Abstract

Light scattering in turbid media is essential for such diverse application areas as paper and print, computer rendering, optical tomography, astrophysics and remote sensing. This thesis investigates angular variations of light reflected from plane-parallel turbid media using both mathematical models and reflectance measurements, and deals with several applications. The model of most widespread use in industry is the Kubelka-Munk model, which neglects angular variations in the reflected light. This thesis employs a numerical solution of the angle resolved radiative transfer problem to better understand how the angular variations are related to medium properties. It is found that the light is reflected anisotropically from all media encountered in practice, and that the angular variations depend on the medium absorption and transmittance and on the angular distribution of the incident light.

If near-surface bulk scattering dominates, as in strongly absorbing or highly transmitting media or obliquely illuminated media, relatively more light is reflected in large polar (grazing) angles. These results are confirmed by measurements using a set of paper samples. The only situation with isotropic reflectance is when a non-transmitting, non-absorbing medium is illuminated diffusely. This is the only situation where the Kubelka-Munk model is exactly valid. The results also show that there is no such thing as an ideal bulk scattering diffusor, and these findings can affect calibration and measurement procedures defined in international standards.

The implications of the presented results are studied for a set of applications including reflectance measurements, angle resolved color and point spreading. It is seen that differences in instrument detection and illumination geometry can result in measurement differences. The differences are small and if other sources of error — such as fluorescence and gloss — are not eliminated, the differences related to instrument geometry become difficult to discern. Furthermore, the angle resolved color of a set of paper samples is assessed both theoretically and experimentally. The chroma decreases and the lightness increases as the observation polar angle increases. The observed differences are clearly large, and it is an open issue how angle resolved color should be handled. Finally, the dependence of point spreading in turbid media on the medium parameters is studied. The asymmetry factor is varied while maintaining constant the optical response in a standardized measurement geometry. It is seen that the point spreading increases as forward scattering becomes more dominant, and that the effect is larger if the medium is low-absorbing with large mean free path. A generic model of point spreading must therefore capture the dependence on all of these medium parameters.

This thesis shows that turbid media reflect light anisotropically, and angle resolved radiative transfer models are therefore necessary to capture this. Using simplified models can introduce errors in an uncontrolled manner. The results presented potentially have consequences for all applications dealing with light scattering, some of which are studied here.
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This thesis consists of the following papers, herein referred to by their respective Roman numerals:


1 Introduction

The interaction between light and matter determines how we perceive an object. In a scattering and absorbing turbid medium, incident light can be absorbed and converted into other forms of energy, such as heat, or scattered one or several times and then exit the medium. The reflected light gives rise to our visual sensation and can also give important information about the light scattering properties of the medium, such as spectral reflectance, transmittance and opacity, and about color sensations such as whiteness. This thesis is about the assessment and interpretation of light reflectance and quantities derived from it, using both measurements and mathematical models.

Understanding how light interacts with matter and the connection to the resulting optical response is important in various industrial sectors. In the paper and print industries light scattering simulations and measurements are used when developing new products, for quality control in production, and in data exchange [1–3]. Furthermore, light scattering is important in the paint industry [4–6], textile industry [7, 8], food industry [9], and is the basis for computer rendering [10] and medical applications such as optical tomography [11]. Propagation of light in turbid media is also studied in research fields such as atmospheric physics [12] and astrophysics [13], and related problems are studied in, e.g., neutron transport [14] and remote sensing [15]. The results presented in this thesis are generic in the sense that they can be used in all applications dealing with light scattering.

1.1 Models of radiative transfer

There are many mathematical models describing radiative transfer (RT), i.e. light propagation in turbid media. The model of most widespread use in industry is the Kubelka–Munk (KM) model [16–18]. The KM model relates the light intensities $i$ and $j$ in two directions or solid angles to the KM scattering and absorption coefficients $s_{KM}$ and $k_{KM}$. The model can be stated as

$$\begin{align}
-\frac{di}{dx} &= -(s_{KM} + k_{KM})i + s_{KM}j \\
\frac{dj}{dx} &= -(s_{KM} + k_{KM})j + s_{KM}i
\end{align}$$

if $x$ denotes distance. In the paper industry the KM model is used together with standardized d/0 measurements [19] to determine the KM scattering and absorption coefficients, which is also a standardized procedure [20]. The paper samples are then illuminated diffusely and the detector is located at zero degrees, i.e. in the normal direction of the paper. The KM model is a simplification of more comprehensive models of radiative transfer, since it ignores all angular variations of the reflected light. Several limitations of the KM model have been reported, such as the dependence between the scattering and absorption coefficients [21–25], and several explanations have been proposed [26–30]. Angular variations of reflected light has
been observed or discussed by other authors [30–33], but not thoroughly or systematically studied.

General RT theory takes into account the angular dependence of the reflected light. It can be traced back to Lommel [34] via Chwolson [35], Schuster [36] and Chandrasekhar [13] among others. The RT equation for plane-parallel laterally homogeneous turbid media can be stated as

$$\frac{dI(s, \theta, \varphi)}{ds} = \sigma_e \left[-I(s, \theta, \varphi) + S\right],$$

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

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

where $a$ is the single scattering albedo defined as $a = \sigma_s/(\sigma_s + \sigma_a)$, $\omega$ is solid angle, $p(\cos \Theta)$ is the phase function and $\Theta$ is the angle between the directions of the incident and scattered light. The phase function describes the angular distribution of the single scattering process. A commonly used phase function is the Henyey–Greenstein phase function [37]. It can be written

$$p(\cos \Theta) = \frac{1 - g^2}{(1 + g^2 - 2g \cos \Theta)^{3/2}}$$

and it contains one parameter, the asymmetry factor $g$, which ranges from $-1$ to $1$ with $g = -1$ meaning complete back scattering, $g = 0$ isotropic scattering and $g = 1$ complete forward scattering. For organic materials such as paper it has been shown that $g$ has values in the interval 0.6–0.9 approximately [38–40], which means that forward scattering dominates.

The mathematics involved when solving the angle resolved RT problem (2) become far more complicated compared to when solving the KM equations (1), and analytical solutions to practically relevant problems cannot be found. Edström presented an implementation, called DORT2002, of a numerical solution of Eq. (2) [41]. The DORT2002 simulation tool is used extensively throughout this work. Monte Carlo methods can also be used to solve Eq. (2), but this is extremely time consuming and the solution contains noise. On the other hand, Monte Carlo methods pose less restrictions on the complexity level of the problem and can be used to solve, e.g., the 5-dimensional (laterally resolved) RT problem and RT in inhomogeneous media [42].

1.2 Purpose of the thesis

The general purpose of this thesis is to use the possibilities of angle resolved radiative transfer models to better understand angular variations in the light reflected
Radiative transfer and Maxwell’s equations

from plane-parallel turbid media. This will make clear how the angular variations depend on the medium parameters and shed new light on the relation between general RT theory and the KM model. Furthermore, the consequences of anisotropic reflectance for several applications will be studied. These include reflectance measurements, angle resolved color and point spreading.

2 Radiative transfer and Maxwell’s equations

The propagation of electromagnetic energy in space is described by Maxwell’s equations. When an object occupies some part of space, Maxwell’s equations have to be solved also inside the object with appropriate boundary conditions in order to obtain the electromagnetic field everywhere. The presence of the object thus alters the electromagnetic field. This is what is meant by electromagnetic scattering. When there are many scattering objects, i.e. when dealing with multi-particle systems, solving Maxwell’s equations becomes intractable and established numerical techniques fail. Therefore, one has to resort to approximate solution methods. Radiative transfer (RT) theory is one such approximate solution method.

The relation between radiative transfer theory and Maxwell’s equations has been unclear until recently, when Mishchenko derived the RT equation from Maxwell’s equations \[43–48\]. Mishchenko’s analysis is a justification of the use of radiative transfer theory to describe light scattering in multi-particle systems in general, and in the applications of this thesis in particular. The main steps in Mishchenko’s analysis are therefore presented here. This reveals the necessary conditions to be fulfilled when applying RT theory. Most details are omitted and the reader is referred to Mishchenko’s work \[43–48\] for a detailed analysis.

The Maxwell equations describing the scattering of a time-harmonic electromagnetic wave by an object occupying a region \(V_{\text{INT}}\) can be stated as

\[
\begin{align*}
\nabla \times \mathbf{E}(\mathbf{r}) & = i\omega \mu_0 \mathbf{H}(\mathbf{r}) \quad \text{,} \quad \mathbf{r} \in V_{\text{EXT}}, \\
\nabla \times \mathbf{H}(\mathbf{r}) & = -i\omega \varepsilon_1 \mathbf{E}(\mathbf{r}) \quad \text{,} \quad \mathbf{r} \in V_{\text{EXT}},
\end{align*}
\]

(5)

\[
\begin{align*}
\nabla \times \mathbf{E}(\mathbf{r}) & = i\omega \mu_0 \mathbf{H}(\mathbf{r}) \quad \text{,} \quad \mathbf{r} \in V_{\text{INT}}, \\
\nabla \times \mathbf{H}(\mathbf{r}) & = -i\omega \varepsilon_2 \mathbf{E}(\mathbf{r}) \quad \text{,} \quad \mathbf{r} \in V_{\text{INT}},
\end{align*}
\]

(6)

where \(\varepsilon\) is electric permittivity, \(\mu_0\) is the permeability of vacuum and \(V_{\text{INT}} \cup V_{\text{EXT}} = \mathbb{R}^3\). Here we have thus assumed that the scattering object and its surrounding are non-magnetic. Provided that we know the electric field \(\mathbf{E}(\mathbf{r})\) we can get the magnetic field \(\mathbf{H}(\mathbf{r})\) by calculating the curl of the electric field. We therefore choose to solve Eqs. (5) and (6) for the electric field. Rearranging Eqs. (5) and (6) and introducing the wave numbers \(k_1 = \omega(\varepsilon_1 \mu_0)^{1/2}\) and \(k_2 = \omega(\varepsilon_2 \mu_0)^{1/2}\) now gives

\[
\begin{align*}
\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - k_1^2 \mathbf{E}(\mathbf{r}) & = 0, \quad \mathbf{r} \in V_{\text{EXT}} \quad (7) \\
\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - k_2^2 \mathbf{E}(\mathbf{r}) & = 0, \quad \mathbf{r} \in V_{\text{INT}}. \quad (8)
\end{align*}
\]
The solution to these equations is given by

\[ E(r) = E^{\text{inc}}(r) + k_1^2 \int_{V_{\text{INT}}} d^3 r' \overset{\rightarrow}{G}(r, r') \cdot E'(r') \left[ m^2(r') - 1 \right], \tag{9} \]

where \( \overset{\rightarrow}{G}(r, r') \) is the free space dyadic Green’s function, \( m = k_2/k_1 \) is the relative refractive index and \( E^{\text{inc}}(r) = E(r) - E^{\text{ sca}}(r) \) is the electric field without the presence of the scattering object. The scattered field \( E^{\text{sca}}(r) \) is given by

\[ E^{\text{sca}}(r) = \int_{V_{\text{INT}}} d^3 r' \overset{\rightarrow}{G}(r, r') \cdot \int_{V_{\text{INT}}} d^3 r'' \overset{\rightarrow}{T}(r, r') \cdot E^{\text{inc}}(r''), \tag{10} \]

where \( \overset{\rightarrow}{T}(r, r') \) is the dyad transition operator. If we now apply the far-field condition \( k_1 r \gg 1 \), i.e. that \( r \) is much larger than the wavelength, and also assume that \( r \) is much larger than the size of the scattering object, we get

\[ E^{\text{sca}}(r) = \exp(ik_1 r) \frac{r}{r} E^{\text{sca}}_1(\hat{r}), \tag{11} \]

which is a spherical wave propagating in direction \( \hat{r} \) with scattering amplitude \( E^{\text{sca}}_1(\hat{r}) \).

We can treat multiple scattering if we consider \( N \) particles in \( V_{\text{INT}} \), each occupying a volume \( V_i \) such that \( \bigcup_{i=1}^{N} V_i = V_{\text{INT}} \). The electric field can now be written as

\[ E(r) = E^{\text{inc}}(r) + \sum_{i=1}^{N} \int_{V_i} d^3 r' \overset{\rightarrow}{G}(r, r') \cdot \int_{V_i} d^3 r'' \overset{\rightarrow}{T}_i(r, r') \cdot E_i(r''), \tag{12} \]

summing the contribution from all particles. The electric field \( E_i(r) \) excites particle \( i \) and is the resulting field from all other particles. It is given by

\[ E_i(r) = E^{\text{inc}}(r) + \sum_{j=1}^{N} \sum_{j \neq i} E^{\text{exc}}_{ij}(r), \tag{13} \]

where

\[ E^{\text{exc}}_{ij}(r) = \int_{V_j} d^3 r' G(r, r') \cdot \int_{V_i} d^3 r'' T_j(r, r') \cdot E_j(r''). \tag{14} \]

Equations (12)-(14) are called the Foldy-Lax equations. They describe the electric field resulting from scattering of \( N \) particles and follow from Maxwell’s equations without simplifying assumptions.

We can now simplify the Foldy-Lax equations if we apply the far-field condition so that all particles are assumed to be much larger than the wavelength and separated by distances also much larger than the wavelength. This means that each particle is excited by the other particles through spherical waves. Omitting details,
the total field can then be expressed as

\[ E(r) = E^{\text{inc}}(r) + \sum_{i=1}^{N} \frac{\exp(ik_1 r_i)}{r_i} \hat{A}_i(\hat{r}_i, \hat{r}^{\text{inc}}) \cdot E^{\text{inc}}(R_i) \]

\[ + \sum_{i=1}^{N} \frac{\exp(ik_1 r_i)}{r_i} \sum_{j=1}^{N} \hat{A}_i(\hat{r}_i, \hat{R}_{ij}) \cdot E_{ij}, \quad (15) \]

where \( \hat{A} \) is the scattering dyad, \( \hat{r}_i \) is directed from particle \( i \) towards the observation point, \( \hat{R}_{ij} \) is directed from particle \( j \) towards particle \( i \). The sums in Eq. (15) can be expanded and solved numerically, but in practice the problem becomes intractable for large \( N \).

By using the so-called Twersky approximation, that neglects scattering paths passing the same particle more than once, Eq. (15) can be simplified further.

