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GRAPHIC DOCUMENTS

Permanently bright? Predicting light-induced changes in white paper: Preliminary results

Marie Kern*
Conservation of Works of Art on Paper, Archives and Library Materials, Stuttgart State Academy of Art and Design (ABK Stuttgart)
Stuttgart, Germany
marie.kern@abk-stuttgart.de

Irene Brückle
Conservation of Works of Art on Paper, Archives and Library Materials, Stuttgart State Academy of Art and Design (ABK Stuttgart)
Stuttgart, Germany
irene.brueckle@abk-stuttgart.de

Georg Josef Dietz
Museum of Prints and Drawings (Kupferstichkabinett), National Museums in Berlin (SMB Berlin), Prussian Cultural Heritage Foundation
Berlin, Germany
g.dietz@smb.spk-berlin.de

Ute Henniges
Conservation of Works of Art on Paper, Archives and Library Materials, Stuttgart State Academy of Art and Design (ABK Stuttgart)
Stuttgart, Germany
ute.henniges@abk-stuttgart.de

Fabienne Meyer
Museum of Prints and Drawings (Kupferstichkabinett), National Museums in Berlin (SMB Berlin), Prussian Cultural Heritage Foundation
Berlin, Germany
f.meyer@smb.spk-berlin.de

Thomas Prestel
Archaeometry and Natural Sciences Laboratory, Dresden University of Fine Arts (HfBK Dresden)
Dresden, Germany
prestel@hfbk-dresden.de

Stefan Röhrs
Rathgen Research Laboratory, National Museums in Berlin (SMB Berlin), Prussian Cultural Heritage Foundation
Berlin, Germany
s.roehrs@smb.spk-berlin.de

Giulia Vannucci*
Rathgen Research Laboratory, National Museums in Berlin (SMB Berlin), Prussian Cultural Heritage Foundation
Berlin, Germany
g.vannucci@smb.spk-berlin.de

*Author for correspondence

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Abstract

To differentiate the light sensitivity of different white papers and gauge the capacity of microfading testers (MFTs) to predict their color changes, the light responses of 38 papers were investigated based

INTRODUCTION

Light exposure is a constant preservation issue for works on paper. Because color and contrast are the main features determining readability of these planar objects, color change can cause significant or complete loss of a work's aesthetic value. Although the light sensitivity of paper-based objects receives ample attention in museum practice, those objects are primarily flagged as light-sensitive based on their media despite the fact that their appreciation also depends on the lightness of the paper support. With the exception of lignin-rich and optically brightened papers, preservation guidelines do not differentiate among the different responses of white paper to light, reflecting a lack of systematic empirical studies of light-induced color change in white papers.

There is an increasing interest in measuring the light-sensitivity of works on paper using a microfading tester (MFT) (Whitmore et al. 1999). This in situ dose-response testing of original objects is regularly employed to test colorants on paper, but it has not been optimized to test paper supports, with some early (Bowen et al. 2002) and recent (e.g., Ford and Smith 2011, Ford and Batterham 2014, Kogou et al. 2015, Pesme 2016) exceptions. A challenge also arises in the use of MFT for white papers, which have previously been reported to have an equal or higher stability than BWS 3 (Blue Wool Scale) according to ISO 105:2004 (Ford and Smith 2011), since a high error in measuring Blue Wool references with MFT makes it hard to differentiate materials in that sensitivity range (BWS 3–4). Thus, whether the established MFT (xenon-arc source) and the newer, automated MFT (LED source; Łojewski et al. 2011) are reliable tools for predicting the light responsivity of white paper is unclear, as is whether the two instruments are equally suitable to the task.

