(s,\theta,\varphi)$ is intensity at depth s at polar angle θ and azimuthal angle φ. Here σ_e is the extinction coefficient and S is a source function. The extinction coefficient is the sum of the scattering and absorption coefficients σ_s and σ_a. The source function accounts for light scattered to θ,φ at depth s from all other angles. It can be written

$$S = \frac{a}{4\pi}\int_{4\pi} p(\cos\Theta)I(s,\theta,\varphi)\mathrm{d}\omega, \tag{2}$$

where a is the single scattering albedo defined as $a = \sigma_s/(\sigma_s+\sigma_a)$, ω is solid angle and $p(\cos\Theta)$ is the phase function describing the angular distribution of each single scattering. Here Θ is the angle between the directions of the incident and scattered light. A commonly used phase function is the Henyey-Greenstein phase function [22]. This phase function has a single parameter g, called the asymmetry factor, describing the angular distribution of the single scattering process. Isotropic single scattering is obtained when $g=0$ while $g=-1$ and $g=1$ give complete backward or forward scattering respectively. In the present work the Henyey-Greenstein phase function is used. Accurate spectral estimations of g are missing in the literature, but by comparing measurements and simulations the asymmetry factor $g=0.8$ is found to represent the measurement data reasonably well for all wavelengths.

To be able to do simulations for comparison with the goniophotometer measurements, numerical values of the scattering and absorption coefficients of the paper samples are necessary. These can be obtained by measuring the reflectance factor of the paper samples in the commonly used and standardized d/0 instrument geometry [23]. This instrument records the d/0 reflectance factor spectrally for wavelengths in the interval 400-700 nm in steps of 10 nm. Using this reflectance data the DORT2002 model can be employed to calculate the scattering and absorption coefficients σ_s and σ_a if the asymmetry factor and the grammage are provided. The grammage can be treated as equivalent to the thickness in this case [24]. The accurate determination of σ_s and σ_a is possible since DORT2002 can simulate the specific illumination and detection conditions of the instrument [17]. When doing spectral calculations, Eq. (1) has to be solved for each wavelength independently giving a set of medium parameters corresponding to the spectrum.

Results

The measured and simulated angle resolved reflectance factor spectra are translated to the $L^*a^*b^*$ color space using the D50 illuminant and 2° observer [25, 26]. Figs. 1, 2 and 3 show the measured and simulated $L^*a^*b^*$ values respectively for the different paper samples. It can be seen that the correspondence is good between measured and simulated data. In particular, the angular variations are present in both measurements and simulations. The L^* value shows a characteristic increase in both measurements and simulations when the detector angle increases. The a^* and b^* values show a similar increase when the samples contain dye, i.e. for samples 3-5. This means that the color in angles near the medium surface is perceived as lighter for all samples and as having less chroma for dyed samples. Since a^* and b^* seem to depend in the same way on observation angle the hue is unchanged when changing the observation angle.

Fig. 4 shows the measured and simulated CIE whiteness W for the non-dyed samples. It can be seen that the whiteness increases as the observation angle increases in the same characteristic way in both measurements and simulations. The correspondence in lightness L^* and whiteness W between measurements and simulations is best for sample 1, i.e. the non-dyed 90 g/m^2 sample. Samples 2-5 are translucent and the measurement background can influence the measurements, while the background in the simulations is a black cavity. This can affect the measurements and simulations differently.

The color change is further illustrated in Fig. 5 where the actual perceived color corresponding to the $L^*a^*b^*$ value is included. Fig. 6 shows how L^* and the chroma C^* defined as $C^* = \left[(a^*)^2+(b^*)^2\right]^{1/2}$ vary with observation angle, also with the perceived color indicated. Displaying the data in this way it can be easily seen that the lightness L^* increases for all samples as the observation angle increases, and that the chroma C^* decreases as the observation angle increases. The change in chroma is larger the more dye the sample contains.

The CIE 1976 color difference in the $L^*a^*b^*$ color space is denoted ΔE^*_{ab} [25] and defined as $\Delta E^*_{ab} = \left[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2\right]^{1/2}$. The maximum color difference for all samples when varying the observation angle is shown in Table 2. It can be seen that the color difference increases as the amount of dye increases. The smallest color difference is obtained for sample 1, the plain white paper. The measured and simulated values show the same tendency, but the measured difference is larger for the white samples while the simulated is larger for the dyed samples.

