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Impact of Aging and Recycling on Optical Properties of Cardboard for Circular Economy

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Abstract: This study investigates the effects of aging and recycling on the optical properties of paperboard, which is key to advancing circular economy practices in packaging. Methods included deinking flotation of cardboard made from sea algae and eco-conventional cardboard of unexposed and exposed samples in a xenon test chamber. Optical measurements were performed on the obtained laboratory paper sheets. Measurements for the chromatic coefficients ΔL*, Δa* and Δb*, as well as the CIE whiteness from comparison of the fluorescent component in the cardboard, were carried out under two light sources, D65 and UV. Regression analysis was used to quantify the statistical significance of these changes over time, i.e., in the aging process. The results revealed significant effects of both aging and recycling on the chromatic coefficients, with ΔL* and Δa* decreasing, while Δb* initially increased before decreasing. The influence of the fluorescent component is reduced by recycling the samples. Opacity measurements showed an initial increase in values that decreased with the aging of the samples, which indicates structural changes in the material. This research contributes to the circular economy by providing insight into the durability and optical properties of recycled cardboard, helping to develop sustainable packaging solutions.

Keywords: circular economy; deinking flotation; optical properties of cardboard; regression

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1. Introduction

The circular economy is important in the paper recycling industry due to environmental, economic and social aspects [1]. It promotes the maximum use of resources while reducing the amount of waste. In the paper production industry, this manifests itself through a reduced need for new raw materials, which preserves forests and reduces deforestation [2]. Furthermore, the consumption of energy and water is reduced compared to the production of paper from raw materials, and the emission of greenhouse gases is also reduced. It is important to emphasize that the abovementioned positive effects for the environment are complemented by economic benefits [3–5]. Paper recycling can be economically viable, reducing production costs and creating new jobs.

The circular economy encourages innovation and new business models, creating additional opportunities for growth and development. Innovative circular economy processes related to paper recycling are applied in paper collection and sorting systems that enable efficient recycling and reuse of materials [6]. Advanced technologies enable the conversion of wastepaper into new products with minimal loss of product quality, minimal recycling of lower-quality paper or paper containing impurities, with maximum use of digital technologies for monitoring and optimizing the recycling process [7–10]. In some countries like Germany, Slovenia, Switzerland and Austria, paper recycling rates exceed 70%, which shows the success of implementing the circular economy [11–14]. Advances in circular economic technology are also reflected in the use of recycled fibers to create new materials, such as bio-composites, which are widely used in the packaging industry and construction [15].

Paper and print aging refers to the gradual physical and chemical deterioration of paper and printed materials over time. Environmental conditions affect the aging or degradation of paper and printing. Exposing the print or paper to light, especially UV radiation, can cause color fading and cellulose degradation, while high temperatures can accelerate chemical reactions that cause the degradation of paper and printed materials [16,17]. Furthermore, moisture in the environment can cause paper to swell and shrink, leading to physical damage, and high humidity can encourage the growth of mold and paper-degrading microorganisms [18–20]. Anthropogenic environmental conditions such as the presence of sulfur dioxide, nitrogen oxides and ozone can also accelerate the chemical deterioration of paper. Acidic papers (e.g., those produced with acidic adhesives) are more susceptible to faster degradation due to the presence of acid that causes cellulose hydrolysis [21,22]. In the literature, aluminum sulfate has negative connotations in accelerating the aging of paper due to the creation of an acidic environment that weakens cellulose fibers over time [23]. Similarly, optical brightening agents (OBAs) cause yellowing as they degrade, especially under exposure to light [24]. Formaldehyde-based resins and certain synthetic polymers can release acids as they break down, leading to brittleness and faster degradation. Even mineral fillers such as kaolin, while beneficial for opacity, can catalyze oxidation, ultimately reducing the durability of the paper under archival conditions [25]. These additives, although they initially improve the properties of the paper, have long-term disadvantages that threaten the stability of the paper. Microorganisms such as bacteria and fungi can cause biological degradation of paper, while insects such as silverfish can physically damage the paper or cardboard and the applied adhesive layer on the paper or cardboard [26,27].

The aging of paper and print is often characterized by color changes, with the paper turning yellow, brown or gray as a result of cellulose and lignin oxidation [28–30]. The strength of the paper decreases and the paper becomes brittle, loses its elasticity and tears easily. Physical changes such as cracks, folds and deformations and phenomena caused by biological activity such as mold and dark spots may appear on paper [31–36]. Due to the influence of light and chemical reactions in the print, the ink fades, or the color of the print changes.