The next step is to introduce the coherency dyadic \( \hat{C}(r) \) defined as

\[ \hat{C}(r) = \langle E(r, t) \otimes E^*(r, t) \rangle_t, \quad (16) \]

which is the time average of the dyadic product of the electric field and its conjugate. The time-harmonic factor is canceled in the coherency dyadic due to the multiplication with the conjugate. This is a desirable feature when looking for a measurable quantity, since otherwise an instrument would register no signal due to the time-harmonic oscillations giving a zero net result over a period of time. Furthermore, if ergodicity is assumed, averaging over time can be replaced by averaging over particle state and position. Equation (16) then becomes

\[ \hat{C}(r) = \langle E(r, t) \otimes E^*(r, t) \rangle_{\mathbf{R}, \xi}, \quad (17) \]

where \( \mathbf{R} \) denotes position and \( \xi \) state. The calculation of \( \hat{C}(r) \) can be simplified by the so-called ladder approximation, where scattering paths are allowed to have no more than one scattering particle in common. This approximation can be used if the number of particles is large. The resulting quantity is called the ladder coherence dyadic and is denoted \( \hat{C}_L(r) \). Furthermore, the ladder specific coherency dyadic \( \hat{\Sigma}_L(r, \hat{q}) \) is defined by

\[ \hat{C}_L(r) = \int_{4\pi} \hat{\Sigma}_L(r, \hat{q}) d\hat{q}, \quad (18) \]

This quantity satisfies an RT equation and is related to the specific intensity column vector \( \mathbf{I}(r, \hat{q}) = [\tilde{I}(r, \hat{q}), \tilde{Q}(r, \hat{q}), \tilde{U}(r, \hat{q}), \tilde{V}(r, \hat{q})]^T \), which also satisfies an RT equation. This RT equation can be written

\[ \hat{q} \cdot \nabla \mathbf{I}(r, \hat{q}) = -n_0 \langle K(\hat{q}) \rangle_\xi \mathbf{I}(r, \hat{q}) + n_0 \int_{4\pi} \langle Z(\hat{q}, \hat{q}') \rangle_\xi \mathbf{I}(r, \hat{q}') d\hat{q}', \quad (19) \]

where \( n_0 = N/V \) is the particle number density, \( \langle K(\hat{q}) \rangle_\xi \) is the single-particle extinction matrix averaged over particle states and \( \langle Z(\hat{q}, \hat{q}') \rangle_\xi \) is the single-particle phase matrix averaged over particle states.
In this thesis we solve the scalar version of the RT equation. To obtain this equation the specific intensity column vector is replaced by its first component, i.e. the specific intensity, and the phase and extinction matrices are replaced by their upper left elements. We thereby ignore polarization. Equation (19) can then be written

\[
\frac{dI(x, y, z; \theta, \varphi)}{ds} = -\sigma_e(x, y, z)I(x, y, z; \theta, \varphi) + \sigma_s(x, y, z) \int_4\pi p(\cos \Theta)I(x, y, z; \theta, \varphi)d\omega,
\]

adopting the notation of this thesis, and we see the similarity with Eq. (2). Denoting the integral term \( S \) and omitting variable dependence, Eq. (20) takes on the simpler form

\[
\frac{dI}{ds} = -\sigma_e I + S.
\]

To go from Maxwell’s equations (5)–(6) to the RT equation (21) we had to make a series of assumptions. These assumptions are listed below.

1. Non-magnetic material: \( \mu_0 = 0 \).
2. Far-field scattering: scattering particles and distances between the particles are much larger than the wavelength.
3. Twersky approximation: neglect scattering paths passing the same particle more than once.
4. Ergodicity: averaging over time is equivalent to averaging over state and position.
5. The number of particles is large: \( N \to \infty \)
6. Ladder approximation: scattering paths can have no more than one particle in common.
7. Ignore polarization.

When dealing with light scattering in paper, assumptions 1, 4 and 5 are obviously fulfilled. Assumptions 3 and 6 are noncontroversial in practice since their effect can be compensated for by the scattering coefficient.

Assumption 2 is reasonable for light scattering in paper. Components of several microns dominate the paper, and the mean free path is also in the order of several microns. Possible effects such as diffraction and interference are probably negligible since coherent light sources are normally not used in paper applications.

As mentioned previously, the KM model is widely used in industrial applications. For the KM model to be exactly valid we have to make further assumptions, as shown in Paper I in this thesis. These assumptions are

8. Non-absorbing medium: \( \sigma_a \to 0 \).
3 Some comments on terminology and notation

Terminology and notation related to light scattering differ somewhat depending on the application area. To avoid confusion, these matters are clarified in this section.

The intensity $I$ in RT theory is called radiance and denoted $L$ in radiometry [49]. The radiance is defined as

$$L = \frac{d^2 P}{\cos \theta dA d\omega}, \quad (22)$$

where $dP$ is radiant power, $dA$ is a surface element, $d\omega$ is a solid angle and $\theta$ is the angle between the normal of $dA$ and $d\omega$. The radiance has the unit $[W \cdot m^{-2} \cdot sr^{-1}]$. Another commonly used quantity is called flux and denoted $F$ in RT theory. It is called irradiance and denoted $E$ in radiometry. The irradiance at a point is the amount of energy per time incident on an infinitesimal surface containing the point. It has the unit $[W \cdot m^{-2}]$ and can be written

$$E = \frac{dP}{dA}. \quad (23)$$

The radiant exitance $M$ is the corresponding quantity for emittance, i.e., at a point it is the amount of energy per time emitted by an infinitesimal surface element containing the point.

A convenient and widely used construction for relating the irradiance to the radiance is the bidirectional reflectance-distribution function (BRDF), denoted $f_r$ [50]. Its general definition is

$$dL(\theta_i, \varphi_i, \theta_r, \varphi_r, E) = f_r(\theta_i, \varphi_i, \theta_r, \varphi_r)dE(\theta_i, \varphi_i), \quad (24)$$

and we see that it gives the reflected radiance as a function of irradiance and angles of incidence (subscript $i$) and reflection (subscript $r$). The BRDF $f_r(\theta_i, \varphi_i, \theta_r, \varphi_r)$ can be obtained by solving the RT equation, and the radiance can be obtained in a specific solid angle by integrating Eq. (24) over this solid angle. This is impossible in practice and a simpler version of the BRDF is often used to represent angle resolved reflectance data given that a certain illumination is used. That is, we are not interested in a BRDF containing information about how changing the illumination affects
the reflected light. This would then correspond to another measurement or simulation setup. We can therefore remove the dependence on \( \theta_i, \varphi_i \) in the BRDF and integrate Eq. (24) over \( \theta_i, \varphi_i \) containing all incident light. Furthermore, if we assume that all measurements are done in-plane, so that we get no dependence on \( \varphi_r \) (which is equivalent to assuming azimuthal symmetry), Eq. (24) becomes

\[
L(\theta_r) = f_r(\theta_r)E. \tag{25}
\]

It follows from Mishchenko’s work [43–48] that the radiance can be measured with a well-collimated radiometer, and we can therefore assess also the BRDF experimentally through Eq. (25) by careful calibration of the measurement instrument.

4 Summary of the papers and contribution of the thesis

The papers included in this thesis all use radiative transfer models to investigate various aspects of angle resolved light scattering. The main contribution of the thesis is new knowledge about the interaction of light with plain-parallel turbid media through the application of these models. This is relevant for virtually any area where light scattering is an important part. The application areas in this thesis include reflectance measurements, angle resolved color and point spreading.

Papers I-II deal with anisotropic reflectance from plane-parallel turbid media, both from a theoretical and an experimental point of view. Paper III analyzes the consequences of anisotropic reflectance for the d/0 and 45/0 instrument geometries. Paper IV shows how the angular variations of light from scattering media result in corresponding angular variations of color. Paper V investigates how point spreading in turbid media is related to the medium parameters and shows that the single scattering anisotropy must be taken into account in a model for point spreading.

4.1 Paper I

Paper I employs the DORT2002 radiative transfer model to show that the light reflected from turbid media is anisotropic in all cases encountered in practice and that the anisotropy depends on the medium properties in a characteristic way. This is illustrated by displaying the simulated BRDF of three media. These show that the BRDF is close to isotropic when the medium is low-absorbing and opaque, while it is anisotropic when there is considerable absorption or transmittance. To understand this, simple expressions are stated showing that the angular variations of the reflectance depend on the relative contribution from different scattering depths in the medium. When near-surface bulk scattering increases, the amount of light reflected in large polar angles increases. This is the case for optically thin or strongly absorbing media, or when the illumination is incident obliquely. The illumination dependence is predicted by theory and verified by simulation in this paper. These
results also lead to the conclusion that there exists no bulk scattering ideal diffusor. If the light scattering properties are ideal it will still reflect anisotropically depending on the illumination. It reflects isotropically only when illuminated diffusely. This has consequences for the wide spread Kubelka-Munk model. It is shown that the Kubelka-Munk model is exactly valid only under very specific conditions. These are zero transmittance, zero absorption and diffuse illumination. Neglecting the angular variations in the reflected light introduces errors that can be 20-40 %, depending on the medium parameters. It is also pointed out that the error introduced when applying the Kubelka-Munk model can be minimized. For example, by placing the detector in an optimal angle in a measurement setup.

Paper I was written in cooperation with Prof. Per Edström. The contribution of the author of this thesis was to perform simulations, to do calculations and main part of the reasoning and writing.

4.2 Paper II

Paper II confirms experimentally the theoretical results from Paper I. The angle resolved reflectance from a set of paper samples varying in dye and filler content and thickness is measured using a goniophotometer. The addition of dye and fillers alters the scattering and absorbing properties of the samples so that their influence on the angle resolved reflectance can be studied. Corresponding simulations are also done using DORT2002 and it is seen that the agreement between measurements and simulations is good. Furthermore, the angle resolved reflectance of a BaSO₄ diffusor is measured and simulated with varying angle of incidence of the illumination. It is seen that the agreement between experiment and theory is good, and that the diffusor is not ideal. Paper II also discusses how anisotropy of the reflected light can affect standardized measurements through e.g. the calibration routine which often involves alleged ideal diffusors.

Paper II was written in cooperation with Prof. Per Edström. The contribution of the author of this thesis was to do measurements, simulations, and main part of the reasoning and writing.

4.3 Paper III

Paper III investigates differences in measured reflectance factor between the 45/0 and d/0 instrument geometries. The effect of external factors, such as sample background, calibration and sample inhomogeneity, on the reflectance measurements is quantified and a method for eliminating them is suggested. The remaining instrument differences can be attributed to the instrument geometry. The 45/0 and d/0 reflectance factors are measured and compared for a set of paper samples. The instruments are also simulated using DORT2002 and it is seen that the inter-instrument differences are of the same magnitude as in the measurements. The inter-instrument differences are small and around 0.1 ΔE*ab for the set of paper samples. An explanation is proposed for the geometry induced inter-instrument differences where results...
from Papers I and II are used. The mean scattering depth in the 45/0 instrument is larger than in the d/0 instrument. This means that the light reflected from a medium with the respective illuminations will differ in angular dependence. Relatively more light will be reflected towards the detector with the 45/0 illumination if the medium is low absorbing, highly scattering and opaque. The d/0 instrument will on the other hand detect a higher reflectance if the medium is strongly absorbing or transmitting, since then more light is reflected with the d/0 illuminations than with the 45/0 illumination.

Paper III was written in cooperation with Prof. Per Edström, M.Sc. Stefanos Avramidis and Dr. Mattias Andersson. The contribution of the author of this thesis was to do part of the writing and reasoning behind the proposed explanation.

4.4 Paper IV

Paper IV deals with the consequences of the findings in Papers I and II for angle resolved color. The same set of paper samples is used here as in Paper I and II, and an additional thicker paper. The angle resolved spectral reflectance factor of the samples is measured with a goniophotometer at InFotonic Center, Joensuu. The medium scattering and absorption coefficients are assessed using d/0 measurements and assuming that $g = 0.8$. In this way the goniophotometer measurements can be simulated using DORT2002 since we then have the necessary parameter setup. The reflectance factor increases as the polar angle increases in both measurements and simulations. Simulations and measurements agree well. The relative increase is larger for wavelengths in the absorption band of the dye. This is agreement with the results of Papers I and II, and the characteristics of the anisotropy are further confirmed by these result. Note that the samples are prepared in such a way to minimize gloss and that the simulations contain no surface scattering. These effects are therefore a result of bulk scattering, and not of surface scattering. The measured and simulated angle resolved reflectance factor spectra are then converted to the $L^*a^*b^*$ color space using the D50 illuminant and $2^\circ$ observer. It is seen that the angular variations in reflectance factor give rise to strikingly large variations in the angle resolved $L^*\ a^*\ b^*$ values. The maximum CIE 1976 color difference $\Delta E_{ab}^*$ is obtained for the heavily dyed paper. It is 23.9 for the measurements and 30.5 for the simulations. This is far above the limit of what is possible to perceive. It is suggested that it is reasonable that the eye can adapt to the angle resolved observation conditions so that the perceived differences are smaller and more realistic. It is still an open issue how to incorporate angular resolution in the calculation of the $L^*a^*b^*$ values.

A result of the simulations and the measurements is that the lightness increases and the chroma decreases when the observation polar angle is increased. An explanation is proposed for this based on the findings in Papers I and II. The lightness increases since the light of all wavelengths is reflected anisotropically with more light in large polar angles. The chroma decreases since light of wavelengths that is absorbed by the dye is reflected even more anisotropically from the medium, with a larger relative increase in large polar angles.
Summary of the papers and contribution of the thesis

4.5 Paper V

Paper V concerns point spreading in turbid media. The same set of dyed paper samples as in Paper II and IV is used to assess relevant medium properties describing the scattering characteristics. The d/0 reflectance factor is measured and the medium properties albedo and mean free path are calculated using DORT2002 for a set of values of the asymmetry factor. In this way the optical response in the d/0 instrument will be the same irrespective of the value of the asymmetry factor. The point spreading is then simulated for all media using the Monte Carlo model Open PaperOpt [42]. A numerical measure is introduced to quantify point spreading. It is seen that the point spreading measure increases linearly with the asymmetry factor for both opaque and thin media. It is largest for opaque media with high albedo and large mean free path. The number of scattering events that the wave packets undergo before leaving the medium is also calculated using the Monte Carlo model. Increasing the asymmetry factor increases the number of scattering events. Media with high albedo have a similar contribution to the reflectance from different scattering orders. Media with low albedo are also similar in this respect. This means that the point spreading is determined by the distance that the multiply scattered light can travel. This distance is larger if the mean free path is large and absorption is low. A medium with high albedo and large mean free path will thus give the largest point spreading, and it will increase further if light is scattered more in the forward direction.

The presented results thus show that the asymmetry factor plays a significant role in point spreading together with the albedo, mean free path and thickness. This means that models of point spreading must take all of these medium characteristics into account in order not to be based on ad hoc assumptions.

Paper V was written in cooperation with L.Sc. Ludovic Coppel and Prof. Per Edström. The contribution of the author of this thesis was to perform simulations and measurements, to do calculations and main part of the reasoning and writing.
5 Discussion

We have seen that the light reflected from turbid media has angular variations in all situations encountered in practice. Angle resolved radiative transfer theory is necessary to describe these variations. Paper I showed that the variations depend on the angular distribution of the incident light and the medium absorption and transmittance. Varying these factors will alter the mean scattering depth in the medium and thereby the angular distribution of the reflected light. Furthermore, we saw in Paper II that the theoretical predictions could be confirmed experimentally through goniophotometric measurements of light reflectance from paper samples.

These findings can potentially have a large impact on many applications dealing with measurements of scattered light. For example, we showed that a non-transmitting, low-absorbing medium, i.e. what is normally thought of as an ideal diffusor, reflects light anisotropically depending on the angular distribution of the illumination. This type of medium, such as a BaSO$_4$ plate, is often used for calibration purposes, and neglecting angular variations can introduce errors. It should be mentioned though, that the error can cancel to some extent in some cases, for example if measurements are done to compare samples. But it is still an uncomfortable situation since this is not controlled.

Paper III showed that this kind of anisotropic light reflectance will result in different instrument readings in instruments with different illumination and detection geometries. Other sources of error in reflectance measurements, such as fluorescence, gloss and different backgrounds, can give far larger differences between instruments. This means that if these other sources of error are not carefully eliminated, the error due to instrument geometry will be difficult to discern. If the measurements are perfect in all other respects, the instrument geometry can have a considerable impact on the measurement results.

Angle resolved color was investigated in Paper IV and the results presented posed several new questions. The measured and simulated differences in color when varying the observation angle were clearly large – larger than what is perceived by the human eye. This means that there must probably be an adaptation step not currently included in the calculation of the $L^*a^*b^*$ color coordinates. This is being investigated by the authors of Paper IV, and will hopefully result in new guidelines on how to handle angle resolved color. The magnitude of the color differences will then probably be smaller, but the characteristic effects (decreased chroma and increased lightness) observed in Paper IV will probably remain. This aspect of anisotropic light reflectance can lead to several new investigations since the color sensation is of such great importance in many applications.