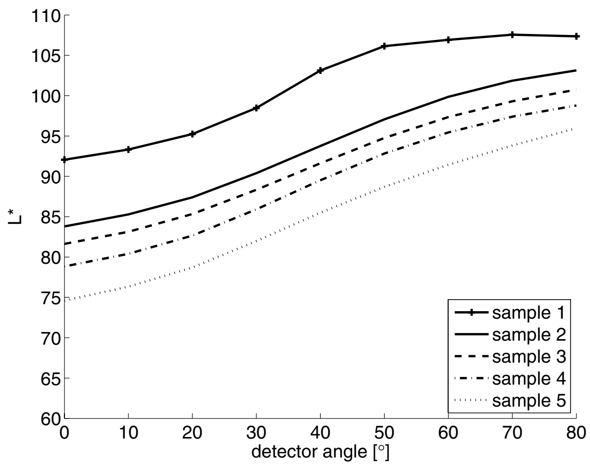
The color differences presented in Table 2 are clearly large. They are far above the limit of what is possible to perceive. However, it is an open issue if these values are directly comparable to the visual perception of the samples. The calculation of the $L^*a^*b^*$ values is performed according to a standardized procedure, but the experiments leading to the color matching functions were originally performed at constant angle of observation. To account for angle resolved observations it is possible that modifications of the standard be necessary. On the other hand, disregarding the magnitude of the differences, the characteristic behavior is plausible and can for example be observed when placing several identical paper sheets on a table and thus varying the observation angle. The paper sheet with the largest angle between the illumination and the direction of observation can then appear whiter. Furthermore, the dependence on dye content, i.e. the degree of absorption, is systematic and in agreement with previously published results [11, 12]. This is further discussed below.

Table 2. The maximum measured and simulated color difference for the paper samples when varying the observation angle.

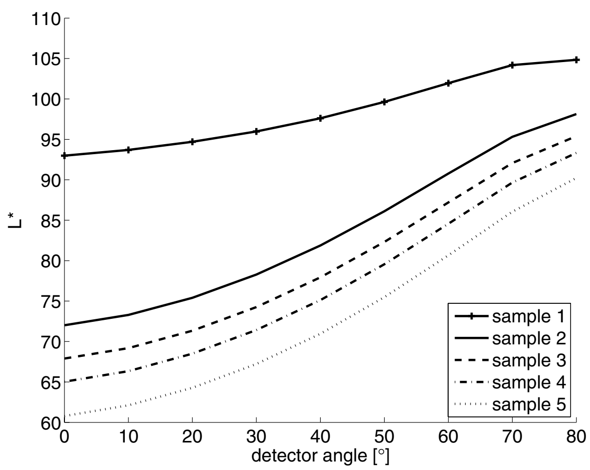
Sample	Max. meas. ΔE^*_{ab}	Max. sim. ΔE^*_{ab}
1	15.5	11.9
2	19.4	25.5
3	19.7	27.3
4	21.4	28.7
5	23.9	30.5

Simulation of Opaque Media

To eliminate the influence of the medium background on the angular color variations, and to further isolate the effect of ab-

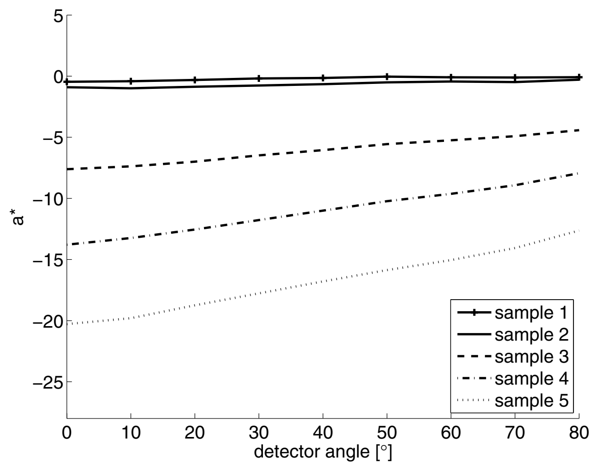


(a) Measured L^* value.