Recycling refers to old paper that has been degraded to a certain degree. The recycling process is designed to remove ink and other contaminants, but the degradation process can affect the quality of the cellulose fibers [37]. It should be noted that with each recycling process, the cellulose fibers shorten and become weaker, which can reduce the strength and quality of the new paper. The success and usability of the process also depend on the ink and chemicals used in printing [38]. Processes such as mixing, washing and chemical treatments during recycling can further degrade paper fibers.

The degradation processes that take place in paper and print are complex and depend on various factors such as paper composition, ink type, environmental conditions, printing techniques and weather factors. In paper processes, cellulose and lignin can react with water, especially at low pH values, causing hydrolysis (decomposition) of cellulose chains [39–42]. The described effect leads to a loss of strength and brittleness of the paper. In the paper of Strlič and collaborators, the influence of the evaporation of organic compounds and hypoxia on the degradation of paper was investigated; it was shown that vapors significantly accelerate the degradation of cellulose, while hypoxia slows down the processes, which is useful for the preservation of archival materials [43]. Bartl et al. emphasize the significant influence of dust on degradation, whereby dust particles facilitate the accumulation of moisture and accelerate oxidative processes [44]. In researching the influence of the environment, Lee and Inabe determined that high temperature and humidity are the key factors that accelerate the aging of naturally used paper [45]. Such results are confirmed by Coppola and colleagues, who point out that controlled conditions extend the life of modern paper materials [46]. Coppola and Modelli investigate the oxidative degradation of recycled and non-recycled paper, noting that recycled paper shows a greater tendency to oxidative processes due to previous cycles of use, which weakens its structure and resistance to further damage [47]. In the context of paper preservation treatments, Bicchieri et al. analyze the influence of gamma radiation, revealing that it can stabilize the paper structure, but with changes that can affect the optical properties [48]. Similar research, such as that of Coppola and colleagues, showed that gamma radiation reduces the presence of microorganisms, but causes changes that affect the long-term stability of paper [49]. Łojewski and co-workers use spectroscopic, chromatographic and chemical methods to follow the progress of degradation in detail, providing an interdisciplinary insight into the structural changes of cellulose during degradation [50]. Tétreault et al. contribute to this topic by developing a model that predicts the degradation of cellulose paper considering factors such as temperature, humidity and the presence of contaminants, providing a basis for the long-term preservation of paper materials [51]. Print degradation can cause ink adsorption into the paper or chemical fixation on the paper surface. The process depends on the type of ink (water-based inks, pigmented inks or toners) [52–55]. Under the influence of light and atmospheric pollutants, inks containing organic pigments can undergo oxidative degradation [56–59]. Print may be subject to color changes due to chemical and physical processes (fading in sunlight). It should not be overlooked that print is subject to physical damage such as scratching, tearing or wear.

The focus of the research is the analysis of the fluorescent component in the paper that can change the perception of colors and the overall visual impression of the paper. Optical brighteners are often added to paper to increase the whiteness and brightness of the paper. By absorbing UV light, they emit blue light, which can change the perception of paper color. Optical whites can affect the visual perception of paper color, so different lighting can cause the paper to look different (daylight versus artificial lighting). The results obtained can be applied to quality control and the development of new products. Understanding fluorescence helps to better control paper quality, ensuring consistency of color and visual appeal across different production runs. Furthermore, information on the influence of fluorescence can help in the development of new paper products with improved optical properties.

2. Materials and Methods

Two cardboard substrates for printing, Shiro Alga Carta and Kromopak, were used as samples. Shiro Alga Carta is a cardboard made from seaweed from the Adriatic Sea (Favini, Italy) (label P2). The use of seaweed as a raw material contributes to the sustainability and ecological suitability of the printing substrate, as it is a renewable raw material whose use contributes to the introduction of balance into the marine ecosystem. Global environmental problems have contributed to increased seasonal marine blooms and beach algae build-up. After drying, the collected algae are ground in a colloidal mill and mixed with FSC fibers [60]. The presence of algae in the cardboard is visible by optical inhomogeneities on the cardboard surface.

Kromopak is an environmentally friendly GC2 cardboard (coated on the front with a cream back) with FSC® and PEFC™ certificates. The FSC® (Forest Stewardship Council) certification ensures that the forest is managed according to strict ecological, social and economic standards, while the PEFC™ (Programme for the Endorsement of Forest Certification) certification guarantees that the wood raw material originates from sustainable forest management. Kromopak is composed of 60% virgin fibers, 30% high-quality post-industrial fibers, unprinted white wood-free paper and 10% coating [61]. It features a three-layer pigment coating on the front and a single-layer coating on the back.