Paper V discusses point spreading in turbid media, and is an example of how results concerning light scattering are important for such diverse fields as halftone image reproduction, light propagation in tissues and computer rendering. We saw that assumptions about the single scattering anisotropy can greatly alter the average lateral distance that incident light travels before exiting the medium, but still give the same optical response in a specific measurement geometry. This illustrates why
angle resolved models and measurements are crucial for a full understanding of light scattering in turbid media. It also shows that model simplification can be a risky business since important phenomena can be ignored and since the simplified model can be used by others, unaware of its limitations.

6 Future work

As mentioned above, it is still an open issue how to handle angle resolved color. Work on this is ongoing and will be presented shortly. The results on anisotropy can partly explain some of the reported shortcomings of the Kubelka-Munk model, such as the inter-dependence of the model parameters. This will be reported by the author elsewhere. Work is currently being done by the author to include fluorescence in general radiative transfer theory. This will give new insight into the effect of fluorescence on anisotropy and phenomena related to it, and will open up possibilities to optimize for example the paper composition for increased whiteness. The parameter estimation problem can be studied further to determine the spectral asymmetry factor \( g \) by solving the inverse RT problem to match goniophotometric measurements. This will give further understanding of light scattering and possibilities to optimize the appearance of light scattering media. Precise goniophotometric measurements also open up the possibility to separate the bulk and surface contributions to the light reflectance, for example when dealing with glossy paper samples. The results presented in this thesis can be used to give guidelines on how to construct instruments that can be used together with radiative transfer theory to better understand the optical response of turbid media.

7 Conclusions

This thesis shows that the light reflected from turbid media is anisotropic in all situations encountered in practice. This can potentially have consequences for all applications dealing with light scattering, including such diverse areas as paper and print, computer rendering, optical tomography and atmospheric physics.

We saw that the angular variations of light reflected from turbid media depend on the medium parameters in a characteristic way. Radiative transfer models have to be angle resolved to capture this dependence. Simplified models that ignore angular variations, such as the Kubelka-Munk model, are widely used in industry. Applying these models to, e.g., reflectance measurements can introduce errors. The magnitude of the error depends on the particular situation and the medium properties, but it is significant for a large range of parameters. Using angle resolved radiative transfer models relieves this uncomfortable situation and opens up for a better understanding of light reflectance from turbid media. The specific instrument geometry can be taken into account and the model parameters are physically objective, i.e. they can be given a meaning outside the model. The explanatory power of the model-measurement compound is therefore far greater in this case compared to when using
simplified models. Furthermore, angle resolved models can give important guidelines when designing new instrument geometries. The instrument geometry can be made optimal in some sense, for example by minimizing the geometry induced error for a given set of medium parameters. Angle resolved radiative transfer also has the advantage of being related to fundamental physical principles, in this case Maxwell’s equations.

The impact of the results presented here on the variety of applications of light scattering is largely unexplored. We saw here that, apart from reflectance measurements, angle resolved color and point spreading is better understood using angle resolved radiative transfer. The color appearance of a light scattering material is of great importance for example in the paper, print and packaging industries. A correct understanding of point spreading is essential, for example, to compensate for optical dot gain in halftone prints and to render realistic computer graphics.

References


Paper I
Anisotropic reflectance from turbid media.

I. Theory

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It is shown that the intensity of light reflected from plane-parallel turbid media is anisotropic in all situations encountered in practice. The anisotropy, in the form of higher intensity at large polar angles, increases when the amount of near-surface bulk scattering is increased, which dominates in optically thin and highly absorbing media. The only situation with isotropic intensity is when a non-absorbing infinitely thick medium is illuminated diffusely. This is the only case where the Kubelka–Munk model gives exact results and there exists an exact translation between Kubelka–Munk and general radiative transfer. This also means that a bulk scattering perfect diffusor does not exist. Angle-resolved models are thus crucial for a correct understanding of light scattering in turbid media. The results are derived using simulations and analytical calculations. It is also shown that there exists an optimal angle for directional detection that minimizes the error introduced when using the Kubelka–Munk model to interpret reflectance measurements with diffuse illumination. © 2010 Optical Society of America

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1. INTRODUCTION

Propagation of light in turbid media, or radiative transfer, has been studied throughout the last century. Radiative transfer theory is applied in a variety of fields such as astrophysics, atmospheric physics and neutron diffusion. It is also used in various industrial contexts as for example paper, printing, graphics, textiles, and the plastics industry.

Early attempts to develop a theory for radiative transfer include Schuster [1] and Kubelka and Munk [2–4]. They proposed two-flux models that consider the radiation, or intensity, in two directions only. The general problem of radiative transfer is described, for example, by Chandrasekhar [5], and efficient numerical solution methods exist, as presented by, e.g., Edström [6]. As opposed to the early intents to present a theory of radiative transfer, the theory now describes the intensity at all locations and in all directions. Modern models thus capture angular variations, or the anisotropy, of the intensity, while previous ones do not. The present work deals with the anisotropy of the intensity when considering plane-parallel media, and it is the purpose to show that anisotropy is present in almost all situations, and that low-resolution models are insufficient for a full understanding of light scattering in turbid media.

Schuster acknowledged the importance of angular variations in the light distribution within the medium, but refrained from investigating the matter further because of its complexity. Other authors have discussed or observed anisotropy and mentioned its potential importance [7–10]. The model developed by Kubelka and Munk (hereafter KM model) does not take angular variations into account. Kubelka [3] claimed their model to be exact when the incident light is diffuse (isotropic), or when collimated illumination incident at 60° from the normal is used. Kubelka argued that the mean path in the medium is proportional to the vertical distance from the medium surface, which is the measure of extension in the KM model, under these circumstances. The KM model assumes isotropic single scattering, and the reasoning is based on the belief that this together with the specified illumination conditions leads to an isotropic intensity throughout the medium.

Several problems with the KM model have been reported, as it is widely used in industrial applications. One example is the dependence between the model parameters [11–15], and explanations have been proposed attributing the behavior to model limitations [7,16–19]. The disagreement between the KM model and general radiative transfer theory has been investigated by Edström [20], who noted that it is most noticeable for optically thin and highly absorbing media. The KM model thus gives incorrect results even where it is generally believed to give exact results.

The purpose of this work is to investigate anisotropy and to explain how it arises in plane-parallel turbid media. Also, the KM model validity is studied by recovering the KM model from general radiative transfer theory using the necessary assumptions, and the error introduced when applying the KM model is investigated. While this paper has a theoretical focus, part II of this work [21] is more applied, dealing with experimental verification of the results and measurement related issues concerning anisotropy.
2. THEORIES OF RADIATIVE TRANSFER

A. General Radiative Transfer

The problem of radiative transfer concerns calculating the radiation field in a turbid medium that absorbs, scatters, and emits radiation. The quantity of interest is the intensity $I$ defined as

$$ I = \frac{\text{d}E}{\cos \theta \text{d}\Omega \text{d}d} $$

(1)

This is thus the flow of energy $\text{d}E/\text{d}t$ through the area $\text{d}a$ within the solid angle $\text{d}\Omega$, where $\theta$ is the angle between $\text{d}a$ and $\text{d}\Omega$. The intensity can represent a set of photons traveling along the same line and is also referred to as light or radiation in this work. Equation (1) holds for all wavelengths $\nu$ but this is not stated explicitly here. If the intensity $I$ travels a distance $\text{d}x$ it will be reduced by

$$ \text{d}I = -\sigma_I \text{d}x, $$

(2)

where $\sigma_I$ is the extinction coefficient. The extinction can be due to absorption or scattering, and the extinction coefficient can thus be divided into two parts:

$$ \sigma_I = \sigma_a + \sigma_s, $$

(3)

where $\sigma_a$ and $\sigma_s$ are the absorption and scattering coefficients. The single scattering albedo $\omega$ is defined as

$$ \omega = \frac{\sigma_s}{\sigma_a + \sigma_s}, $$

(4)

and the optical thickness $\tau$ as

$$ \tau = \omega \delta, $$

(5)

The albedo and the optical thickness are dimensionless measures of the medium’s characteristics.

The contribution to the intensity from scattering is accounted for by the source function $S$ defined as

$$ S = \frac{\omega}{4\pi} \int_{4\pi} p(\cos \theta) \text{d}w, $$

(6)

where $p(\cos \theta)$ is the phase function that describes the probability for scattering at an angle $\theta$ from the direction of incidence. Adding this source term to Eq. (2) gives the equation of radiative transfer:

$$ \frac{\text{d}I}{\text{d}x} = \sigma_s(-I + S). $$

(7)

To account for other phenomena, such as for example fluorescence, an additional term has to be included in Eq. (6). This is not within the scope of the present work.

B. Kubelka–Munk Model

The KM model reduces the problem of radiative transfer to light traveling in two directions in a homogeneous medium characterized by the KM scattering and absorption coefficients, $s_{KM}$ and $k_{KM}$. If light of intensity $i$ travels a distance $\text{d}x$, the intensity will be reduced by $(s_{KM} + k_{KM})\text{d}x$. The amount scattered will reappear as intensity in the other direction, $j$. The same conditions hold for this intensity. This gives the following equations for the changes in intensity:

$$ \frac{\text{d}i}{\text{d}x} = -(s_{KM} + k_{KM})i + s_{KM}j, $$

$$ \frac{\text{d}j}{\text{d}x} = -(s_{KM} + k_{KM})j + s_{KM}i, $$

(8)

where $i$ is the intensity in the downward direction and $j$ in the upward.

3. ANISOTROPIC REFLECTANCE FROM TURBID MEDIA

A. DORT2002 Simulations

To investigate the anisotropy of light reflected from plane-parallel turbid media, the simulation tool DORT2002 developed by Edström [8] is employed. (The DORT2002 software is freely available from the authors.) This tool implements a numerical solution of Eq. (7). To illustrate the phenomena under consideration here, the single scattering albedo and optical thickness of the medium are assigned numerical values. To exclude anisotropy originating from anisotropic single scattering the phase function $p(\cos \theta) = 1$ is used.

In Fig. 1 the distribution of the reflected light, the bi-directional reflectance distribution function (BRDF), is calculated for three media. The BRDF is proportional to the intensity $I$ [22]. isotropic illumination is used, and the supposed validity conditions of the KM model are thus fulfilled. The first medium in Fig. 1a is a low absorbing thick medium, and it can be seen that the reflected light is close to isotropic. In Fig. 1b the BRDF for a low absorbing thin medium is shown. It can be seen that the light distribution is highly anisotropic in this case, with more light being reflected at larger polar angles. Similarly, for the highly absorbing thick medium in Fig. 1c, the BRDF is also highly anisotropic with more light being reflected at larger polar angles. It is thus observed that increasing the absorption or the transmittance increases the anisotropy in this sense.

![Fig. 1. Three-dimensional BRDF for three media. The single scattering albedo is denoted by $\omega$ and the optical thickness by $\tau$. The supposed validity conditions of the KM model are used; that is, isotropic illumination and isotropic single scattering. A shaded half sphere is included for reference. The resulting intensity is anisotropic, since more light is reflected into larger polar angles for (b) thin and (c) highly absorbing media.](image-url)
B. Analytical Calculations

In order to determine what causes the anisotropic reflectance, simple analytical relations can be stated.

If light is scattered at a depth $d$ inside a medium with extinction coefficient $\sigma$, the probability for the light to exit the medium without further scattering is

$$\int_{0}^{\infty} \sigma_x \exp(-\sigma x) dx = \exp(-\sigma d / u),$$

(9)

where $\sigma_x \exp(-\sigma x)$ is the path length probability density and $u = \cos \theta$ with $\theta$ the exit angle. This type of expression is often denoted Beer’s law.

It can be seen that the probability depends strongly on the exit angle $\theta$ and decays with $\theta$. This is shown in Fig. 2 where the probability for an exit direction for different scattering depths is illustrated schematically. It can be seen that the distance to the surface strongly affects the angular distribution of the reflected light. A larger distance to the surface causes the relative amount of radiation exiting the medium at larger polar angles to decrease. Thus, the presence of near-surface bulk scattering alters the angular distribution of the reflected light.

Taking into account the distribution of the depth of the first scattering event gives

$$\int_{0}^{\infty} \int_{x}^{\infty} \sigma_x \exp(-\sigma x_1) \sigma_x \exp(-\sigma x_2) dx_1 dx_2 = \frac{1}{1+1/u},$$

(10)

The situation treated is illustrated in Fig. 3 and the expression holds for an infinitely thick medium, i.e., when transmittance is not present, with normally incident light.

Equation (10) depends on exit angle only, while Eq. (9) depends on optical thickness and exit angle. This is a consequence of introducing a length scale in Eq. (9) when specifying a scattering depth. Equation (10) says that, independently of the characteristics of an infinitely thick medium, the probability for normally incident light to be reflected from the first scattering event decays with exit angle as $1/(1+1/u)$. This can be understood intuitively since the same extinction coefficient governs both the probability of first scattering and the exit path of the light.

Equations (9) and (10) state a fundamental result. They show that angular variations in the reflectance must depend on the relative contributions from different depths in the medium. This is the only behavior that distinguishes different media, if the reflectance is dominated by light originating from a certain depth, its angular distribution will be determined by this light. That is, if near-surface bulk scattering dominates, the angular amount of light reflected into larger polar angles will increase, when compared to media with other depths dominating. The intensity $I$ is thus anisotropic if scattering from a certain depth dominates the reflectance. This holds no matter the order of the scattering. The crucial factor is the starting depth of photons exiting the medium. Changing the angle of incidence can change the relative contribution to the reflectance from different depths, but the result stated still holds.

C. Conclusions about Anisotropy

What causes the anisotropy can be understood in the light of the result obtained from Eqs. (9) and (10). In a low absorbing thin medium, near-surface bulk scattering will dominate, since light penetrating the medium further has a high probability of being transmitted. Similarly, in a highly absorbing thick medium, near-surface bulk scattering will dominate since the light has a high probability of being absorbed when penetrating the medium further. Thus, transmission and absorption are two characteristics determining the anisotropy, since they govern the relative amount of near-surface bulk scattering. Two extreme cases are a thick non-absorbing medium, giving close to isotropic reflectance, and a thin, highly absorbing one reflecting most anisotropically.

A third factor that affects the relative amount of near-surface bulk scattering is the illumination. If the illumination is incident near the normal direction, the light will on average penetrate the medium further, thus causing the first scattering to occur deeper inside the medium. If the illumination is incident obliquely the light will on average scatter closer to the surface. This is shown in Fig. 4 where the BRDF of an infinitely thick non-absorbing medium is calculated with DORT2002 for two types of illumination. This medium is chosen in order to eliminate anisotropy originating from medium characteristics. It can be seen that the medium reflects isotropically if the illumination is isotropic, and that the reflectance decreases for larger polar angles when normally incident illumination is used, i.e., it is anisotropic in the way predicted here. In fact, the situation shown in Fig. 4(a) is the only case giving a perfectly isotropic intensity. This is realized
where \( t \) is the intensity of the incident light. Now, if \( a = 1 \) and assuming no transmittance, the intensity of the reflected light is \( I_0 \). Assuming that solutions to Eq. (7) are unique, this is the only case where the reflected intensity is isotropic. This is a reasonable assumption, especially for a transmittance with isotropic single scattering. But this phenomenon affects standardized industrial measurements involving perfect diffusers, which are often used in calibration procedures to allow for measurement of absolute reflectance. This will be further investigated by the authors in future publications. It is noteworthy that if the diffuser were constructed from some material involving only surface scattering, i.e., with no light penetrating the medium, this phenomenon could possibly be avoided.