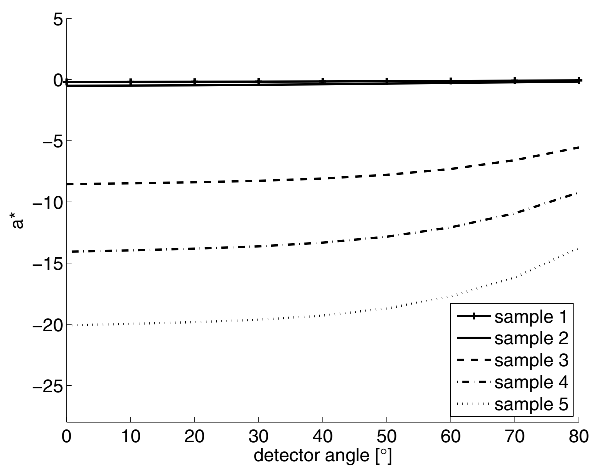


(b) Simulated L^* value.

Figure 1. *The measured and simulated L^* values for all paper samples.*



(a) Measured a^* value.



(b) Simulated a^* value.

Figure 2. *The measured and simulated a^* values for all paper samples.*

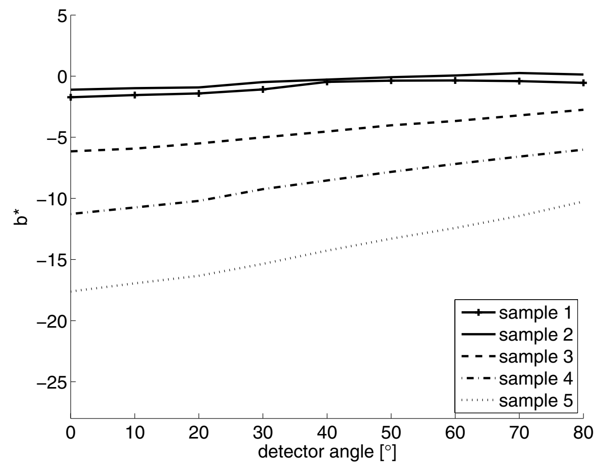
sorption, opaque media can be simulated using DORT2002. As previously, the medium scattering and absorption coefficients obtained from the d/0 measurement data are used, and the medium thickness is increased until the point of no transmittance. Fig. 7 shows how lightness and chroma of an opaque medium change with the angle of observation. It can be seen that the characteristic angular variations of lightness and chroma are present also in this case when the medium background has no influence. Comparing Figs. 6(b) and 7 it can be seen that the difference in color between the samples is smaller when there is no transmittance, since they overlap in the L^*C^* plot. It is interesting to note that the difference in chroma of the dyed samples when increasing the observation angle is larger for opaque media than for translucent media. Also, the lightness of the non-dyed samples varies less with observation angle if the medium is opaque.

Discussion

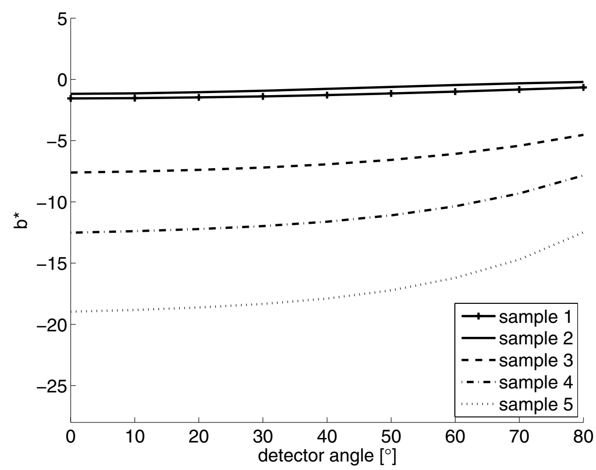
It has been shown in this work that the color of light reflected from paper has angular variations. This was shown by measuring the angle resolved reflectance of a set of paper samples with minimal surface effects such as gloss. By doing the corresponding simulations with the radiative transfer based DORT2002 model it was seen that the simulations display the same characteristic angular variations as the color of the samples. Since the agreement between measurements and simulations is good, a result of this work is that the papers used can be considered to be turbid media and that their interaction with light is accurately described by radiative transfer theory. This means that a large part of the light reflected from the papers comes from bulk scattering. Furthermore, this work showed that the color of turbid media decreases in chroma and increases in lightness when the angle of observation approaches the medium surface. This phenomenon was present in both measurements and simulations. These results hold for the media studied here and the particular measurement setup used, but the results can be generalized using knowledge about what causes the anisotropic reflectance of turbid media.