The printing substrates were subjected to an accelerated aging process in a xenon test chamber, Solarbox 1500, manufactured by CO.FO.ME.GRA. according to ISO 12040:1997 [62]. Cardboard samples were exposed to the real atmosphere of a closed space at middle latitudes for 7, 14, 28, 56 and 112 days. Before and after the accelerated aging process, the samples were recycled using the alkaline chemical deinking flotation process according to the INGEDE 11 standard [63]. Samples of laboratory paper sheets were made before and after the exposure process in the chamber and before and after the deinking flotation process on the Rapid Köthen sheet forming device, according to the ISO 5269-2 standard [64]. Table 1 lists the designations of the samples.

Table 1. Designations of samples.

Printing Substrate	Process Stage	Non-Aged	Aged 7 Days	Aged 14 Days	Aged 28 Days	Aged 56 Days	Aged 112 Days
Alga carta	Before flotation	AC_0_BF	AC_7_BF	AC_14_BF	AC_28_BF	AC_56_BF	AC_112BF
	After flotation	AC_0_AF	AC_7_AF	AC_14_AF	AC_28_AF	AC_56_AF	AC_112_AF
Kromopak	Before flotation	K_0_BF	K_7_BF	K_14_BF	K_28_BF	K_56_BF	K_112_BF
	After flotation	K_0_AF	K_7_AF	K_14_AF	K_28_AF	K_56_AF	K_112_AF

Optical measurements were performed on the produced sheets of paper on a specialized spectrophotometer for paper and cardboard, Color Touch 2, manufactured by Technidyne. Table 2 presents the measured parameters along with the corresponding standards.

Table 2. Optical measurements and standards used.

Parameters	Standards
Chromatic coefficients L*, a*, b* (light source D65)	ISO/CIE 11664-1:2019 [65], ISO/CIE 11664-2:20 [66], ISO/CIE 11664-4:2019 [67]
Chromatic coefficients L*, a*, b* (UVEX light source)	ISO/CIE 11664-3:2019 [68]
CIE witness	ISO 11475:2017 [69]
Opacity	ISO 2471:2008 [70], T 425 Om-06 [71]

Fluorescence spectroscopy was used to analyze the fluorescent properties of the paper. This technique enables the identification and quantification of fluorescent components in the paper, that is, the measurement and evaluation of the influence of fluorescence on the overall color and appearance of the paper. Standard light sources D65 and UVEX were used for the measurement. D65 light simulates natural daylight with a coral temperature of around 6500 K and is the standard light source used in the paint industry. A UV light source (UVEX) emits ultraviolet light that is not visible to the naked eye but can cause components in the paper to fluoresce. Fluorescent components in the paper absorb UV light and re-emit it as visible blue light. Paper without optical brighteners reflects light in accordance with its natural color, while a paper with fluorescent components shows increased reflection in the blue spectrum when exposed to UV light. By comparing the measurement results under the D65 and the UV light sources, fluorescent components in different paper samples can be quantified.

In the research, the optical properties of paper were measured and described using CIE Lab color components. CIE Lab is a colorimetric model that quantifies color based on human color perception and uses three components:

*L (lightness): represents brightness and colors.

a*: represents the position of the color on the red–green axis.

b*: represents the position of the color on the yellow–blue axis.

The use of the CIE Lab model enables a more detailed analysis of the color of the paper, due to the complex action of the fluorescent component in the paper.

In addition to the above, the CIE whiteness and opacity of laboratory paper sheets were measured before and after the deinking flotation process. CIE is a metric used to quantify the whiteness of paper under certain lighting conditions. The CIE (International Commission on Illumination) has developed a formula to standardize the measurement of whiteness, allowing consistent comparison of materials. CIE whiteness is usually measured under standard illumination (D65) and a standard observer (CIE 1964 10° standard observer). Opacity is a measure of the amount of light that passes through a material. For paper and cardboard, opacity refers to how much of the print on the back of the sheet can be seen through the paper. High opacity means less light passes through and, therefore, less print is visible on the other side. Opacity is measured by comparing the reflection of a leaf with a black background and the reflection of the same leaf with a white body.

$$\Delta L^* = \beta_0 + \beta_1 \cdot \text{Time} + \beta_2 \cdot \text{Treatment} + \epsilon \quad (1)$$

$$\Delta a^* = \beta_0 + \beta_1 \cdot \text{Time} + \beta_2 \cdot \text{Treatment} + \epsilon \quad (2)$$

$$\Delta b^* = \beta_0 + \beta_1 \cdot \text{Time} + \beta_2 \cdot \text{Treatment} + \epsilon \quad (3)$$

$$\Delta \text{ISO WI}^* = \beta_0 + \beta_1 \cdot \text{Time} + \beta_2 \cdot \text{Treatment} + \epsilon \quad (4)$$

where

- ΔL^* represents the change in L* value (brightness).
- Δa^* represents the change in the position of the color on the red–green axis.
- Δb^* represents the change in the position of the color on the yellow–blue axis.
- ΔISO WI* represents the change in CIE whiteness.
- β_0 is the intercept (constant).
- β_1 is a coefficient that represents the effect of time on the change in brightness.
- β_2 is the coefficient representing the effect of the treatment on the brightness change.
- ϵ is a model error or residual.