4. VALIDITY OF THE KUBELKA–MUNK MODEL

The reasoning up to this point has consequences for the widespread KM model, since this model assumes an isotropic light distribution throughout the medium. To investigate the validity of the KM model, general radiative transfer theory as expressed by Eq. (7) is manipulated below to see under which conditions the KM model can be recovered. In this way the validity conditions of the model can be stated.

The KM model assumes isotropic single scattering, and therefore the phase function \( p(\cos \theta) = 1 \) is used. Equation (7) then becomes

\[
\frac{dl}{ds} = -s \frac{\sigma_s}{4\pi} \int_{4\pi} I d\omega. \tag{12}
\]

If the intensity is divided into two parts, \( I^r \) and \( I^s \), representing polar angles in the intervals \([0, \pi/2]\) and \([\pi/2, \pi]\), respectively, such that \( I = I^r + I^s \), Eq. (12) can be written

\[
\frac{dl^r}{ds} = -s \frac{\sigma_s}{4\pi} \int_{4\pi} (I^r + I^s) d\omega
\]

\[
\frac{dl^s}{ds} = -s \frac{\sigma_s}{4\pi} \int_{4\pi} I^s d\omega. \tag{13}
\]

If \( dx \) denotes infinitesimal distance from the medium’s lower surface and upwards as in the KM model, the infinitesimal distance \( ds \) in any direction can be written \( ds = dx/u \) where the definition \( u = \cos \theta \) with \( \theta \) the polar angle is used. The distance measure can then be changed and Eq. (13) becomes

\[
\frac{dl^r}{dx} = -s \frac{\sigma_s}{4\pi} \int_{4\pi} (I^r + I^s) d\omega
\]

\[
\frac{dl^s}{dx} = -s \frac{\sigma_s}{4\pi} \int_{4\pi} I^s d\omega. \tag{14}
\]

Equation (14) can be integrated over polar angle (or \( u \)) since \( u \) and \( x \) are independent:

\[
\int_0^1 \frac{dl^r}{dx} du = \int_0^1 -s \frac{\sigma_s}{4\pi} \frac{1}{4\pi} \left( (I^r + I^s) d\omega \right) du
\]

\[
\int_0^1 \frac{dl^s}{dx} du = \int_0^1 -s \frac{\sigma_s}{4\pi} \frac{1}{4\pi} \left( I^s d\omega \right) du. \tag{15}
\]

If \( I \) is assumed isotropic throughout the medium, the integrations in Eq. (15) can be performed trivially. If \( I \) is somewhere anisotropic, or if there is no other knowledge about \( I \), there is no possibility for further analytic simplifications of general radiative transfer theory, and one would have to resort to standard solution procedures for...
the complete radiative transfer problem. Thus, the assumption that the intensity be isotropic is essential, and assuming this Eq. (15) becomes

\[
\begin{align*}
\frac{1}{2} dI' &= -\left(\sigma_a + \sigma_s I'\right) + \frac{1}{2} \sigma_s I'' + \frac{1}{2} \sigma_a I' \\
\frac{1}{2} dI' &= -\left(\sigma_a + \sigma_s I'\right) + \frac{1}{2} \sigma_s I'' + \frac{1}{2} \sigma_a I'.
\end{align*}
\]

(16)

Equation (16) can be rewritten to facilitate comparison with Eq. (8), i.e., the KM model:

\[
\begin{align*}
\frac{dI'}{dx} &= -\left(\sigma_a + 2\sigma_s I'\right) + \sigma_s I'' \\
\frac{dI'}{dx} &= -\left(\sigma_a + 2\sigma_s I'\right) + \sigma_s I''.
\end{align*}
\]

(17)

It can be seen that Eqs. (8) and (17) are equivalent if \( I' = -i, I' = -j, \sigma_s = \sigma_{ka}, \) and \( \sigma_a = \sigma_{ka}/2. \) The first two equalities are just a matter of the definition of the coordinates. The factor \( \frac{1}{2} \) in the last equality appears, because the KM model scattering coefficient is defined to scatter radiation into the opposite direction only, while it is equally distributed over all directions in general radiative transfer theory (when assuming isotropic single scattering).

The assumptions made to recover the KM model from radiative transfer theory are thus:

1. Isotropic single scattering.
2. Isotropic intensity throughout the medium.

If any of these conditions does not hold, the KM model is not exactly valid and applying it will introduce errors. The magnitude of the error will, of course, depend on each particular situation. Intuitively one would expect a larger error in situations with strong anisotropy, that is, when \( \sigma' = 0 \) and/or \( \sigma'' = 0. \)

Another consequence of this is that an exact translation between the KM model and general radiative transfer cannot exist if these conditions are not fulfilled. Any translation will then necessarily be an approximation. Translations proposed by other authors [25–31] thus hold exactly only when the validity conditions are fulfilled, or approximately in certain given cases. This means that the KM scattering and absorption coefficients are inherently model dependent, and that they cannot be given a physical interpretation.

5. CONSEQUENCES FOR THE KUBELKA–MUNK MODEL

Based on the reasoning in Subsection 3.C, where anisotropy was explained, the actual validity conditions of the KM model can now be stated. Since the situation in Fig. 4(a), i.e., when an infinitely thick non-absorbing medium is illuminated isotropically (diffusely) is the only one leading to isotropic intensity throughout the medium, the conditions of exact validity are:

1. Non-absorbing and infinitely thick medium.
2. Isotropic illumination.

3. Isotropic single scattering.

If these hold the KM model and general radiative transfer theory coincide and give the same result. If they do not hold the intensity is anisotropic and an angle resolved model is necessary to describe the anisotropy. In this case the KM model and general radiative transfer theory are incommensurable.

All media will in practice absorb some light, so the first KM validity condition can thus never be fulfilled. Applying the KM model will then always introduce some error. However, it is not impossible that the KM model give satisfactory results in some applications. This is further investigated in Subsection 5.A.

A. Deviation between General Radiative Transfer and the Kubelka–Munk Model

The reflectance factor \( R \) can be defined as the flux of energy reflected in a solid angle divided by the flux reflected from an ideal perfect diffusor in the same solid angle when both media are under the same illumination:

\[
R(\phi) = \frac{\int_{\Delta \omega} I_{\text{rad}} \cos \theta d\omega}{\int_{\Delta \omega} I_{\text{rad}} \cos \theta d\omega}.
\]

(18)

\( I_{\text{rad}} \) is the intensity of the radiation reflected from the perfect diffusor, \( I_{\text{rad}} \) the intensity of the radiation reflected from a general surface, and \( \phi \) is the solid angle. Since \( I_{\text{rad}} \) is independent of direction the denominator can be written \( \int_{\Delta \omega} I_{\text{rad}} \cos \theta d\omega = I_{\text{rad}} \Delta \omega \) if the integration is performed over the upper half-sphere, that is, if the total reflectance is considered. Normalizing the incident flux, the definition \( I_{\text{rad}} \Delta \omega \equiv 1 \) can be used. The total reflectance factor \( R_T \) then becomes

\[
R_T = \int_{\Delta \omega} I_{\text{rad}} \cos \theta d\omega.
\]

(19)

The KM model neglects the variations in \( I_r \), and the KM total reflectance factor \( R_{T,KM} \) can then be written

\[
R_{T,KM} = I_{r,max} \pi,
\]

(20)

where \( I_{r,max} \) is a constant value assigned to the intensity, in a real situation usually by some measurement. The intensity \( I_r \) is then implicitly assumed to represent the intensity in all directions. \( R_{T,KM} \) will, through \( I_{r,max} \) depend on the detection angle, but the correct value \( R_T \) is not changed. Depending on the particular problem and how \( I_r \) is chosen, the deviation between the correct general radiative transfer theory and the approximate KM model will vary. This deviation, or error, is a compound consisting of the combination of measurement setup and KM model interpretation. To give an upper bound for the error and to illustrate the dependence on medium properties (single scattering albedo \( \omega \) and optical thickness \( \tau \), \( I_r \) is here chosen as the intensity deviating the most from the intensity corresponding to the correct total reflectance factor. Denoting this intensity \( I_{r,max} \) it is calculated as
In a measurement situation this would correspond to placing the detector in the angle with the same polar argument as $I'_{\text{max}}$. Using the angle resolved model DORT2002 to solve the radiative transfer problem and then inserting the solution in Eq. (19), the relative error $(R_{T,\text{KM}} - R_T)/R_{T,\text{KM}}$ can be calculated for a set of medium parameters. The supposed validity conditions of the KM model are used, i.e., diffuse illumination and isotropic single-scattering.

In Fig. 5(a) the relative error in percent is shown for optical thickness $\tau \in [0.5, 30]$ and albedo $a \in [0.5, 0.999]$. It can be seen that the error is 20–40% in a large region, and that it increases asymptotically for thin media. It is close to zero only for optically thick and highly scattering media. This is in accordance with the results presented above in Subsection 3.A, where it was shown that the anisotropy is large in exactly the cases shown here having a large relative error. The error is a consequence of the anisotropy.

Using Eq. (21), the DORT2002 simulations show that $I'_{\text{max}}$ corresponds to intensities $I_r$ having polar arguments near $\pi/2$, i.e., grazing angles, and that the amount of reflected light is systematically overestimated. That is, $R_T < R_{T,\text{KM}}$ in all cases for the specific choice of parameters. It must then hold that $R(\phi') < R_T$ for some solid angle $\phi'$, since the reflectance factor in Eq. (18) becomes the total reflectance factor $R_T$ when the integration in Eq. (18) is performed over the whole hemisphere. This is investigated in Figs. 5(b)–5(d) where the relative error is shown when $I'_r = I_r(0^\circ)$, $I'_r = I_r(35^\circ)$, and $I'_r = I_r(55^\circ)$, respectively, still using diffuse illumination and isotropic single-scattering. It is obvious that $R_T > R_{T,\text{KM}}$ when $I'_r$ approaches the normal direction, and the reflectance factor is thus underestimated in this case. The error is then around 20–30%. When $I'_r = I_r(55^\circ)$ the error is small, and one can conclude that for some intensity $I'_{\text{max}}$ it must hold that $I'_{\text{max}} = R_T/\pi$. There is thus necessarily an intensity $I'_{\text{min}}$ such that the corresponding measurement direction, that corresponds to the total reflectance. This is shown in Fig. 6 where the mean relative error is calculated for different detection polar angles $\theta$ over the set $\tau \in [0.5, 30]$, $a \in [0.5, 0.999]$ using DORT2002. Figure 6 shows that in an average sense and over the chosen set of medium parameters, it holds that $I'_{\text{max}} = I(\theta) = I(51^\circ)$. In a measurement situation this means that to minimize the error introduced by the KM model the reflectance factor detected around $\theta = 51^\circ$ from the normal should be used.

It is interesting to note that common practice in the paper industry is to use the standardized d/0 instrument geometry [32] for reflectance measurements. This instrument has diffuse illumination and measures the reflectance in the normal direction. That is, $I'_r = I_r(0^\circ)$, which is not the optimal angle of measurement according to the results presented here. Though, it should be mentioned that calibration of a measurement procedure normally improves the situation, since the anisotropy then implicitly can be partly compensated for. Practical and instrument related issues are discussed in part II [21] of this work.

6. DISCUSSION AND CONCLUSIONS

It has been shown in this work that the reflectance from turbid media is always anisotropic with the exception of a diffusely illuminated infinitely thick non-absorbing medium. The anisotropy depends on the relative contributions of different scattering depths in the medium, and...
near-surface bulk scattering increases the relative reflectance at polar angles close to the medium surface. Near-surface bulk scattering dominates in thin and highly absorbing media, since light penetrating the medium further has a high probability of being transmitted or absorbed. Another factor affecting the relative contributions from different depths is the illumination. Obliquely incident illumination causes the light to scatter closer to the surface and consequently the reflectance at large polar angles to increase, and vice versa with normal incidence. Situations with strong anisotropy are thus

- Highly absorbing media.
- Optically thin media.
- Non-diffuse (anisotropic) illumination.

These results also lead to the conclusion that the supposed perfect diffuser cannot reflect diffusely independently of the illumination. This can have practical consequences since it is often used in calibration routines.

The actual validity conditions of the KM model were derived. It is necessary that the intensity be isotropic throughout the medium for the KM model to give exact results. Since this work showed that isotropic intensity is obtained only when an infinitely thick non-absorbing medium is illuminated diffusely, this is consequently the validity condition of the model together with the requirement of isotropic single scattering. Assuming anisotropic single scattering will increase the anisotropy of the intensity, thus further increasing the error. Depending on the medium properties and the way the intensity is approximated or measured, the magnitude of the error introduced when applying the KM model will vary. This work showed that the error can be 20–40% in a large region of medium parameters. It was also shown that there exists an optimal angle where the KM model error is minimized for a given medium or experimental setup. This is a fact that can be exploited, for example, when designing setups for measuring reflectance.

ACKNOWLEDGMENTS

This work was financially supported by The Swedish Governmental Agency for Innovation Systems (VINNOVA), which is gratefully acknowledged.

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II

Paper II
Anisotropic reflectance from turbid media.
II. Measurements

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1. INTRODUCTION
Radiative transfer theory has been studied throughout the last century to describe the propagation of light in turbid media. Different models of varying degree of complexity have been applied in several scientific and industrial fields, ranging from astrophysics and neutron diffusion to color predictions in plastics, textile, and paper. The first theoretical attempts were made by Schuster [1], while the general radiative transfer theory is usually ascribed to Chandrasekhar [2]. The most widespread industrial model is the one presented by Kubelka and Munk [3–5] (hereafter denoted KM), but now efficient general solution methods exist, such as DISORT [6] and DORT2002 [7].

Schuster acknowledged the importance of angular variations in the light distribution within a medium, but found the matter too complex for further investigation. Other authors have observed angular variations, or anisotropy, and have discussed its potential importance [8–10]. One important difference between two-flux models like KM and more general solution methods like DORT2002 is that the latter can describe and handle the angular resolution of light. The KM model assumes an isotropic single scattering process, and is further based on the implicit assumption that this, together with a perfectly diffuse (Lambertian) illumination, will give isotropic light intensity throughout the medium. This is not at all true, although several industrial applications rely on this assumption. The disagreement between KM and general radiative transfer theory has been investigated by Edström [11], who noted that the error is larger for optically thin and highly absorbing media.

Part I of this work [12] (appearing consecutively in this issue) theoretically investigates anisotropy, explains how it arises in plane-parallel turbid media and shows that anisotropy is present in all real situations. It is shown that the anisotropy depends on the relative contributions of different scattering depths in the medium, and that near-surface bulk scattering increases the relative reflectance in large polar angles. Increasing absorption, transmittance, or the average angle of incidence of the illumination causes the amount of near-surface bulk scattering to increase, and consequently also the relative reflectance in large polar angles increases. In this sense the anisotropy increases as the amount of near-surface bulk scattering increases. These results hold even for an isotropic single scattering process, and asymmetric single scattering will obviously introduce further anisotropy. It is then concluded that there can be no perfect bulk-scattering diffruser, which has practical impact on, e.g., calibration routines. It is further shown that anisotropy makes instruments measuring only in one direction overestimate or underestimate the total reflectance, depending on the particular instrument geometry and medium properties. Part I also derives validity conditions for the KM model, and shows that errors can be 20–40% in a large region of medium parameters. But it is also shown that an optimal detection angle can be determined that minimizes the KM model error for a given medium or experimental setup, which could be exploited for designing reflectance measurement setups.