Explaining goniochromism in turbid media

Using the conclusions of Neuman and Edström [11, 12], angular variations of color in turbid media can be given a physically based explanation derived from first principles. Neuman and Edström showed that the relative reflectance in polar angles near the medium surface is increased when the amount of near-surface bulk scattering increases. This is the case when the medium is strongly absorbing or transmitting, or when the illumination is obliquely incident. In the case studied in the present work, the absorption varies between the samples and wavelengths since light of some wavelengths is absorbed more when dye is added to the paper. The light intensity is thus affected differently depending on the wavelength and reflected more or less anisotropically.

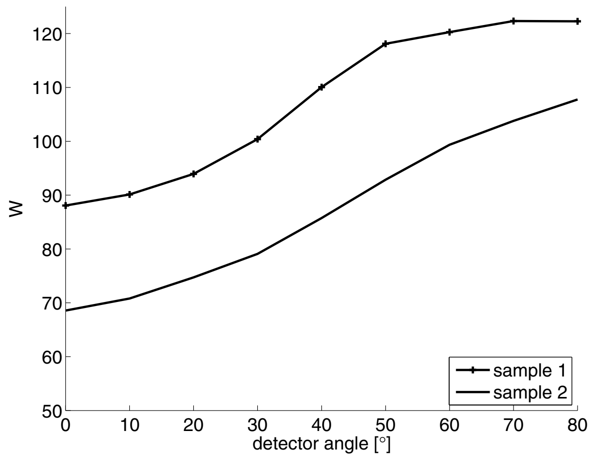


(a) Measured b^* value.

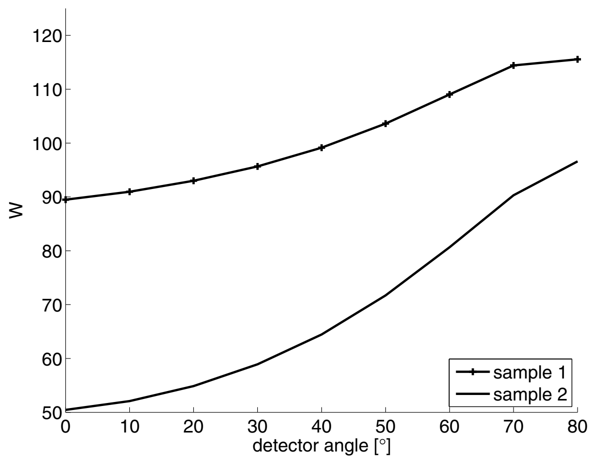


(b) Simulated b^* value.

Figure 3. *The measured and simulated b^* values for all paper samples.*



(a) Measured whiteness values.

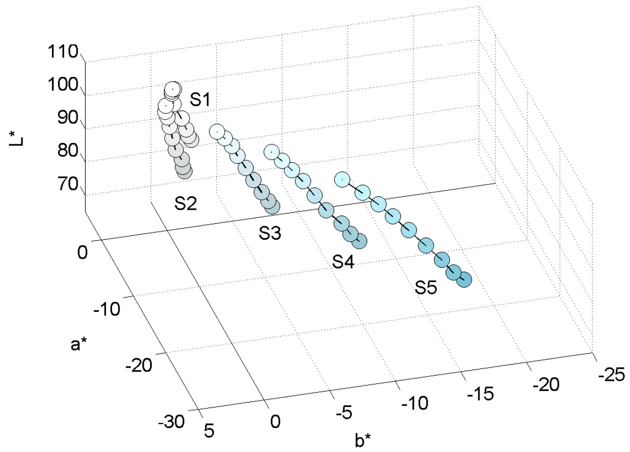


(b) Simulated whiteness values.