The results obtained were interpreted to confirm the knowledge obtained by processing the measured data and to determine the measurement trends and representativeness of the data. Analyzing the impact of recycling on material characteristics provides insight into important information that supports the use of recycled materials, improving sustainability and resource efficiency.

The research used multiple linear regression, which is used to analyze the relationship between one dependent variable and several independent variables. In this analysis, the dependent variables are ΔL, Δa, Δb and ΔCIE WI, while the independent variables are time and treatment. The model is formulated as

$$Y = \beta_0 + \beta_1 \cdot X_1 + \beta_2 \cdot X_2 + \epsilon \quad (5)$$

where

- Y dependent variable (ΔL, Δa, Δb, ΔCIE WI).
- X_1 i X_2 independent variables (Time i Treatment).
- β_0, β_1, β_2 regression coefficients.
- ϵ is the model error.

The regression results provide information on whether time and treatment are statistically significant for each of the dependent variables.

3. Results

Brightening or bleaching agents are often added to recycled materials to improve their appearance. These agents increase the ISO brightness making the material whiter and brighter. Such treatments can be dated by measuring the difference in lightness measured by two standard world sources as explained earlier. ΔL^* is higher for paper made from algae, indicating the absence of substances that have a fluorescent component, in contrast to conventional cardboard where a small presence of such substances, probably optical brighteners, is observed. When studying individual measurements, it can be noticed that the value of L* is higher for conventional cardboard samples. As algae contribute to the inhomogeneity of the paper and have a blue-green tone, they contribute to the reduction of the L* value. So, for example, the L* value for unaged algae cardboard samples before the deinking flotation process is 91.25 for the D 65 light source, while it is 89.72 for the UV source. When studying conventional cardboard, the values are 93.68 for the D65 source and 93.46 for the UV light source (Figure 1).

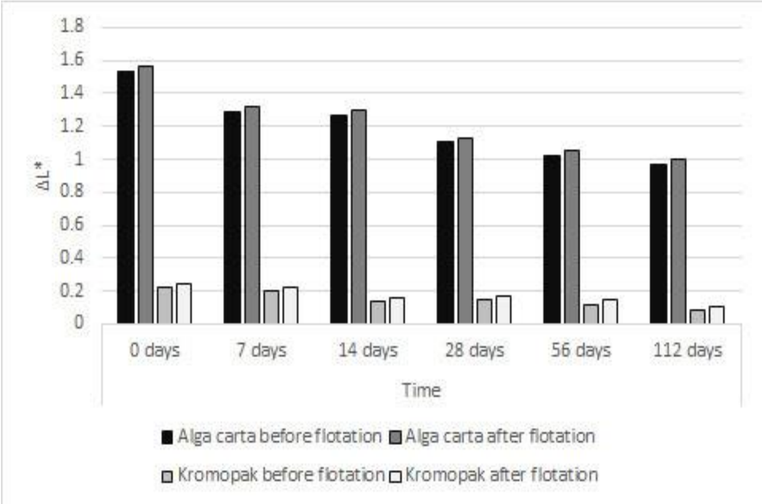

Figure 1. The effect of aging and deinking flotation on the change in the lightness of samples measured with standardized light sources D65 and UV.

During cardboard recycling, deinking procedures are used, which usually contribute to an increase in the ISO brightness value, because the removal of ink particles and impurities results in a cleaner and whiter material as can be seen in Figure 1. The recycling process includes mechanical and chemical treatments that can shorten the cellulose fibers in the cardboard, which can affect how light is absorbed and reflected. Degraded fibers can scatter light differently, which can affect the light.

The aging process in algae cardboard samples and conventional cardboard contributes to chemical decomposition processes: In algae cardboard samples, the process of oxidation and degradation of pigments such as chlorophyll can occur. These changes can lead to a loss of color intensity and a shift towards lighter cardboard colors. Samples of conventional paperboard are mainly composed of cellulose fibers and lignin, which can degrade over time due to exposure to light, heat and oxygen. Lignin tends to turn yellow as it oxidizes, which can reduce the lightness of the board. The aging process also leads to physical changes such as increased brittleness and roughness of the surface, which can contribute to changes in light scattering and affect measurement results.