Previous research indicated that the aging response and long-term color stability of white paper depends significantly on its key components as well as on potential additives. Next to the cellulosic fiber base, early handmade papers contain calcium carbonate, gelatin, and alum (Barrett and Mosier 1995, Barrett et al. 2016) but also contaminant iron ions (Hon 1975, Niehus et al. 2012, Henniges et al. 2020), and in papers since the 19th century, lignin and rosin sizing are likely to be found; in addition, contemporary papers contain optical brightening agents (OBAs) (Mustalish 2000). Yet so far, only ligneous papers have been extensively researched regarding light exposure effects (as summarized by Paulsson and Parkas

on museum exposure, accelerated aging, and testing with two different MFTs (xenon and LED). According to the preliminary results, a third of the papers were as sensitive as Blue Wool Standards (BWS) 2–3, and aged rag paper and some papers containing optical brightening agents (OBA) were as responsive as ligneous papers; however, OBA papers were not adequately represented by the MFT results. Instrument drift for the two MFTs was found to differ in terms of drift direction, but not intensity, which must be considered when interpreting the results. Visibility testing of MFT spots revealed that spots 0.2 mm in size and with fading below ΔE_{00} 5 will not be visible to most observers. The introduction of UV in MFT seems promising for papers that contain materials sensitive to UV, except OBA-containing papers.

2012, 5596–5598), with far fewer studies examining the effects of iron (Czepiel 1960, Bicchieri and Pepa 1996, Daniels 2002, Niehus et al. 2012).

In this study, we examined the impact of paper composition on the color changes caused by light exposure and compared xenon-MFT and LED-MFT to determine their efficacy in assessments of the light sensitivity of white paper. Our preliminary analyses addressed (a) white papers in indoor exposure, (b) instrumental noise, (c) the visibility of MFT-measured spots, and (d) measurements of OBA-containing paper.

MATERIALS AND METHODS

Four paper sample groups were evaluated: (1) rag papers (pre-industrial papers), (2) wood pulp papers (early industrial papers), (3) papers with OBAs, and (4) filter papers impregnated with key chromophores of aged paper (Rosenau et al. 2011). The papers in each group included typical additives expected to stabilize or destabilize a paper's light response. Papers in groups 1, 2, and 4 were lab-prepared, and papers in group 3 were commercially available papers. Additionally, those in groups 1 and 2 were supplemented by selected historical papers.

Colorimetric measurements were performed after each irradiation according to the graphic industries standard ISO 13655:2009 and using white standard backing (M1 measurement/D50, UV-included mode). The measurements were made using an X-Rite i1Pro UV/VIS spectrophotometer (tungsten light source and UV-LED, ring illumination, 45/0° measuring geometry), with an instrumental error of 0.1 ΔE_{00} (mean of 10 consecutive measurements on white standard) and the average value calculated based on five repeats per spot on three spots per paper.

The **xenon-MFT** was the Oriel Fading Test System (model 80190) equipped with a UV- and IR-filtered xenon arc lamp (Osram 75W Xe) and a sampling head extended with an endoscope camera. The reflected spectra were analyzed using a photodiode array detector (control development, model PDA-512). Fiberglass optics (Oriel P/N 78251 and 78367) were used to transport the light. The diameter of the illuminated spot was evaluated being 330 microns; the test duration was set to 15 min (corresponding to ~1.6–1.7 Mluxh). The instrument was calibrated using a Labsphere Spectralon as the white standard. The **LED-MFT** (Instytut Fotonowy, Cracow, Poland) is equipped with a neutral white LED source. The measurement spot was 0.58 mm. A Fotonowy Fotolon white standard reference (equivalent to the Labsphere Spectralon) was used for calibration. The geometry of the illuminating and receiving probes was 0–45° for both instruments. The illuminance of the xenon-MFT ranges from 6.3–6.6 Mlx with both UV and IR filters in place. With UV introduced, the illuminance ranges reaches values of around 8 Mlx. The illuminance of the LED-MFT ranges from 6.1 to 6.5 Mlx. Illuminance was measured using an Almemo datalogger 8590-9A and a FLA 623 sensor adapted for the measurement of direct light. Spectra were recorded in the wavelength range of 420–735 nm for the xenon-MFT and 400–750 nm for the LED-MFT.