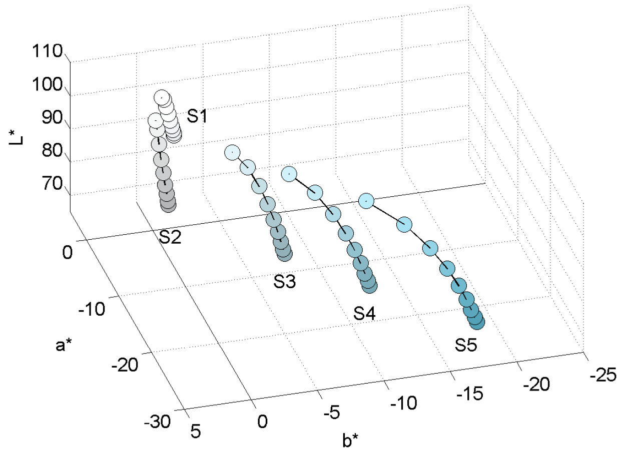
Figure 4. *The measured and simulated whiteness values for the paper samples with no dye.*

The relative reflectance in large polar angles is higher for wavelengths with strong absorption than for wavelengths with low absorption. Due to this the chroma decreases and color information is partly lost in these angles, because the difference between wavelengths becomes less pronounced. Lightness increases in large polar angles when light of all wavelengths has a higher relative reflectance in these angles. This is the case for transmitting media or when the illumination is obliquely incident, which affects all wavelengths equally. This is supported by the measurements presented in this work. For example, Fig. 6 shows that the increase in lightness is larger for the 30 g/m^2 sample than for the 90 g/m^2 sample. This is due to the higher transmittance of the 30 g/m^2 sample. Fig. 7 shows that the lightness increases also when there is no transmittance. This is because light of all wavelengths is absorbed to some extent, and primarily because the illumination is incident in 45° which causes light of all wavelengths to be reflected anisotropically with a higher relative reflectance in large polar angles. Diffuse illumination would give very small color variations in this case.

Several predictions about the angular variations of color in

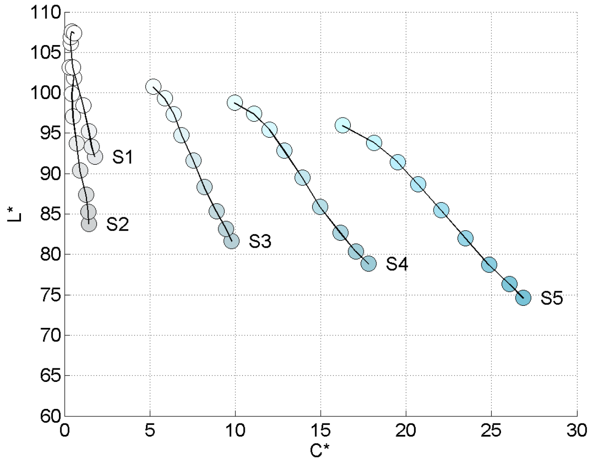


(a) Measured $L^*a^*b^*$ values.

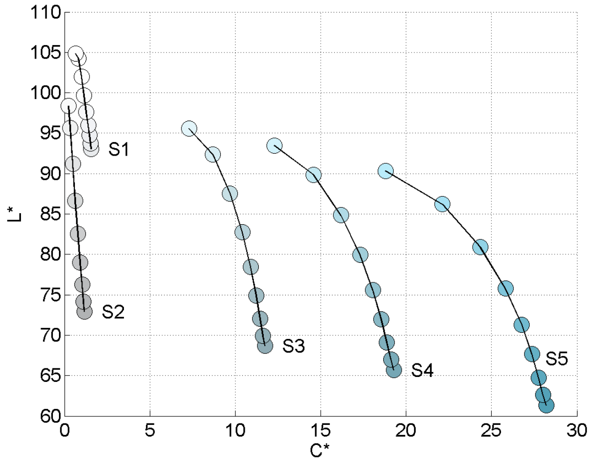


(b) Simulated $L^*a^*b^*$ values.

Figure 5. *The measured and simulated $L^*a^*b^*$ values for all paper samples for different directions of observation. Each dot corresponds to an observation angle and the color indicates the observed color. The observation angle ranges from 0° to 80° and increases when moving upwards and to the left along the line connecting the dots. S1-5 in the figures denote sample 1-5. It can be seen that the lightness increases for all samples and that the chroma decreases for the dyed samples when increasing the observation angle. This phenomenon is present in both measurements and simulations.*



(a) Measured L^* and C^* values.



(b) Simulated L^* and C^* values.