The aging of cardboard contributes to the processes of oxidation and UV degradation. The UV degradation process can break down lignin and other components in the paperboard, causing color changes that increase a* values (more towards red). Recycling procedures can affect the quality of the recycled fibers, and due to the occasionally lower quality of the fibers, the color may be uneven, which will be further contributed to by the possible presence of impurities. When conventional cardboard is studied under UV light, the fluorescent components are active and emit light in the visible spectrum. The described effect changed the perception of the color of the more environmentally friendly cardboard in this research, increasing a* towards redder shades. When studying paperboard with algae, the algae in the paper may have natural fluorescent components that react to UV light. The described phenomenon resulted in increased color changes towards greener shades due to the natural color of the algae. For the non-aged sample, a* is −0.81. Samples of conventional cardboard and cardboard with algae after the deinking flotation process have a lower a* value, so it is obvious that the recycling process affects the reduction of fluorescent components in the paper (Figure 2). The aging process of paper contributes to the same trend in both samples.

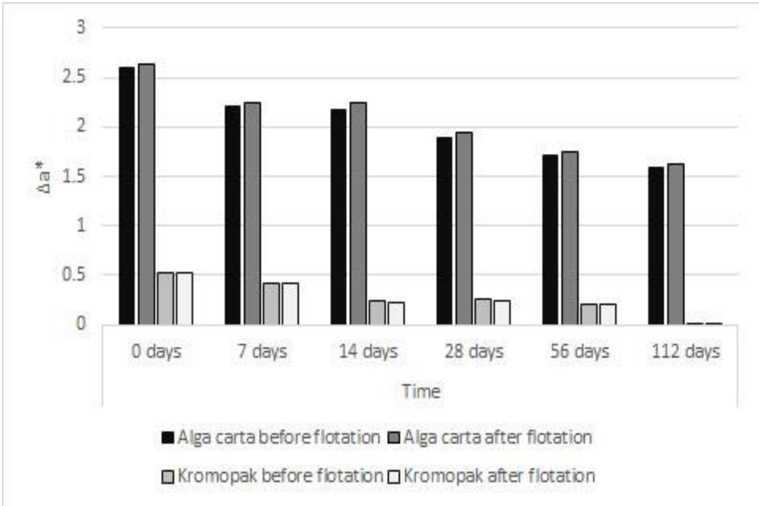

Figure 2. The effect of aging and recycling on the change in the chromatic coefficient Δa* of samples measured with standardized light sources D65 and UV.

Oxidation processes, UV degradation and chemical changes caused by paper aging processes usually result in a change in color towards yellowish or brown tones. These changes affect the chromatic coefficient b*, increasing the b* value towards more positive tones (more yellow) (Figure 3).

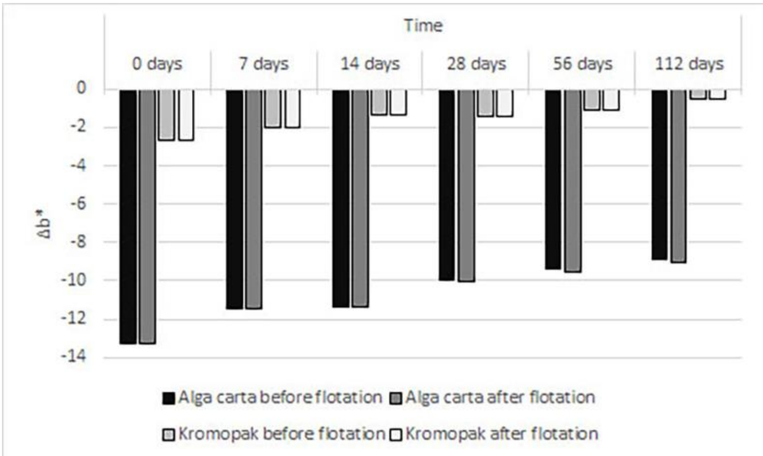

Figure 3. The effect of aging and recycling on the change in the chromatic coefficient Δb* of samples measured with standardized light sources D65 and UV.

The trend of cardboard color change from the blue to the yellow region during the aging process was additionally highlighted by measuring the coefficient b* in the UV region. The aforementioned can be seen in the representation of the differences in the coefficient b* in Figure 3. The differences obtained between the measured values of b* related to the aging process and the source of UV light are most evident in this chromatic coefficient.

Yellowish shades caused by the aging of cardboard in the processes of oxidation and UV degradation also affect the reduction of the CIE whiteness value here, because the cardboard appears less bright and more colored. The measurement of the difference in whiteness under D65 and UV light sources with conventional cardboard and cardboard with algae showed that fluorescent components did not affect the measured values. Variations in measurements are caused by aging or recycling processes. When studying the absolute values of the measurements, it is evident that the CIE whiteness decreases when measured with a UV light source; however, higher values of the difference in whiteness under different light sources were measured for the algae cardboard samples. Algae cardboard may contain natural fluorescent compounds that behave differently under UV light compared to artificial optical brighteners. These compounds may be less stable or less effective under UV light, resulting in lower CIE whiteness and brightness values (Figures 1 and 4).