The purpose of part II (this paper) is to experimentally verify the theoretical findings in part I and to discuss measurement related issues concerning anisotropy and consequences for industrial applications. A specific aim is thus to use measurements to verify the theoretical predictions from part I regarding increased anisotropy—in the form of increased relative reflectance in large polar angles—for highly absorbing and optically thin media as well as oblique illumination. Another aim is to verify the...
predicted anisotropic reflectance of an alleged perfect dif-
funser. One additional aim is to discuss consequences for
some of these findings on practical measurements and in-
dustrial applications, e.g., for standardized diffuse re-
fectance measurements, their calibration, and their inter-
pretation using the KM model. One final aim is to discuss
the possibility to design a measurement setup with a cor-
responding data interpretation model that is in some re-
spect optimal.

This part of the paper is more applied, and the reader is
referred to part I [12] for a theoretical treatment, in-
cluding definitions and equations.

2. METHOD

In order to experimentally investigate angle resolved re-
fectance and how it depends on medium properties, a se-
ries of paper handsheets was made and their reflectance
measured with a goniophotometer. The handsheets con-
tain different amounts of dye and vary in grammage. In-
creasing the amount of dye is equivalent to decreasing the
single scattering albedo in the wavelengths corresponding
to the region of absorption of the dye. The grammage can
be treated as equivalent to the paper thickness [13], and
is proportional to the paper optical thickness. The hands-
sheets thus allow for an investigation of how the deviation
from an isotropic intensity distribution depends on albedo
and medium optical thickness. A corresponding in-
vestigation was also made for a “perfect reflecting diffu-
sor” using a sample that is provided by instrument sup-
pliers for calibration purposes. In this case the illu-
mination was varied to study its influence on anisotropy.
To compare measurements with the theoretical pre-
dictions in part I of this work, simulations were made
with the angle resolved DORT2002 model. (The DORT2002
software is freely available from the authors.)

A. Material

The paper handsheets used in this work were prepared
with a Formette Dynamique. All handsheets were made
from a mix of equal amounts of kraft and birch pulps hav-
ing Shopper/Riegler numbers of 18° and 23° SR, respec-
tively. Sheets of two approximate grammages, 30 and
50 g/m², were prepared. All sheets contain 0.02% PAM,
0.2% Bentonit, 0.35% Cerestar, and 22% filler (PCC Syn-
carb FO 474-MJ). The amount of blue dye was changed
from 0 to 1% of the fiber weight in four steps. This gives

eight sheets in total and is summarized in Table 1. The
dye used was Levacell Fast Blue KS-6GLL Liquid, manu-
factured by Lanxess. This is a cationic direct dye, and it
does not affect the structure of the paper. The sheets were
dried in a cylinder dryer for 5 min at a temperature of
105°C and a pressure of 1.50 bars.

This process assures that the paper sheets have mini-
mal surface effects, such as, e.g., gloss. In this way they
can be considered to be purely bulk scattering turbid me-
dia, and their interaction with light is thus accurately de-
scribed by radiative transfer theory.

B. Measurements

The angle resolved measurements were made with a Zeiss GP3
goniophotometer. This instrument uses colli-
mated illumination from an incandescent lamp, and the
detector can be adjusted to simulate the specific illumination
angle of the measurement setup, and a numerical
Y-filter [14] is implemented. The experimental results can
be used in this case in order to study its influence on the reflectance
from the diffusor. The barium sul-
phate diffusor delivered with the instrument was used.
The angle of incidence of the illumination was varied in
this case to study its influence on the reflectance
from the diffusor.

C. Simulations

The DORT2002 model [7] is adapted to light scattering
simulations in paper and print, has been successfully ap-
plied to real paper industry problems [11,15,16], and has
been thoroughly evaluated [17,18]. DORT2002, being
angle resolved, is in this work used to predict the paper
samples’ reflectance and the corresponding dependence on albedo and optical thickness.

The same model is also used to predict the anisotropic re-
reflectance from the “perfect diffusor.” DORT2002 is ad-
justed to simulate the specific illumination and detection
geometry of the measurement setup, and a numerical
Y-filter [14] is implemented. The experimental results can
d thus be compared with the theoretical results.

In order to obtain values for the paper samples’ albedo
and optical thickness, the samples were measured with a Datacolor SF450 Elrepho complying with ISO2469 [19].
This instrument has d/0 geometry, meaning that the illu-
mination is diffuse and the detector is centered around 0°,
i.e., the normal direction of the paper. Using these illu-
minance data, calculations were performed with DORT2002
adjusted to this instrument geometry (more details are
given by Edström [16]). This procedure gives numerical
values for the paper samples’ albedo and optical thickness
that can be used when simulating the goniophotometer.

| Table 1. Overview of the Paper Handsheets Used in This Work |
|-------------------|------------------|-------------------|
| Sheet No. | Grammage (g/m²) | Dye (% of fiber weight) |
| 1      | 31.80           | 0.00               |
| 2      | 31.69           | 0.25               |
| 3      | 32.54           | 0.50               |
| 4      | 32.27           | 1.00               |
| 5      | 64.97           | 0.00               |
| 6      | 65.73           | 0.25               |
| 7      | 66.91           | 0.50               |
| 8      | 65.87           | 1.00               |
described in 2.B. Subsection 2.B. The asymmetry factor of the Henyey–Greenstein phase function [20] describing the anisotropy of each single scattering, usually denoted $g$, is set equal to 0.8, which is the only reported value for $g$ in a paper application [21]. This corresponds to forward scattering dominating the single scattering process, since $g=1$ is complete forward scattering and $g=0$ is isotropic single scattering.

3. RESULTS

A. Dependence of Anisotropy on Medium Absorption and Thickness

To study the dependence of anisotropy on absorption, opaque pads of paper sheets 1–4, with varying amounts of dye, are measured and simulated. In this way the effect of absorption is isolated, since there is no transmittance. Goniophotometer measurement data and the corresponding simulations are shown in Fig. 1. The data are presented in terms of the bidirectional scattering distribution function (BRDF, $f_r$), which is proportional to the intensity $I$ and the reflectance factor $R$ [22]. The BRDFs are normalized to coincide at polar angle $\theta=0^\circ$ in order to highlight the dependence of the anisotropy on absorption. That is, if the BRDF of sample $i$ is denoted $f^i_r$, the normalized BRDF $\tilde{f}_r^i$ is $\tilde{f}_r^i(\theta)=f^i_r(\theta)f_r(0^\circ)$, where the dependence on polar angle $\theta$ is included. It can be seen in Fig. 1 that the anisotropy increases as the absorption increases since relatively more light is reflected at large polar angles.

The dependence of anisotropy on medium optical thickness can be studied by using measurement data from paper sheets of varying grammage and an opaque pad of paper sheets. Sheets 1 and 5 are used since they do not contain dye, thus isolating the effect of transmittance. In Fig. 2 these data together with the corresponding model predictions are shown. It can be seen that the anisotropy increases in the same characteristic way in both measurements and simulations as the medium optical thickness decreases. Relatively more light is reflected at large polar angles for thin media, i.e., when transmittance is significant, and the anisotropy is larger in this sense for thin media. The reflectance closest to isotropic (Lambertian) is
as expected obtained from the opaque pad of paper sheets. These results are in complete agreement with the findings of part I of this work.

B. Dependence of Anisotropy on Illumination: The Non-Diffuse Reflectance of the Perfect Diffusor

The dependence of anisotropy on illumination can be investigated by measuring and simulating the reflectance from a highly scattering and non-transmitting medium, i.e., what is normally believed to be a perfect diffusor. This eliminates effects of absorption and transmittance. The barium sulphate diffusor delivered with the Zeiss GP3 goniophotometer is used here, and in the DORT2002 model the albedo is set equal to one, and the optical thickness is increased until the point of zero transmittance. Isotropic single scattering, i.e., $g_{\text{HS11005}} = 0$, is assumed in the simulations. Assuming $g_{\text{HS11005}} \neq 0$ would introduce further anisotropy, and isotropic single scattering is a necessary condition for isotropic reflectance in this case. In both measurements and simulations the illumination angle is varied, and incident angles $0^\circ$, $30^\circ$, $45^\circ$, and $60^\circ$ are used. When illuminating in $0^\circ$ the reflectance cannot be measured for polar angles $\theta < 30^\circ$ because of physical constraints of the instrument. Figure 3 shows the measured and predicted BRDFs with varying illumination angle.

Fig. 3. Measured BRDFs of (a) a barium sulphate diffusor and predicted BRDFs of (b) a non-absorbing and non-transmitting medium with varying illumination angle. Angles $0^\circ$, $30^\circ$, $45^\circ$, and $60^\circ$ are used. It is seen that the relative reflectance at large polar angles increases as the illumination angle increases. The reflectance can thus be anisotropic both from an alleged perfect diffusor (a) and an ideal bulk scattering diffusor (b). Lambertian reflectance is included for reference.

The relative reflectance is larger in the normal direction if the illumination is incident around the normal direction. These results are also in agreement with the results from part I of this work.

4. DISCUSSION AND CONCLUSIONS

It has been seen that the measured and model predicted reflectance from various turbid media, including an alleged perfect diffusor, show the same type of dependence between anisotropy and medium absorption, optical thickness, and illumination. Increasing absorption and illumination angle or reducing the thickness has the same characteristic effect on the anisotropy, since this increases the relative reflectance at large polar angles. This is in complete agreement with the conclusions of part I of this work. The correlation between measurements and simulations shows that an explanation of the observed phenomenon can indeed be found within the framework of radiative transfer theory. Using the conclusions from part I, the results can be understood by considering the amount of near-surface bulk scattering. Increasing absorption or illumination angle, or reducing the optical thickness increases the amount of near-surface bulk scattering, which leads to higher reflectance at large polar angles. This holds since light deeper inside a highly absorbing or transmitting medium has a high probability of being absorbed or transmitted, and since the light has to travel a larger distance to reach a certain depth if the illumination angle is increased. The angular distribution of the reflected light is thus determined by the relative contributions to the reflectance from different depths in the medium, and the relative contributions are determined by the absorption, optical thickness, and illumination.

A. Consequences for the Kubelka–Munk model

It has been seen in this work that the light reflected from common paper samples can be highly anisotropic. The samples studied are comparable to, e.g., common office paper or newsprint, so the phenomenon is present in everyday situations. Because of this, the KM model is invalid in practically all situations, since it is based on the false assumption that the light is isotropic throughout the medium. The amount of error introduced when assuming this was indicated in part I of this work to be 20–40% in a large region of parameters, but this holds under the assumption of isotropic single scattering. Anisotropic single scattering will increase the error further. To reduce this error, angle resolved models are necessary, but it is also possible to customize a measurement setup for a particular set of samples as described in part I. The determination of the KM scattering and absorption coefficients is a standardized procedure [23], but it is important that anisotropy be taken into account in these calculations.
B. Effect of Anisotropy on Established Instruments

If reflectance measurements are performed with instruments having different illumination and detection geometries, anisotropy has to be taken into account to be able to compare the measurements. If assuming isotropically reflected light the inter-instrument difference will vary depending on the measurement setup and the sample properties. This is further investigated for the common 45/0 [24] and d/0 [19] geometries by the authors [25].

Calibration routines involving perfect diffusors are affected by the anisotropy of the light reflected from the diffusor if the anisotropy is not taken into account. Judging from Fig. 3, the barium sulphate diffusor, which is a common used material, probably has anisotropic single scattering, since in the measurements the light tends to be scattered more in the forward direction compared to the backward direction. This further increases the effect of anisotropy, and it is more severe if directed illumination is used since illuminating diffusely to some extent reduces this effect. Though, calibration can in many situations improve the results if the total reflectance is overestimated or underestimated in the same way in both calibration and actual measurements. But without taking angular variations into account the measurements will contain errors due to the anisotropy, and standards for reflectance measurements should be modified accordingly.

C. Optimal Instrument Geometry for Reflectance Measurements

As described in part I of this work an optimal instrument in the sense that it minimizes the error in the KM model can be readily constructed for a set of samples. The instrument detection geometry would have to be modified if changing the error tolerance or the set of samples, and the modification would require angle resolved calculations. In order to construct a generic instrument without need for recalibration it is necessary with several detection angles. Given that the reflectance is fairly monotone with respect to polar angle, it could be enough with two or three detection angles. Combining accurate measurements in several directions with an angle resolved model such as DORT2002 would give access to the objective and open up possibilities for further investigations. How- ever, other phenomena such as surface roughness or gloss can affect the measurements, and to stay optimal these have to be included in the model.

5. FUTURE WORK

Measuring the spectral reflectance in several directions allows for an investigation of the spectral dependence of the asymmetry factor. This work is planned by the authors and will be published shortly. Furthermore, fluorescence is present in many situations. By including this in an angle resolved model it can be understood more thoroughly. Also, effects of surface roughness, gloss, or the refractive index have to be taken into account to develop a more comprehensive model. The authors believe that better instruments for reflectance measurements can be constructed, and that they can be combined with angle resolved models such as DORT2002. This would allow for a better characterization of the light scattering properties of the medium and open up possibilities to optimize, for example, the optical appearance.

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Paper III
Geometry Related Inter-Instrument Differences in Spectrophotometric Measurements

Per Edström, Magnus Neuman, Stefanos Avramidis and Mattias Andersson

KEYWORDS: Reflectance measurements, Spectrophotometry, Instrument geometry, Measurement differences, Radiative transfer, Anisotropy, Absorption, Transmittance

SUMMARY: The L&W Elrepho d/0 and the Spectrolino 45/0 instruments are examined using paper samples with differences in geometrical characteristics. External factors that influence the measurement in general. The model/measurement agreement is satisfactory as the characteristic behavior is captured in reflected light. The simulated and measured differences are shown to depend on the anisotropy of the sample, and explained using DORT2002, and the instrument calibration. The simulated and measured differences are found to agree in magnitude, caused by instrument geometry. The simulated and measured differences are found to agree in magnitude, and the differences are mapped against sample properties. It is observed that the 45/0 instrument detects higher reflectance from paper samples with negligible absorption and transmittance. When there is considerable absorption (dyed samples) or transmittance (thin samples), the d/0 instrument detects higher reflectance. The physical mechanism behind this behavior is studied and explained using DORT2002, and the instrument differences are shown to depend on the anisotropy of the reflected light. The model/measurement agreement is satisfactory as the characteristic behavior is captured in almost all cases studied. This new understanding is important for facilitating accurate data exchange between the paper and graphic arts industries, but also for interpretation of reflectance measurements in general.

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The paper and the graphic arts industries use different spectrophotometer instruments to assess the optical properties of printed and unprinted paper through reflectance measurements. The instruments differ significantly in geometrical characteristics since the illumination and detection conditions are not the same (ISO 2469, ISO 5-4, ISO 13655, DIN 5033), and the different instrument types have been reported to yield incomparable measurements (Radencic et al. 2008; Seghi 1990). Explanations have been proposed (Spooner 1994) based on differences in spectrophotometer properties like detector linearity, spectral calibration, spectral bandwidth treatment, transulence phenomena and other edge effects.

The standardized instruments for measuring reflectance used in the paper and graphic arts industries have a d/0 or 45/0 geometry, respectively. The d/0 geometry, used in the paper industry, illuminates the paper sample diffusely and measures reflectance in 0°, i.e. in the normal direction. The 45/0 geometry, used in the graphic arts industry, illuminates the sample with directed light incident in 45° from the normal to the paper and measures reflectance in 0°. Apart from the geometrical differences, the instruments differ in calibration routines, sample background, measurement spot size and UV content of the illumination. Furthermore, there exist few if any standardized methods for communication of optical properties between the paper and graphic arts industries. Independently of each other, they have developed standardized methods for the measurement of optical properties, optimized for the different requirements and purposes in each industry. The differences between the color measurement methods used in the two industries do inarguably cause problems in both color management and proofing. Fluorescence is by far the biggest problem, and steps have been taken to address this in 45/0 instruments with adjustable UV content.