For **indoor light exposure**, three sets of 38 paper samples (Figure 1) were light-exposed under different indoor and museum conditions at the

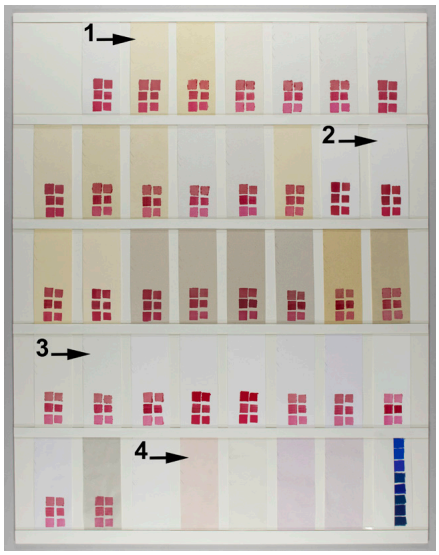

Figure 1. Samples mounted for display at the Kupferstichkabinett, Berlin; paper categories 1–4 are indicated

Museum of Prints and Drawings (Kupferstichkabinett) in Berlin (Table 1), including placement in temporary exhibitions at 50 lux and permanent LED illumination in a separate "light lab" at 100 lux. The samples were framed behind UV-filtering museum glass (Mirogard VSG). As demonstrated by the different rates of fading of ISO Blue Wool Standard references, the total light dose varied between settings and was greatest in the foyer, which featured mixed fluorescent and daylight (350–628 lux), mimicking a gallery-style exposure (Table 1). At ~1 Mlx·h exposure, the foyer setting revealed the first light-induced changes in the papers lying within or above a just noticeable difference (JND) of 1.5 ΔE_{00}. The following observations were based on this setting.

Table 1. BWS 1–3 response to exposure settings (museum exhibition, light lab, museum foyer) expressed as CIELAB ΔE_{00} (measured 20 September 2022) and the respective light doses required for JND (according to Michalski 2018)

	ΔE_{00}			Dose for 1 JND (no UV)
	exhibition	"light lab"	foyer	
BWS 1	1.51 (± 0.26)	6.04 (± 0.38)	7.48 (± 0.42)	0.3 Mlx·h
BWS 2	0.51 (± 0.14)	1.35 (± 0.11)	1.64 (± 0.26)	1 Mlx·h
BWS 3	0.55 (± 0.23)	0.63 (± 0.15)	0.74 (± 0.18)	3 Mlx·h

COLOR CHANGE OF PAPERS DURING MUSEUM FOYER EXPOSURE

Twelve out of 33 tested papers (excluding the chromophore-doped filter papers) displayed a light sensitivity > BWS 3 (Table 2). This placed them within the mid- to lower range of the "sensitive" category (BWS 1–3) as defined in most museum guidelines based on the ISO Blue Wool ratings, e.g., CIE 157:2004 and CEN/TS 16163:2014. As expected, this category included all lignin-rich papers included in the group containing early industrial papers. The presence of rosin sizing (prepared with alum) did not adversely impact the color stability of these papers (Table 2).

In the other paper groups, the majority of non-ligneous paper samples were more stable than BWS 3, with notable exceptions. Even behind UV-filtering glass, some optically brightened papers had a sensitivity between BWS 2 and BWS 3 (Tables 2, 3). However, our results suggest that the amount of OBA present in the paper and the paper composition also have an impact, because at least one OBA-free contemporary paper fell within a similar range and only four out of eight OBA papers were more sensitive than BWS 3 (Table 3). Also, among the group of rag papers, the historical gelatin-sized Whatman paper was the most sensitive (BWS range 1.5–2) as displayed considerable photo-bleaching (Table 2). In the context of conservation practice, this observation showed that some historical handmade papers are prone to undesired photo-bleaching that will impact their benignly aged appearance, causing a color shift.

Within the group of filter papers doped with the three key chromophores that typically form in aging papers, one (DHNQ) was also affected by photo-bleaching, one (DHBQ) by only yellowing, and one (DHAP) darkened slightly. These effects match observations of historical rag papers discolored through light exposure (Dietz et al. 2016).