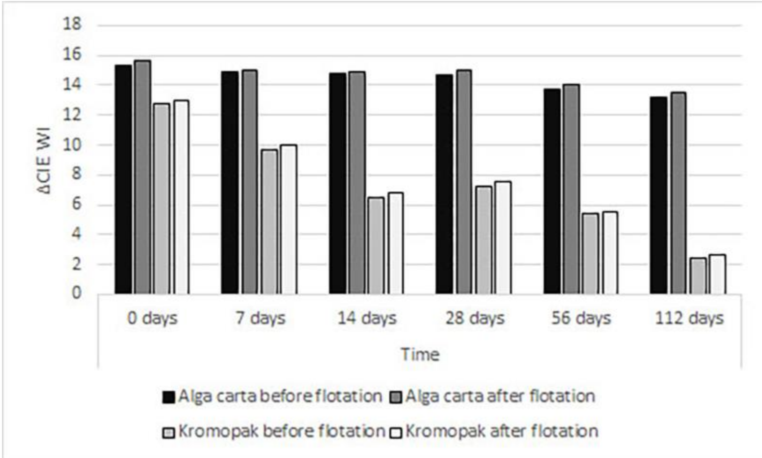

Figure 4. The effect of aging and recycling on the change in the CIE whiteness of samples measured with standardized light sources D65 and UV.

Fluorescent components in algae cardboard are natural substances such as phycobiliproteins, chlorophylls, carboxylic acids, carotenoids, lutein and zeaxanthin, as well as flavonoids and phenolic compounds. Some algae produce carboxylic acid derivatives that may have fluorescent properties. Some flavonoids and phenolic compounds in algae may have fluorescent properties, especially under UV light. Phycocyanin and allophycocyanin absorb light in the red part of the spectrum and emit blue light. Chlorophylls, which include chlorophyll a and b present in all photosynthetic organisms including algae, absorb light in the blue and red part of the spectrum and emit fluorescence in the red part of the spectrum. Phycobiliproteins, like phycoerythrins, absorb light in the green part of the spectrum and emit red light. Carotenoids such as lutein, zeaxanthin and astaxanthin can have a certain degree of fluorescence. Astaxanthin is found in some microalgae and gives them their red color. The contributions of some of these substances influenced the previously mentioned results of a* and b* chromatic coefficients (Figures 2 and 3).

When studying the opacity results, it is visible that in the early stages of aging it increases; this could be due to the creation of microstructural imperfections within the fibers or between the fibers, which can increase light scattering and, thus, increase the opacity (Figure 5). Further aging of the samples can lead to changes in the chemical composition of the cardboard caused by the oxidation process of cellulose fibers, which can change the color and transparency, and the process of lignin degradation, which can reduce the opacity. Over time, the fibers can become more brittle, which can affect the overall density and arrangement of the fibers, affecting the reduction in opacity and contributing to increasing the porosity of the cardboard, which can also reduce its opacity. Recycling has an additional effect on fiber damage through mechanical and chemical means. Cellulose fibers can be shortened, which can reduce opacity because shorter fibers block the passage of light less effectively. The chemicals used in recycling also contribute to fiber damage. In the recycling process, there can often be a partial or complete loss of optical brighteners, fillers and coatings, which can significantly reduce the opacity of the cardboard.

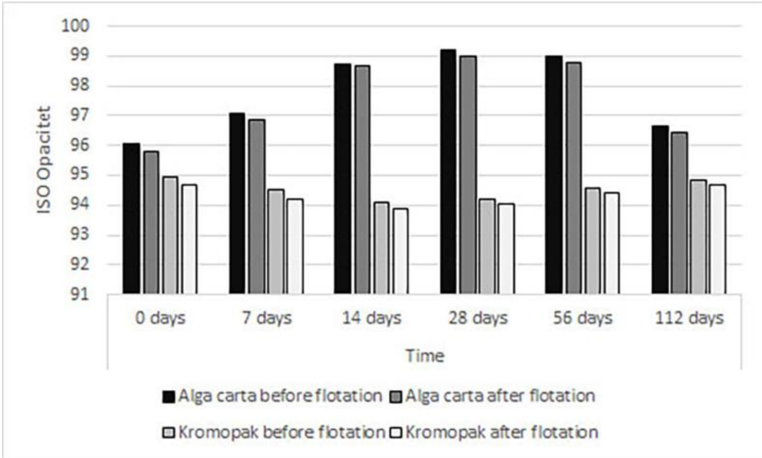

Figure 5. The effect of aging and recycling on the opacity of samples measured with standardized light sources D65.