The optical model in most frequent use in the industry, the Kubelka-Munk (KM) model (Kubelka, Munk 1931; Kubelka 1948), considers light travelling in the two main directions up and down, and the medium is characterized by its phenomenological KM scattering and absorption coefficients s and k. However, the KM model does not allow for investigation of the influence of the angle resolved intensity, and thereby of the instrument geometry, on the detected reflectance factor, R.

To accurately model the instruments’ geometry it is convenient to use radiative transfer theory (Chandrasekhar 1960), where the medium is
characterized by its physically objective scattering and absorption parameters (cross sections) $\sigma_s$ and $\sigma_a$. For a medium with an isotropic single-scattering process, there are simple approximate relations (exact relations do not exist (Neuman, Edström 2010a)) between the sets of scattering and absorption parameters, $s = \frac{1}{2} \sigma_s$ and $k = 2 \sigma_a$. The relation between scattering and absorption is often indicated using the dimensionless single scattering albedo, $a = \sigma_s / (\sigma_s + \sigma_a)$. Radiative transfer theory describes in detail the interaction of light and turbid media, such as paper, and it allows for the calculation of the angle resolved intensity of light. In this way the geometrical differences in illumination and detection conditions can be included in the model and simulated. The KM model has been shown to be a simplified case of the more general radiative transfer theory (Edström 2004, Neuman, Edström 2010a, Neuman, Edström 2010b). Edström (2005) has developed the computational tool DORT2002\(^1\) for numerical solution of the radiative transfer problem, and this tool has been thoroughly evaluated (Edström 2008; Edström 2009). This tool is adapted to applications in the paper industry and it is based on the same assumptions about the paper medium as Kubelka-Munk theory uses (a continuum of scattering and absorption sites), but it is generalized to handle angular variations in the light. Pierce and Marcus (1996) have presented a comparison of KM and radiative transfer models for 45/0 and d/0 measurements, but their focus was on the differences between the two theoretical models and not on the influence of instrument geometry.

The purpose of the present work is to investigate the inter-instrument differences between instruments of d/0 and 45/0 type, and to examine to what extent they can be explained by taking the instrument geometry into account. This work also aims to quantify these differences, and to suggest what can be done to minimize them. It is the purpose to show how both proper measurement routines and better measurement data interpretation can facilitate this, and to show the size of the errors made if this is not considered.

### Methods and Materials

Paper samples were produced and measured, and the measurements were simulated as described in separate sections below. The difference in the detected reflectance factor from d/0 and 45/0 spectrophotometers was measured and simulated for a wide variety of different paper samples. A measure of the reflectance factor difference was introduced as $\Delta R = R_{d/0} - R_{45/0}$; therefore a positive $\Delta R$ means that the 45/0 detects larger reflectance factor than the d/0 and vice versa. This was used to quantify the differences between instrument types, and both theoretical and experimental arguments were used to explain the findings.

#### Paper samples

To study the effect of scattering, absorption and paper thickness on the differences between the two types of instruments, a series of samples with low grammage and varying filler and dye content was prepared with a Formette Dynamique. All samples were made from a mix of equal amounts of kraft and birch pulps, having Shopper/Riegler numbers 18º and 23ºSR respectively. Sheets of approximate grammage of 30 g/m² were prepared, each with and without filler (PCC SYNCRAB FO 474-MJ). All sheets contain 0.02% PAM, 0.2% Bentonit and 0.35% Cerestar. The amount of blue dye was changed from 0 to 1% of the fiber weight in four steps, which gives 8 types of sheets in total. The dye used was Levacell Fast Blue KS-6GL Liquid, manufactured by Lanxess. This is a cationic direct dye and it is assumed that it does not affect the structure of the paper. The sheets were dried in a cylinder dryer for 5 minutes at a temperature of 105ºC and a pressure of 1.50 bars. The samples do not contain any FWA. These samples are denoted Series 1 and their properties are shown in Table 1.

<table>
<thead>
<tr>
<th></th>
<th>Four samples, no fillers</th>
<th>Four samples, 22 % fillers</th>
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<tr>
<td>0.00 % dye content</td>
<td>0.00 % dye content</td>
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<tr>
<td>0.33 % blue dye content</td>
<td>0.33 % blue dye content</td>
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<tr>
<td>0.67 % blue dye content</td>
<td>0.67 % blue dye content</td>
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<tr>
<td>1.00 % blue dye content</td>
<td>1.00 % blue dye content</td>
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</table>

The spectral variation of scattering and absorption parameters for one of the samples in Series 1 is illustrated in Fig 1. The similarity between the phenomenological KM parameters $s$ and $k$ and the objective parameters $\sigma_s$ and $\sigma_a$ of radiative transfer theory is evident.

A second series of highly scattering samples was also used in this work. Three different filler contents and three different grammages were chosen and prepared with a small-scale experimental paper machine. All samples were made from a mix of 25% long fiber and 75% short fiber pulps, both having Shopper/Riegler numbers 25ºSR. Sheets of approximate grammage 80, 100 and 120 g/m² were

\(^{1}\) DORT2002 is freely available from the authors.
Fig 1. Comparison of the physically objective scattering and absorption parameters $\sigma_s$ and $\sigma_a$ of radiative transfer theory and the phenomenological KM parameters $s$ and $k$, for the 30 g/m² sample containing 0.33% dye and no fillers from Table 1. The parameter sets are obtained by using DORT2002 and KM respectively on standardized paper industry reflectance factor measurements. The similarity of the two parameter sets is evident.

prepared, each with filler contents of approximately 0%, 15% and 30% PCC. All sheets contain 0.05% Bentonit SH, 0.01 % Percol 178 and 0.19-0.24% C*Bond HR35844 together with 0.13-0.17% Kemsize 200. The samples do not contain any dye or FWA. These samples are denoted Series 2 and their properties are shown in Table 2.

Table 2. Series 2 consists of nine samples that vary in grammage and amount of filler. No samples contain dye.

<table>
<thead>
<tr>
<th>Three samples, no fillers</th>
<th>Three samples, 15 % fillers</th>
<th>Three samples, 30 % fillers</th>
</tr>
</thead>
<tbody>
<tr>
<td>80 g/m²</td>
<td>80 g/m²</td>
<td>80 g/m²</td>
</tr>
<tr>
<td>100 g/m²</td>
<td>100 g/m²</td>
<td>100 g/m²</td>
</tr>
<tr>
<td>120 g/m²</td>
<td>120 g/m²</td>
<td>120 g/m²</td>
</tr>
</tbody>
</table>

Fluorescence and surface characteristics influence strongly the reflectance measurements, and introduce differences between the d/0 and 45/0 instruments that are far more significant than the geometrical differences studied in this work. It has been shown (Andersson, Norberg 2007) that fluorescence can introduce errors up to 7ΔE*ab. This is, among others, due to the different treatment of UV (illumination, calibration and filtering) between the instruments. The interested reader can find more on this in Radencic et al. (2008).

Samples included in this work were uncalendered uncoated papers with low gloss and no added FWA. Gloss measurements were made with a Zehntner glossmeter. Ten measurements were made on each sample, and the gloss was found to be low with an average of 1.0 for 20° and 2.9 for 60° and 75°. The surface roughness averaged at 250 Bendtsen for the samples with high filler content, and at 550 Bendtsen for the samples with no fillers.

Measurements

The d/0 instrument used in this work was a L&W Elrepho, which complies with ISO 2469 (1994). It is calibrated with specific reference samples in accordance with the ISO hierarchy. The illumination is diffuse, using an integrating sphere with a gloss trap. The detection uses a 4° aperture sensor directed along the normal to the sample, and requires a sample area of 34 mm radius. The illuminated area is larger than the measured area, which avoids edge effects. Its repeatability is given by the manufacturer to be 0.01 ΔE*ab, where the CIE 1976 color difference in the L*a*b* color space is given by:

$$\Delta E_{ab} = \sqrt{\left(\Delta L^*\right)^2 + \left(\Delta a^*\right)^2 + \left(\Delta b^*\right)^2}$$

(Wyzecki, Stiles 2000).

Instruments with 45/0 geometry have different characteristics depending on the manufacturer. The 45/0 instrument used in this work was a GretagMacbeth Spectrolino, which complies with DIN 5033 (1992). It is calibrated using tiles provided by the manufacturer for each specific instrument. The illumination is of ring-type at 45° to the normal to the sample. The detection uses a 4° aperture sensor directed along the normal to the sample, and requires a sample area of 4 mm radius. The illuminated area is equal to the measured area, which means there will be some edge effects. Its repeatability is given by the manufacturer to be 0.03 ΔE*ab.

The samples in Series 1 and 2 were measured in these d/0 and 45/0 instruments. Reflectance factor values were recorded at every 10 nm in the wavelength range of the respective instrument, and they were used to calculate measured inter-instrument differences and for comparison with simulations. All conversions from measured reflectance spectra to L*a*b* were done using the color weighting functions for D65 illuminant and 10° observer.

Several external factors can influence the measurements. The most important are the measurement background, the instrument calibration and the inhomogeneity of the paper sample. Special care has to be taken to minimize these factors. This facilitates the isolation of the effects of the instrument geometry for the present study, but it also illustrates the error made when not recognizing and minimizing these external factors. The following three points were employed in this work.
A highly absorbing black matte surface was used as background – the same in both instruments. The measured reflectance factor for the background is about 0.05, and the spectral variation is shown in Fig 2.

The calibration of the instruments was done with the instrument-specific tiles and routines supplied by the manufacturer (the 45° devices must unfortunately be calibrated individually – a tile from another instrument of the same type from the same manufacturer gives miscalibration).

A set of five measurements over different random spots was averaged to reduce the effect of the sample inhomogeneity.

Simulations

The radiative transfer based light scattering simulation tool DORT2002 (Edström 2005) was set up to simulate the standardized d/0 and 45/0 reflectance measurements. In addition to this, the different instruments treat UV differently (illumination, calibration and filtering), but such effects were avoided in this work by using non-fluorescent samples.

Simulated inter-instrument differences

Fig 3 shows the simulated difference in \( R_0 \), i.e. the reflectance factor from a single sheet (left), and the simulated difference in \( R_\infty \), i.e. reflectance factor from an opaque pad (right). The simulations are made for a range of scattering and absorption coefficients for 30 g/m² and 80 g/m² samples. A single point on the surface thus corresponds to one specific wavelength with those scattering and absorption properties. The inter-instrument differences predicted by the model are clearly of small magnitude; the maximum difference is approximately 0.02 reflectance factor units. Based on Fig 3, several features can be distinguished:

- With high absorption and moderate scattering (I), an instrument with d/0 geometry is predicted to detect higher reflectance factor than an instrument with 45° geometry. This parameter region corresponds to dyed papers.
- For samples with very low scattering and moderate to high absorption, i.e. dark samples (II), the d/0 instrument is predicted to detect higher reflectance factor than the 45° instrument.
- For very light samples (III), the 45° instrument is predicted to detect higher reflectance factor than the 45° instrument.
- The parameter region around minimal absorption and scattering (IV) corresponds to an emulsion or air spray, rather than paper samples, and is thus not relevant in paper applications.

Results

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- The parameter region around minimal absorption and scattering (IV) corresponds to an emulsion or air spray, rather than paper samples, and is thus not relevant in paper applications.
Fig 3. The simulated reflectance differences $\Delta R_0$ (left) and $\Delta R_\infty$ (right) plotted against the range of scattering and absorption coefficients for 30 g/m$^2$ (upper) and 80 g/m$^2$ (lower) samples. The middle area (V corresponding to lightly dyed samples) is where inter-instrument differences are small. The area of low absorption (III – corresponding to white paper) is where d/0 measures larger $R$ than d/0, and the rest of the area (I and II – mostly corresponding to dyed or printed paper) is where d/0 measures larger $R$ than 45/0. The inter-instrument differences $\Delta R$ are clearly small – below 0.02 reflectance factor units.

- The large flat region (V) corresponds to lightly dyed papers. In this region, the inter-instrument difference is very small.
- High transmittance tends to decrease the inter-instrument difference in the case of low absorption since then $\Delta R_0 < \Delta R_\infty$. Very light paper samples with low grammage are thus predicted to result in small inter-instrument differences.

The simulations predict that white plain paper would have the largest inter-instrument differences, and that instruments with 45/0 geometry would detect higher reflectance factor for this type of paper, but that the differences would be smaller for low grammages. As the absorption is increased, the d/0 would eventually detect a higher reflectance factor. This means that white and dyed paper exhibit significantly different characteristics in this respect.

**Measurements**

The measured inter-instrument differences are small, as can be seen in Fig 4, and the dyed samples have smaller differences than the non-dyed ones. Both size and sign of the inter-instrument differences are in agreement with the simulated model predictions, as are the variations with sample characteristics.

Since the inter-instrument differences are so small (it should be noted that they are close to instrument precision, so some irregular variations are likely to be present), they can be better observed by plotting the reflectance factor difference $\Delta R = R_{45/0} - R_{d/0}$.

In Fig 5, $\Delta R$ is presented for the non-dyed samples of 80 g/m$^2$ and 120 g/m$^2$. For these samples, the color difference reaches 0.2 $\Delta E^*_{ab}$. The presence of fillers increases the differences for samples of higher grammage.
Fig 4. Spectral $R_0$ and $R_\infty$ reflectance factors are plotted for both instrument types for a thin blue dyed sample (a) (Series 1, 30 g/m$^2$, 1% dye, 22% fillers), and for a non-dyed sample (b) (Series 2, 80 g/m$^2$, no dye, 30% fillers). The inter-instrument differences are small, but they are larger for the non-dyed sample. This is in accordance with the predictions from the model simulations.

Fig 5. Measured $\Delta R_0$ (left) and $\Delta R_\infty$ (right) for non-dyed samples of grammages 80 g/m$^2$ (upper) and 120 g/m$^2$ (lower) and three different filler contents from Series 2. The inter-instrument differences are small, and generally positive. For the samples of lower grammage, this effect is not present due to influence of transmittance$^4$. The inter-instrument differences are small, generally positive (larger values for 45/0) and approximately instrument difference – what is seen in Fig 5 (a) is therefore mostly the different degree of background influence on different spots, and not instrument differences.

$^4$ In fact, the inhomogeneity of samples of lower grammage and low filler content makes the deviation between the small measurement spots of the 45/0 instrument have the same size as the entire inter-instrument differences.
constant with only small variations over the wavelength range. This is in accordance with the model predictions. Unexpectedly, the $\Delta R_\infty$ difference of the samples decreases in the red wavelength region, and in some cases $\Delta R_\infty$ is negative.

In Fig 6, the measured $\Delta R$ is shown for samples with varying amount of blue dye. $\Delta R$ is generally small, very close to the instrument precision. For the wavelengths corresponding to low absorption of the dye, all samples have $\Delta R > 0$ (45/0 detecting larger values). The dyed samples show similar characteristics with each other, with $\Delta R < 0$ for wavelengths corresponding to high absorption of the dye. The differences are larger in the $R_\infty$ measurements than in $R_0$ for these samples of low thickness. This is all in agreement with the model predictions.

The $\Delta E^*_{ab}$ measure of color difference can also be utilized for illustrating the inter-instrument differences with a single, aggregate value. It should be noted, however, that since this is a weighted spectral average, it gives a measure that may conceal some spectral aspects. In Tables 3 and 4, thin dyed samples exhibit the smallest differences overall in the $R_\infty$ measurement. The same samples also show $\Delta R_\infty < \Delta R_0$. Tables 3 and 4 also show that $\Delta R_0$ is larger for the undyed samples of higher grammage than for the dyed samples of lower grammage. All these experimental findings are in accordance with the model predictions above.