Figure 6. *The measured and simulated L^* and C^* values for all paper samples with the actual perceived color indicated. The observation angle ranges from polar angle 0° (right lower part) to 80° (upper left part). It can be seen that the lightness of all samples increases when the observation angle increases, and that the chroma decreases with observation angle. The change in chroma increases as the dye amount increases.*

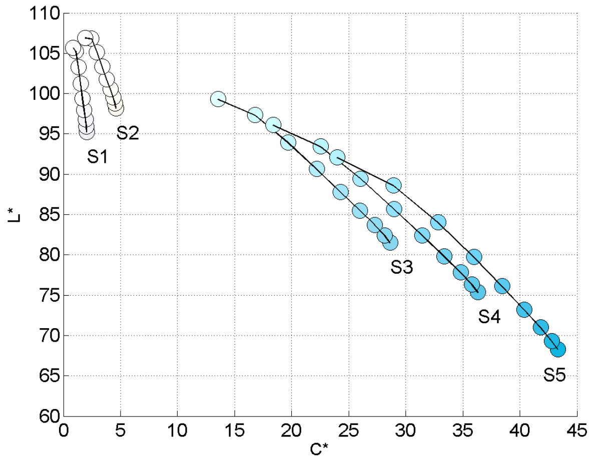


Figure 7. *Simulated L^* and C^* values for opaque samples with scattering and absorption coefficients corresponding to samples 1-5. It can be seen that the characteristic angular variations of lightness and chroma are present also when the medium background has no influence.*

turbid media can be made based on the conclusions of Neuman and Edström. Since the light is reflected anisotropically when there is absorption, transmittance or non-diffuse illumination, the only case with no angular variations of color of this kind is when a non-absorbing and non-transmitting medium is illuminated diffusely. This is an idealized situation never encountered in practice, which means that all media display angular variations of color.

Another type of anisotropy is obtained when the illumination is incident in the normal direction of the medium. In this case the reflectance is dominated by light scattered deeper inside the medium since the light on average penetrates the medium further. This results in anisotropy of the reflected light with more light being reflected around the normal direction, and the reflectance factor decreases in larger polar angles. This affects light of all wavelengths in the same way, so the chroma of the reflected light will be unaffected but the lightness will decrease in large polar angles.

These results can be summarized as

- Chroma and color information is lost in polar angles close to the medium surface. This is because the absorption varies between wavelengths, and light of wavelengths with strong absorption is reflected more anisotropically.
- Transmittance and equal absorption of all wavelengths leads to an increase in lightness in polar angles close to the medium surface. This is because light of all wavelengths is reflected with the same type of anisotropy.
- Obliquely incident illumination causes the lightness to increase in large polar angles.
- Normally incident illumination causes the lightness to decrease in large polar angles.

When there is a combination of dyed samples, transmittance and non-diffuse illumination, the angular variations of color will be determined by the relative influence of each of the factors.

Conclusions

This work has shown that angular color variations are indeed present in turbid media and that plain paper display these variations. This is potentially important for all applications where the perceived appearance of a material is of interest. An example is when comparing the whiteness of different papers. Whiteness is a desirable property of paper, but the results presented here show that the same paper can be perceived differently depending on how it is placed in relation to the light source, and that the transmittance influences the perceived appearance in a characteristic way. Also in the field of packaging this is important since the color appearance should normally not vary with the angle of observation.

The underlying mechanism of the color variations in turbid media was explained, and this knowledge can be used to optimize the color appearance in specific applications.

As pointed out in this work, the $L^*a^*b^*$ color space is not adapted to angle resolved observations of color since the experiments leading to the color matching functions were originally performed at constant angle of observation. It is a reasonable assumption that since the light intensity is higher in larger polar angles, the eye should adapt to this situation and the perceived color differences should be smaller than those presented here. It is still an open issue how to incorporate angular resolution in the calculation of $L^*a^*b^*$ values.

Future Work

The angular variation of color in paper can be further understood if the spectral dependence of the asymmetry factor g, describing the anisotropy of each single scattering, is known. This research activity is planned by the authors. Furthermore, by distinguishing surface phenomena such as gloss and micro roughness from bulk scattering and absorption, discrepancies between reflectance measurements and radiative transfer simulations can be understood.

A perception study testing the findings presented here could investigate the actual perceived color differences. Such a study could also investigate the validity of color calculations in angle resolved situations.

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