4. Discussion of Statistical Analysis of Results

Regression is used to model the relationship between one dependent variable (being predicted) and one or more independent variables (being used for prediction). The most used form of regression is linear regression, where it is assumed that there is a linear relationship between the dependent and independent variables. In the context of this research, regression analysis can help in several ways. It can quantify the influence of independent variables, i.e., how aging (time) and the type of cardboard (conventional versus algae) affect the optical properties of cardboard. The regression model can be used to predict future changes in optical properties based on past data. Regression analysis also makes it possible to assess the statistical significance of the results (*p*-values), which helps determine whether the observed changes are truly significant or could be the result of chance.

The value of R-squared (R^2) equal to 0.968 indicates that the model explains 96.8% of the variation in ΔL* (Table 3). This is a very high value, which means that the model fits the data well. The small value of the data probability (F-statistic), 2.00×10^{-7}, indicates that the independent variables are significantly related to the dependent variable. When studying treatment [T.K] with a value of −1.0444, it can be concluded that treatment "K" reduces ΔL* by 1.0444 units compared to treatment "AC". It is significant (*p*-value = 0.000). Furthermore, time affects the processes in such a way that ΔL* decreases by 0.0026 units with each additional day. It is also significant (*p*-value = 0.013).

Table 3. Regression data for ΔL*.

Parameters	Value
R-squared	0.968
Probability (F-statistic)	2.00×10^{-7}
Treatment [T.K]	−1.0444
Time	−0.0026
Probability (Omnibus)	0.027
Durbin–Watson	0.859
Jarque–Bera (JB)	3.331

The Omnibus probability value of 0.027 shows that there is a moderate deviation from normality. Values between 0.05 and 0.01 indicate moderate deviation. The Durbin–Watson value is 0.859: a value close to 2.0 indicates no serial correlation in the residuals, while values below 1.0 indicate a positive serial correlation. The Jarque–Bera (JB) value of the residual normality test of 3.331 and small *p*-values indicate that the residuals may be non-normally distributed (Table 3).

The results for Δa* show an R-squared (R^2) score of 0.973 indicating that the model explains 97.3% of the variation in Δa* (Table 4). This is a very high value, which means that the model fits the data as well as the previously mentioned calculation. Such a conclusion is supplemented with a probability (F-statistic) value of 8.06×10^{-8}, which is a small value and indicates that the independent variables are significantly related to the dependent variable. Such data is important for confirming the interpretation of the results obtained. The treatment [T.K] calculation value of −1.7572 shows that treatment "K" reduces Δa* by 1.7572 units compared to treatment "AC". It is significant (*p*-value = 0.000). The calculation of the time value as −0.0057 shows that Δa* decreases by 0.0057 units with each additional day. It is also significant (*p*-value = 0.002).

Table 4. Regression data for Δa*.

Parameters	**Value**
R-squared	0.973
Probability (F-statistic)	8.06×10^{-8}
Treatment [T.K]	−1.7572
Time	−0.0057
Probability (Omnibus)	0.148
Durbin–Watson	0.967
Jarque–Bera (JB)	1.426

Furthermore, the Omnibus probability value of 0.148 shows that there are no significant deviations from normality (values above 0.05 indicate normality), while the value for the Durbin–Watson parameter of 0.967, which is close to 2.0, indicates that there is no serial correlation in the residuals. A value of 1.426 in the test for the normality of the residuals for the Jarque-Bera (JB) parameter is close to 0, indicating the normality of the residuals (Table 4).

The model explains 97.8% of the variation in Δb*, which is the highest value in this research and shows that the model fits the data well (Table 5). The value of the probability (F-statistic) is also small like L in the earlier examples and is 3.24×10^{-8}, which indicates a significant connection between the independent variables and the dependent variable. When studying the treatment [T.K] parameter value of 9.2122, it can be concluded that treatment "K" increases Δb* by 9.2122 units compared to treatment "AC". It is significant (*p*-value = 0.000). The influence of weather on Δb* shows that Δb* increases by 0.0234 units with each additional day. It is also significant (*p*-value = 0.004) (Figure 5).

Table 5. Regression data for in Δb*.

Parameters	**Value**
R-squared	0.968
Probability (F-statistic)	3.05×10^{-8}
Treatment [T.K]	9.2122
Time	0.0234
Probability (Omnibus)	0.114
Durbin–Watson	1.038
Jarque–Bera (JB)	1.716

Furthermore, the Omnibus probability value of 0.114 shows that there are no significant deviations from normality, while the value for the Durbin–Watson parameter of 1.038, which is close to the value of 2.0, indicates that there is no serial correlation in the residuals. A value of 1.716 in the test for the normality of the residuals for the Jarque–Bera (JB) parameter is close to 0, indicating the normality of the residuals (Table 5).