Table 3. $\Delta E^*_{ab}$ differences of Spectrolino from L&W Elrepho of sample Series 1 (30 g/m²).

<table>
<thead>
<tr>
<th>w, g/m²</th>
<th>$R_0$</th>
<th>$R_\infty$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% d</td>
<td>0.05</td>
<td>0.06</td>
</tr>
<tr>
<td>0.33% d</td>
<td>0.03</td>
<td>0.07</td>
</tr>
<tr>
<td>0.67% d</td>
<td>0.04</td>
<td>0.03</td>
</tr>
<tr>
<td>1% d</td>
<td>0.08</td>
<td>0.06</td>
</tr>
</tbody>
</table>

Table 4. $\Delta E^*_{ab}$ differences of Spectrolino from L&W Elrepho of sample Series 2 (0% dye).

<table>
<thead>
<tr>
<th>w, g/m²</th>
<th>$R_0$</th>
<th>$R_\infty$</th>
</tr>
</thead>
<tbody>
<tr>
<td>80</td>
<td>0.19</td>
<td>0.15</td>
</tr>
<tr>
<td>100</td>
<td>0.18</td>
<td>0.15</td>
</tr>
<tr>
<td>120</td>
<td>0.10</td>
<td>0.14</td>
</tr>
</tbody>
</table>
In general, the measured inter-instrument differences are in agreement with the model predictions. The measurement results can be summarized as:

• Dyed paper samples have \( \Delta R > 0 \) (45/0 detecting larger values) in wavelengths corresponding to low absorption of the dye, and \( \Delta R < 0 \) (d/0 detecting larger values) for wavelengths corresponding to high absorption of the dye.

• Very light samples have \( \Delta R > 0 \)

• Samples without dye exhibit the largest instrument differences.

• For thin papers \( \Delta R \) is larger than \( \Delta R_0 \).

• For low absorbing thick papers the inter-instrument difference further increases with the presence of filler.

It should be noted for comparison that external factors, if not carefully minimized, affect the measurements to a much larger extent than the recorded inter-instrument difference caused by instrument geometry. This was tested by doing measurements without following the points described in the Methods section that were designed to minimize such influences. Using different background for the \( R_0 \) measurements caused differences up to 3.5 \( \Delta E^* \). Calibrating a 45/0 instrument with a tile from another instrument of the same type caused differences up to 0.8 \( \Delta E^* \). Differences up to 0.3 \( \Delta E^* \) were obtained when carrying out five measurements on different spots/positions on the samples. In addition to this, it has been shown (Andersson, Norberg 2007) that fluorescence can introduce errors up to 7 \( \Delta E^* \), which is far more significant than the geometrical differences studied in this work. The interested reader can find more on this in Radencic et al. (2008). All these factors were, as mentioned above, minimized in the procedure used in this work.

**Explaining geometry related inter-instrument differences**

Neuman and Edström (2010a, 2010b) explain in some detail what causes anisotropy in the light reflected from turbid media such as paper. The conclusions are based on the fact that the angular distribution of the reflected light depends on the relative contributions to the reflectance from different depths in the medium. If near-surface bulk scattering dominates, relatively more light is reflected in large polar angles, i.e. in angles close to the medium surface. If light reflected from larger depths in the medium dominates the reflectance, relatively more light is reflected in the normal direction of the paper. Consequently, a series of factors applicable to paper can be distinguished:

1. Thin samples are dominated by near-surface bulk scattering since light penetrating the medium further has a high probability of being transmitted. These samples thus reflect more light in angles close to the paper surface.

2. Highly absorbing samples are dominated by near-surface bulk scattering since light penetrating the medium further has a high probability of being absorbed. These samples thus also reflect more light in angles close to the paper surface.

3. Illumination with more light incident in angles close to the medium surface results in the domination of near-surface bulk scattering since the light has to travel larger distance to reach larger depths and thus has a higher probability of being absorbed. This leads to more light being reflected in angles close to the surface.

4. Normally incident illumination results in more light scattering deeper inside the medium since the path to reach a certain depth is shorter with this illumination. This leads to more light being reflected close to the normal direction of the sample.

Every situation or measurement setup constitutes a combination of these four factors and the angular distribution of the reflected light will depend on the relative influence of each of them.

The probability \( P \) of directed light incident in polar angle \( \theta' \) to reach a depth larger than \( b \) in a medium with extinction coefficient \( \sigma \) is:

\[
P(x > b) = \int_{\theta_1}^{\theta_2} \exp(-\sigma x) \, dx' = \exp(-\sigma \, \frac{b}{\cos \theta'})
\]

The characteristic depth where the light intensity is half the intensity of the incident light can be expressed as \( b^{0.5} = \ln(2) \cos \theta' / \sigma \). Changing the angle of incidence \( \theta' \) of the directed illumination therefore changes this characteristic depth. If the illumination is not directed, such as the illumination of the d/0 instrument geometry or a perfectly diffuse illumination, the average of the cosine function over the polar angles of the illumination must be used.

For the d/0 instrument with gloss trap, this becomes

\[
\int_{\theta_1}^{\theta_2} \cos x \, dx' (\theta_2 - \theta_1) = 0.5637, \text{ where the angles are the extension of the gloss trap, and}
\]

the characteristic depth thus corresponds to a directed illumination incident at 55.7°. Since 45° is smaller than this, it can be concluded that the characteristic depth \( b^{0.5} \) is larger for the 45/0 geometry, and consequently that the incident light of this instrument geometry reaches relatively larger depths in a medium than the illumination of the d/0 geometry. The reflected light in the 45/0 geometry, part of which is detected by the instrument, is then
dominated by light coming from larger depths in the paper compared to the d/0 geometry. The d/0 instrument geometry thus has a relatively larger part of near-surface bulk scattering compared to the 45/0 geometry.

According to Neuman’s and Edström’s results described above, the angular distribution of the reflected light differs between the instruments, with more light thus being reflected in polar angles close to the paper surface in the d/0 geometry. This is illustrated in Fig 7 where simulations of the BRDF (bidirectional reflectance distribution function) from a non-absorbing (albedo $a = 1$) and non-transmitting medium with varying illumination is shown. The BRDF is proportional to the reflectance factor and the intensity of the reflected light (Nicodemus 1977).

When absorption or transmittance increases, the relative amount of near-surface bulk scattering also increases. This means that factors 1. and 2. above determine the angular distribution of the reflected light to a larger extent than factors 3. and 4. In Fig 8 (a) the simulated BRDF is shown for a highly absorbing and non-transmitting medium, and in Fig 8 (b) the BRDF is shown for a highly transmitting non-absorbing medium. It can be seen that the angular distribution of the reflected light is affected according to what is expected. That is, since factors 1. and 2. dominate, the relative amount of light reflected in large polar angles increases.

Since the medium is non-absorbing and non-transmitting, the illumination angle is the only factor affecting the angular distribution of the reflected light. It can be seen that the angular dependence is in agreement with the reasoning above. The d/0 illumination results in relatively more light being reflected in larger polar angles while the 45/0 illumination results in a higher reflectance around the normal direction. Since the detector of both instruments is placed in the normal direction, the 45/0 instrument will detect a higher reflectance value in the case of a non-absorbing and non-transmitting sample. It is thus incorrect to interpret the detected reflectance as total or diffuse reflectance, or as independent of instrument geometry.

Fig 7. The BRDF of a non-absorbing and non-transmitting medium with varying illumination. A perfectly diffuse illumination would give a constant BRDF and thus an isotropic intensity of the reflected light. The plotted d/0 illumination (including gloss trap) gives close to isotropic intensity but slightly more light is reflected in large polar angles, which also holds for directed illumination from 60º. The 45/0 illumination results in slightly more light being reflected around the normal direction, an effect that is even more pronounced for directed illumination from 10º. This is in accordance with expectations and model predictions.

Fig 8 also shows that the BRDF is lower for all polar angles when the illumination is incident closer to the normal direction, i.e. when the illumination has a larger characteristic depth. This is because the larger characteristic depth results in a smaller probability of light exiting the medium at all, since the light is more likely to be transmitted or
absorbed. Due to this fact, when there is sufficient amount of absorption or transmittance \( (a = 0.95 \text{ or } w = 0.01 \text{ kg/m}^2) \), the d/0 instrument illumination (including gloss trap) will detect a higher reflectance value than the 45/0 instrument illumination for all polar angles, and consequently higher reflectance also at the detector location at 0º.

**Discussion**

The d/0 and 45/0 instrument geometries studied in this work vary in detection and illumination conditions. The angular distribution of the illumination differs significantly between these, and this causes the reflectance factor detected by the respective instrument to differ. This will in turn yield different color values or scattering and absorption parameters for identical samples, which is obviously not desired. The effects of instrument geometry when external factors are minimized can be explained. External factors such as the sample background, the instrument calibration and the sample inhomogeneity affect the inter-instrument differences far more (up to 4-5 \( \Delta E_{ab} \)) than the instrument geometry (of order 0.1 \( \Delta E_{ab} \)), but such effects can be minimized by proper routines. This is summarized in Table 5. Fluorescence can affect the inter-instrument differences even more (up to 7 \( \Delta E_{ab} \)). This can currently only be eliminated by using samples with a minimum of fluorescence, and a general solution calls for standardization of the UV content of the illumination, of calibration routines, and of the use of various filters. The gloss, or specular reflections from the samples, is on the other hand inarguably a factor that firstly will affect the measurements, and secondly, cannot be eliminated. The uncoated samples used here, have rough surfaces and due to this, specular reflections will be included in the measurements. Furthermore, the surface reflection from uncoated paper is very complex, and will likely be handled differently by the two instruments due to the different geometries and designs of the gloss traps. However, in order to reduce the influence of gloss, the samples used here have low gloss values (with an average of 1.0 – 2.9, see section on paper samples). Although gloss is present, it is reasonable to assume that its contribution is small compared to the bulk scattering. No polarization filters were used in the measurements.

There are obviously several color measurement instrumentation error mechanisms which are not considered in this study. Among these are detector linearity, spectral calibration, spectral bandwidth treatment, translucence phenomena and other edge effects. These, and many others are more thoroughly described elsewhere (Spooner 1994, Berns 2000). Moreover, it should be noted that this study uses only measurements on unprinted uncoated papers. Printed samples, glossier samples and larger specular reflection from rough surfaces will for instance put other demands on the gloss trap, spectral calibration and the linearity.

**Table 5. Causes of inter-instrument differences and size of error.**

<table>
<thead>
<tr>
<th>Cause of error</th>
<th>Size of error</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluorescence and gloss</td>
<td>7</td>
</tr>
<tr>
<td>Using different background for the ( R_s ) measurements</td>
<td>3.5</td>
</tr>
<tr>
<td>Calibrating a 45/0 instrument with a tile from another instrument of the same type</td>
<td>0.8</td>
</tr>
<tr>
<td>Sample inhomogeneity and small measurement spot of the 45/0 instrument</td>
<td>0.3</td>
</tr>
<tr>
<td>Instrument geometry</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Using the KM model for interpretation of measurement data is a part of the problem with inter-instrument differences when calculating a paper’s light scattering properties, since it does not recognize the anisotropy (variations in the angular distribution) of the reflected light. An angular resolved model is needed to deal with the situation, and radiative transfer models like DORT2002 can account for geometrical factors in the interpretation of measurement data, which is the key to explain the effect of instrument geometry on inter-instrument differences. This makes it possible to compare different measurement situations objectively, and it recognizes the ubiquitous anisotropy of the reflected light.

A number of results were predicted with DORT2002, and they were confirmed by measurements. The inter-instrument differences when external factors are minimized are small, very close to the instrument precision. Instruments of 45/0 type will detect higher reflectance factor values than d/0 instruments for highly scattering and nearly opaque samples, while the opposite holds for absorbing or translucent samples. The inter-instrument differences are larger for thicker samples, and increase when absorption is negligible and increase further when fillers are present. Knowledge on how near-surface bulk scattering introduces anisotropy of the reflected light makes it possible to predict how this affects detector readings for different instrument geometries. Instruments of 45/0 type will always have relatively more light reaching a certain depth of the sample, but this will
affect detector readings differently depending on sample characteristics. The geometry related inter-instrument differences can be explained as follows.

- Low absorption and low transmittance results in a higher detector reading for 45°/0 than for d/0 instrument geometry. This is because anisotropy causes relatively more light to reflect in near-normal angles (towards the detector) with the illumination of the 45°/0 geometry (factors 3-4 above dominate, see Fig 7).
- Higher absorption or transmittance results in a lower detector reading for 45°/0 than for d/0 instrument geometry. This is because less light is being reflected altogether for the deeper-reaching 45°/0 illumination since more will be absorbed or transmitted (factors 1-2 above dominate, see Fig 8).

These results are in accordance with the theoretical predictions and measurements presented in this work.

Conclusions

A number of paper samples with different properties were examined with the L&W Elrepho d/0 and the Spectrolino 45°/0 instruments. The external factors that influence the measurements were studied, and a methodology for their minimization was presented. The experimental measurements showed that the geometry related inter-instrument difference when all external factors are minimized is small and of order 0.1 ΔE*ab.

It was also observed that the 45°/0 instrument detects higher reflectance from paper samples with negligible absorption and transmittance. When there is considerable absorption (dyed samples) or transmittance (thin samples), the d/0 instrument detects higher reflectance. The physical mechanism behind this behavior was studied and explained, using the DORT2002 radiative transfer model. The measured inter-instrument differences were also compared with the model predictions. The model/measurement agreement was satisfactory as the characteristic behavior was captured in almost all cases. Some variations are to be expected, since the measured differences are close to the instrument precision.

The literature has many statements that 45°/0 and d/0 instruments do differ, and this is usually supported by measurements. Results have not yet been presented that actually explain and quantify the sources of these differences, but this is now done. This work shows the size of a number of external factors, and shows how to minimize them to reduce the inter-instrument differences to a very low level. Further, this work shows how to use the DORT2002 model to predict accurately the remaining differences, which are caused by instrument geometry. This is confirmed by measurements, and the differences are mapped against sample properties. Finally, supported by both simulations and measurements, the physical mechanism behind the inter-instrument differences is explained, showing how absorption and transmittance cause anisotropy in the reflected light and thereby affect detector readings differently. This new understanding is important for facilitating accurate data exchange between the paper and graphic arts industries, but also for interpretation of reflectance measurements in general. This work also illustrates the usefulness of higher order models like DORT2002 for understanding and explaining phenomena not accounted for in simpler models.

Acknowledgements

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Literature


DIN 5033 (1992): Colorimetry; spectrophotometric method, Deutsches Institut für Normung e. V.


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Paper IV
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 goniospectrophotometric 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 anisotropies 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 goniospectrophotometer at Joensuu University, Finland. It has a halo- gen 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 goniospectrophotometer is calibrated by adjusting its readings for the same illumination and detection angles.

The samples are then measured in the goniospectrophotometer 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/m².