The model explains 87% of the variation in ΔCIE WI. When the treatments are studied, treatment "K" decreases ΔCIE WI by 7.1077 units compared to treatment "AC" (*p*-value = 0.000), while ΔCIE WI decreases by 0.0460 units with each additional day (*p*-value = 0.007) (Table 6). Furthermore, the data from the Omnibus probability of 0.041 shows that there are some deviations from normality, while the value for the Durbin–Watson parameter of 0.867, which is somewhat lower than the ideal value of 2.0, indicates potential positive serial correlation in the residuals. A value of 3.157 in the test for the normality of the residuals for the Jarque-Bera (JB) parameter indicates some skewness and kurtosis, suggesting that the residuals are not perfectly normal.

Table 6. Regression data for in ΔCIE WI.

Parameters	Value
R-squared	0.87
Probability (F-statistic)	0.000103
Treatment [T.K]	−7.1077
Time	−0.0460
Probability (Omnibus)	0.041
Durbin–Watson	0.867
Jarque–Bera (JB)	3.157

All models have high R-squared values, which means that they describe well the variation in the dependent variables. The independent variables are significant (all *p*-values are very low), meaning that changes in treatment and time are significantly associated with changes in ΔL* and Δb*.

General by studying the effects of the treatment on the measured values, the "K" treatment significantly reduces ΔL* (lightness) and Δa* (redness/greenness), while it significantly increases Δb* (yellowness/blueness) compared to the treatment "AC". Over time, ΔL* (lightness) and Δa* (redness/greenness) decrease, while Δb* (yellowness/blueness) increases. When studying the influence of time on ΔCIE WI (whiteness) it can be noticed that its values decrease. When studying the models, it can be concluded that all models have high R-squared values, which means that they describe very well the variations in the dependent variables. All model parameters are significant, indicating that treatment and time have a significant effect on changes in color and whiteness. From all that has been said, it can be concluded that the "K" treatment and time are important factors in determining changes in color and whiteness and that the "K" treatment tends to reduce lightness and whiteness while increasing the yellowness compared to the "AC" treatment.

5. Conclusions

This research aimed to examine how aging and recycling processes affect the optical properties of paperboard, with a particular focus on paperboard made from algae compared to conventional paperboard. The research included examining the influence of possible fluorescent components in operation on the results of measurements with two light sources: D65 (daylight simulation) and UV light. In the research, a regression analysis was performed to confirm and quantify the changes.

When studying the values of the differences in the chromatic coefficient ΔL* (brightness), a trend of decreasing values with time can be observed, which could probably be attributed to the degradation of the fibers and the chemical changes that cause the darkening of the cardboard. The recycling process further reduces the ΔL* values because the fluorescent components are lost and presumably the fibers become less reflective. The results of the difference in chromatic coefficients Δa* show a decrease in redness over time, probably due to oxidation and other chemical reactions that neutralize the reddish tone of the cardboard. Aging and recycling significantly affect Δb*. In the initial stages of aging, Δb∗ increases due to the very likely degradation of fibers and chemical changes, while in the later stages, it decreases, probably due to the loss of fluorescent components. Algae paperboard showed a different pattern of Δb* change compared to conventional paperboard, which can be attributed to the presence of specific fluorescent components from algae. in the spectral composition of light, and different reactions of natural and synthetic fluorescent materials to UV light. Together, these factors contribute to a reduction in perceived whiteness and brightness when samples are measured under UV light. In the initial stages of aging, the results show that opacity increases probably due to increased roughness and microscopic changes on the paperboard surface. With further aging, the opacity decreases due to the degradation of the fiber structure. When studying the effect of the recycling process on opacity, it can be observed that the opacity changes as the fibers are likely to be damaged and have less ability to block light.

This research contributes to a better understanding of how biological materials and their fluorescent pigments affect the optical properties of cardboard during aging and recycling. An additional contribution is that the developed regression models provide quantitative tools for predicting changes in ΔL*, Δa*, Δb*, ΔCIE WI and opacity during paperboard aging and recycling. The obtained results contribute to the circular economy in several ways. The results obtained from the analysis of optical changes depending on the processes of pattern aging and cardboard recycling can help manufacturers to use and develop materials that retain the desired optical properties for longer. This reduces the need to produce primary materials, which is crucial for a circular economy. Research has shown that recycling reduces the impact of the fluorescent component, which facilitates material reprocessing and reduces the need for resources (optical whiteners) in future recycling cycles. By introducing optimal techniques, such as flotation to remove ink, the need for additional treatments and additives is reduced, thereby improving material efficiency and reducing production costs.

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