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 ShopperRiegler numbers 18° and 23° SR respectively. Sheets having approximate grammage 30 g/m² 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-6GGL 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>
<thead>
<tr>
<th>Sample</th>
<th>Dye content</th>
<th>Grammage [g/m²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>zero</td>
<td>92.25</td>
</tr>
<tr>
<td>2</td>
<td>zero</td>
<td>31.80</td>
</tr>
<tr>
<td>3</td>
<td>low</td>
<td>31.69</td>
</tr>
<tr>
<td>4</td>
<td>medium</td>
<td>32.54</td>
</tr>
<tr>
<td>5</td>
<td>high</td>
<td>32.27</td>
</tr>
</tbody>
</table>

Table 1. Paper samples

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

$$
\frac{dI(s, \Theta, \Phi)}{ds} = \sigma_g \left[-I(s, \Theta, \Phi) + S \right],
$$

(1)

where $I(s, \Theta, \Phi)$ is intensity at depth $s$ at polar angle $\Theta$ and azimuthal angle $\Phi$. Here $\sigma_g$ is the extinction coefficient and $S$ is a source function. The extinction coefficient is the sum of the scattering and absorption coefficients $\sigma_s$ and $\sigma_a$. The source function accounts for light scattered to $\Theta, \Phi$ at depth $s$ from all other angles. It can be written

$$
S = \frac{a}{4\pi} \int_{4\pi} p(\cos\Theta)I(s, \Theta, \Phi)d\omega,
$$

(2)

where $a$ is the single scattering albedo defined as $a = \sigma_a / (\sigma_a + \sigma_s)$, $\omega$ is solid angle and $p(\cos\Theta)$ is the phase function describing the angular distribution of each single scattering. Here $\Theta$ 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 gonio photometer 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 d0/0 instrument geometry [23]. This instrument records the d0 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 $\sigma_g$ and $\sigma_a$ 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 $\sigma_g$ and $\sigma_a$ 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/m² 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 $C^* = (\vert a^* \vert^2 + \vert b^* \vert^2)^{1/2}$ 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 $\Delta E^*_{ab}$ [25] and defined as $\Delta E^*_{ab} = (\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2)^{1/2}$. 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.

### 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-

---

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

<table>
<thead>
<tr>
<th>Sample</th>
<th>Max. meas. $\Delta E^*_{ab}$</th>
<th>Max. sim. $\Delta E^*_{ab}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>15.5</td>
<td>11.9</td>
</tr>
<tr>
<td>2</td>
<td>19.4</td>
<td>25.5</td>
</tr>
<tr>
<td>3</td>
<td>19.7</td>
<td>27.3</td>
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<tr>
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<td>21.4</td>
<td>28.7</td>
</tr>
<tr>
<td>5</td>
<td>23.9</td>
<td>30.5</td>
</tr>
</tbody>
</table>
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.
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/m² sample than for the 90 g/m² sample. This is due to the higher transmittance of the 30 g/m² 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
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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References


**Author Biography**

Magnus Neuman received his MSc in Engineering Physics from Umeå University (2005). Since 2009 he is a postgraduate student at Mid Sweden University and a part of PaperOpt, a nation-wide paper optics research project in Sweden. His research focuses on the development and application of radiative transfer models in paper and print.
Paper V
Point spreading in turbid media with anisotropic single scattering

Magnus Neuman, Ludovic G. Coppel, and Per Edström

Abstract: Point spreading is investigated using general radiative transfer theory. We find that the single scattering anisotropy plays a significant role for point spreading together with the medium mean free path, single scattering albedo and thickness. When forward scattering dominates, the light will on average undergo more scattering events to give a specific optical response in reflectance measurements. This will significantly increase point spreading if the medium is low absorbing with large mean free path. Any fundamental and generic model of point spreading must capture the dependence on all of these medium characteristics.

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OCIS codes: (030.5620) Radiative transfer; (100.2810) Halftone image reproduction; (170.3660) Light propagation in tissues; (290.2558) Forward scattering; (290.4210) Multiple scattering; (290.7050) Turbid media.

References and links

1. Introduction

The point spread function (PSF) describes the response of a medium to a point light source. The lateral distribution $D(x, y)$ of the reflected light is obtained by integrating the PSF and the illumination $I$ over the reflecting medium, that is

$$D(x, y) = \int_{-\Delta x}^{\Delta x} \int_{-\Delta y}^{\Delta y} \text{PSF}(x, y; u, v) I(u, v) du dv$$

(1)

where $\Delta x$ and $\Delta y$ delimit the medium. In the present work we deal with laterally homogeneous turbid media with point source illumination. The PSF is then isotropic, i.e., it depends on radial distance only, and the reflected light is given by $D(r) = \text{PSF}(r) I$ if the illumination is incident at radial coordinate $r = 0$.

The PSF is a medium characteristic and is related to the scattering and absorbing properties of the medium. Several attempts have been made to model the PSF or its Fourier transform, the modulation transfer function (MTF). Oittinen [1] proposed a Kubelka–Munk (KM) [2] based model of the PSF of paper substrates. Engeldrum and Pridham [3] found that Oittinen’s model poorly described layered substrates and that it described plain papers such as newsprint well only at low spatial frequencies. Arney et al. [4] showed that the Oittinen model overestimated the effect of absorption on the PSF, and proposed a simple model based only on the KM scattering coefficient. Gustavson [5] and Emmel [6] used an exponential approximation of the PSF. Mourad [7] considered lateral fluxes in a KM framework leading to a much more complicated form of the KM based PSF that better fit experimental data from Arney et al. [4].

The PSF is closely connected to the phenomenon known as optical dot gain, when printed dots are perceived larger than their actual physical size due to lateral spreading of light. This is important in graphic arts where continuous tones are reproduced through halftoning using dots varying in size and spacing. Optical dot gain then makes the printed image appear darker than expected, which has to be compensated for. Point spreading is also important in computer rendering for a realistic appearance of rendered images [8], and in optical tomography where light scattering is used for medical diagnosis [9].

General radiative transfer (RT) theory can also be used to study the PSF and optical dot gain of a medium. While the KM model approximates the light intensity within a medium with two diffuse fluxes, general RT theory is angle resolved. A basic reference for RT is Chandrasekhar [10]. Considering light of a single wavelength, the familiar RT equation can be stated as

$$\frac{df(x, y, z; \theta, \phi)}{ds} = \sigma_e [I(x, y, z; \theta, \phi) + S].$$

(2)

The symbols have their usual meaning; $I(x, y, z; \theta, \phi)$ is intensity at position $(x, y, z)$ at polar angle $\theta$ and azimuthal angle $\phi$, $s = (x^2 + y^2 + z^2)^{1/2}$ is distance, $\sigma_e$ is the extinction coefficient
and $S$ is a source function. The extinction coefficient is the sum of the scattering and absorption coefficients $\sigma_s$ and $\sigma_a$ and also the inverse of the mean free path $\ell_e$. The source function accounts for light scattered to $\theta, \phi$ at position $(x,y,z)$ from all other directions. It can be written

$$S = a \frac{\alpha}{4\pi} \int_{4\pi} p(\cos \Theta) f(x,y,z; \theta, \phi) d\omega,$$

where $a$ is the single scattering albedo defined as $a = \sigma_s / (\sigma_s + \sigma_a)$, $\omega$ is solid angle and $p(\cos \Theta)$ is the phase function. Here $\Theta$ is the angle between the directions of the incident and scattered light. The phase function describes the angular distribution of the single scattering process. A commonly used phase function is the Henyey–Greenstein (HG) phase function [11] which contains a single parameter, the asymmetry factor $g$. It ranges from $-1$ to $1$ with $g = -1$ meaning complete back scattering, $g = 0$ isotropic scattering and $g = 1$ complete forward scattering. Also, $g$ is the average of the cosine of the scattering angle and the first moment in an expansion of any phase function. For organic materials such as paper it has been shown that $g$ has values in the interval $0.6-0.9$ approximately [12–14].

We can characterize a turbid medium with the albedo $a$, the mean free path $\ell_e$ and the asymmetry factor $g$. These medium characteristics are independent of position in a homogeneous medium but normally vary between wavelengths. Equation (2) can then be solved for each wavelength if the wavelengths are independent. This is not the case in fluorescing media, but in the present work we do not consider fluorescence.

Considering only one spatial coordinate Eq. (2) becomes a 3-dimensional problem and can be solved numerically using e.g. the method of discrete ordinates (DORT) and a Fourier expansion of the azimuthal dependence [15]. In the present work we are interested in laterally resolved (5-dimensional) radiative transfer and we choose to use Monte Carlo simulations since this problem has not been solved using DORT-methods. Coppol et al. [16] presented an open source Monte Carlo model called Open PaperOpt (available at http://openpaperopt.sourceforge.net), adapted to simulations of light scattering in paper and print that is suitable for the problem at hand.

Previous work on point spreading using general RT theory includes Gustavson [5] who developed an approximate Monte Carlo model of radiative transfer in turbid media to study point spreading, but considered isotropic single scattering only. Furthermore, Sormaz et al. [17] presented a method to speed up Monte Carlo simulations and used optical dot gain as an illustrating example. Chen et al. [18] compared measurements of transmittance through paper with the HG phase function, in this case representing the angle resolved transmittance. This is not correct if light can be multiply scattered, which is the case for paper media. When multiple scattering is considered the framework of general RT theory is necessary since the KM approach to the radiative transfer problem has been shown insufficient [19, 20].

The general purpose of the present work is to investigate point spreading using the framework of RT theory, which has not previously been done exhaustively. More specifically we want to determine how the point spreading is related to the medium parameters $a, \ell_e$ and $g$. In this way we can indicate if this dependence can be captured by a simplified model and give important guidelines when developing simplified models of point spreading.

### 2. Method

#### 2.1. Material

Relevant numerical values for the medium parameters $\ell_e$ and $a$ are assessed using a set of paper samples. We choose two lightly dyed and two non-dyed 30 g/m² paper samples with and without fillers, giving four samples in total. Samples varying in filler content differ in mean free path $\ell_e$ since the addition of fillers significantly increases scattering, thus decreasing the
mean free path. The addition of a blue dye increases absorption, thus decreasing the albedo, in the wavelength interval 550–700 nm approximately, and the effect of increased absorption on point spreading can be investigated by studying wavelengths in this interval. No samples contain fluorescent whitening agents. We denote the samples M1–M4, where M1 contains no dye or filler, M2 contains dye but no filler, M3 contains no dye but filler and M4 contains both dye and filler.

2.2. Estimation of medium parameters using DORT simulations

The paper industry uses standardized reflectance measurements to assess the reflectance factor in d/0 geometry (diffuse illumination and detection in the normal direction) [21]. We use these measurements to determine $\ell_e$ and $a$ by measuring the reflectance factor from a single sheet and an opaque pad of paper sheets. We then get a well-posed optimization problem that can be solved for $\ell_e$ and $a$, e.g., by using the RT based DORT2002 model [15, 22] (freely available). This model can accurately describe the illumination and detection conditions of the d/0 instrument. The asymmetry factor $g$ is varied from 0 to 0.8 in steps of 0.2, and the inverse RT problem is solved for each of these $g$ values. We include $g = 0$ since this is an assumption in the KM model. Each parameter setup $[a, \ell_e, g]$ will then give the same optical response in the d/0 instrument for the particular medium studied, despite the variations in $g$, and the medium thus has the same scattering power irrespective of the $g$ value.

2.3. Monte Carlo simulations of the PSF

When we have the parameter setup $[a, \ell_e, g]$ we can estimate the PSF of a medium with a given thickness $t$ through Monte Carlo simulations. We do simulations using both the thickness $t$ corresponding to 30 g/m$^2$ paper and an opaque medium where $t \to \infty$ in order to study the influence of transmittance on the PSF. The thickness of the paper samples is measured with a micrometer and found to be 65 \( \mu \)m. We simulate illumination incident normally on a point and to minimize noise 10$^8$ wave packets are simulated in each run. We choose the wavelength of light that is most heavily absorbed by the blue dye (620 nm) to represent the four media M1–M4.

The PSF is obtained by averaging the simulation data over azimuthal angle. We then introduce a single number metric $r$ to represent point spreading. This metric is the mean radial distance a wave packet travels before exiting the medium, i.e.,

$$r = \left[ \sum_i \text{PSF}(r_i) \right]^{-1} \sum_i r_i \text{PSF}(r_i).$$

The metric we introduce here is in the distance domain, as opposed to the frequency domain metric $k_p$ used by other authors and introduced by Arney [4], which is the frequency where $MTF = 0.5$. A distance domain metric is more useful when dealing with point spreading since the frequency resolution of the reflected light then is of minor interest.

Furthermore, we calculate using the Monte Carlo simulations the mean number of scattering events that a wave packet undergoes before leaving a medium. This allows for important conclusions to be drawn about the dependence of point spreading on the medium characteristics.

3. Results

3.1. Medium parameters

Table 1 shows the values of $a$ and $\ell_e$ obtained from d/0 measurements when $g$ is varied. As expected M2 and M4 have the lowest albedos and M3 and M4 have the shortest mean free path. We can see that the mean free path decreases as $g$ increases. This can be understood intuitively.
since if the light is scattered more in the forward direction when it impinges on the medium surface, the medium must be highly scattering in order to reflect the measured amount of light towards the detector. In this way the scattering power is the same irrespective of the \( g \) value.

<table>
<thead>
<tr>
<th>( g )</th>
<th>( a )</th>
<th>( \ell_e ) [( \mu m )]</th>
<th>( a )</th>
<th>( \ell_e ) [( \mu m )]</th>
<th>( a )</th>
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</tbody>
</table>

3.2. Point spread simulations

The data in Table 1 is used in Monte Carlo simulations with the Open PaperOpt model. Fig. 1 shows how our PSF metric \( \tau \) varies with \( g \) for the different media and for the two different thicknesses. It can be seen that \( \tau \) increases with \( g \) for all media. The high albedo medium with large mean free path (M1) has the largest mean radial distance \( \tau \). The low albedo medium with short mean free path (M4) has the smallest \( \tau \). We can also see that \( \tau \) is larger for the opaque media, with the most noticeable difference for M1. Hence, transmittance can obviously have a significant effect on point spreading. We can thus conclude that point spreading, as represented by \( \tau \), depends on asymmetry factor, albedo, mean free path and medium thickness.

![Fig. 1. Point spreading as represented by \( \tau \) for two thicknesses of media M1–4 when \( g \) is varied. Point spreading increases with \( g \) for all media. M1 (high albedo and large mean free path) has the largest point spreading and the most noticeable difference when varying the thickness. M4 (low albedo and short mean free path) has the smallest point spreading.](image1)

Figure 2 shows the average number of scattering events that the wave packets undergo before exiting the medium. It can be seen that increasing \( g \) increases the number of scattering events. This holds for both thin and opaque media, but when the mean free path \( \ell_e \) is large (as for M1 and M2) the light is scattered fewer times in thin media. When the effect of transmittance is...
eliminated (Fig. 2(b)) there is only a small difference in the number of scattering events when varying the mean free path (M1 vs. M3 and M2 vs. M4).

Based on these observations we can relate point spreading to the scattering properties of a medium. Media with high albedo (M1 and M3) have a similar contribution to the reflectance from different scattering orders. Media with low albedo (M2 and M4) are also similar in this respect. This means that the point spreading is determined by the distance that the multiply scattered light can travel. This distance is larger if the mean free path is large and absorption is low. A medium with high albedo and large mean free path will thus give the largest point spreading, and it will increase further if light is scattered more in the forward direction.

4. Discussion and conclusions

We have shown that the asymmetry factor \( g \) plays a significant role in point spreading together with the albedo, mean free path and thickness of a medium. Given an optical response from a medium, assumptions about the \( g \) value will greatly alter the point spreading. Any model for point spreading must take these medium parameters into account and describe their relative influence in order to based on physics rather than ad hoc assumptions. Thus, models of point spreading cannot be single parameter models. A generic model of point spreading can be on the form

\[
PSF(r) = \sum_i \alpha_i \exp(-\beta_i r)
\]

where the constants \( \alpha_i \) and \( \beta_i \) can be determined from measurements or simulations and thereby related to \( a, \ell_e \) and \( g \). This could be a topic for future work.

This is of importance for example when predicting the optical dot gain to adjust halftone prints. By using a model based on fundamental knowledge it can be generic in the sense that the dot gain of any paper substrate can be estimated. Furthermore, a thorough understanding of lateral diffusion of light is important for fields such as image rendering and optical tomography.

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