Table 2. Color changes of paper samples corresponding to < BWS 3 after ca. 1 Mlx·h exposure in the museum foyer (measured 20 September 2022). Samples marked * are lab-made model sheets, components in *italics* are present in minor amounts

Paper type	Non-fibrous components	ΔE_{00}	Sensitivity (BWS range)
pre-industrial papers			
Whatman rag paper, ca. 1801	gelatin, calcium, alum, *iron*	2.94 (± 0.13)	1.5–2
early industrial papers (all contain lignin)			
unbleached hardwood sulfate*		2.48 (± 0.89)	< 2
unbleached hardwood sulfate*	sizing (rosin, alum)	2.42 (± 0.10)	< 2
unbleached hardwood sulfate*	iron	1.60 (± 0.20)	2
unbleached hardwood sulfate*	sizing (rosin, alum), iron	1.49 (± 0.13)	2
low-quality machine paper, ca. 1980	sizing (rosin, alum), kaolin	0.98 (± 0.09)	2.5–3
newsprint (NYT), ca.1990s	sizing (rosin, alum)	2.03 (± 0.04)	< 2
contemporary papers (all contain AKD and starch size)			
fine art watercolor paper, hardwood (Canson Montval)	calcium carbonate, talcum	1.17 (± 0.01)	2.5
raw baryta paper, softwood (Adox Fine Art Baryta)	sizing (rosin, alum), OBA, coating (with gelatin)	1.13 (± 0.03)	2.5–3
office copying paper, softwood (Papyrus Plano Speed)	calcium carbonate, OBA, org. pigments	1.55 (± 0.12)	2
notepad paper, softwood and minor portion of groundwood	calcium carbonate, OBA, org. pigments	1.20 (± 0.05)	2.5
recycling newsprint	calcium carbonate, kaolin, OBA, org. pigments	2.00 (± 0.08)	< 2
chromophore-impregnated filter papers			
plain paper (reference)	-	0.48 (± 0.26)	>3
2,5-dihydroxy-1,4-benzoquinone (DHBQ)	-	4.76 (± 0.15)	1.5
2,5-dihydroxyacetophenone (DHAP)	-	1.32 (± 0.09)	2–2.5
5,8-dihydroxy-1,4-naphthoquininone (DHNQ)	-	4.25 (± 0.16)	1.5
mixture (DHBQ, DHAP & DHNQ)	-	3.04 (± 0.19)	1.5–2

Table 3. All tested contemporary papers and corresponding BWS after ca. 1 Mlx·h exposure in the museum foyer. With the exception of C7 (rosin sized, no fillers), all papers were sized with alkyl ketene dimer (AKD) and contain cationic starch and calcium carbonate fillers and contained calcium carbonate fillers

No.	Paper	Pulp	g/m²	OBA	BWS range
C1	fine art watercolor paper (Hahnemühle "Cézanne")	cotton linters	300	no	< 3–3
C2	fine art watercolor paper (Canson Montval)	bleached hardwood	300	no	2.5
C3	fine art drawing paper (Fabriano Academia Disegno)	bleached hardwood	200	minor	3
C4	fine art inkjet paper (Hahnemühle inkjet Photo Rag)	cotton linters	308	minor	> 3
C5	fine art inkjet paper (Hahnemühle inkjet Photo Rag Bright White)	cotton linters	308	yes	3
C6	fine art drawing paper (Impuls drawing paper)	bleached hardwood	220	yes	> 3
C7	raw baryta paper (Adox Fine Art Baryta)	bleached softwood	190	yes	2.5–3
C8	office copying paper (Papyrus Plano Speed)	bleached softwood	80	yes	2
C9	notepad paper	bleached softwood, some groundwood	80	yes	2.5
C10	recycling newsprint	recycling pulp (incl. groundwood)	45	residue	< 2

MFT INSTRUMENT DRIFT

A further aim of this study was to quantify and identify the origin of instrumental noise and to develop measures aimed at its minimization. This was done by first measuring instrument drift during the MFT runs carried out on the white reference standard (Spectralon or Fotolon) used for the

calibration. The resulting color change, expressed as ΔE_{00}, associated with this MFT run, represented the instrumental noise that accompanied the results of the tested materials. The drift measurements shown in Figures 2 and 3 occurred with both MFTs during optimized procedures, which included a "warm up run" of the lamp and filters for 1 h to allow the working temperature to be reached before the first measurement, a white and dark current calibration before each run, and the selection of suitable filters, i.e., those that produced the least drift. The optimized ΔE_{00} drift values for the xenon-MFT were near zero (between 0.0 and 0.2 ΔE_{00}) but were associated with rather high error bars, indicative of significant variances. There was no preferential hue shift, and the drift occurred primarily in the L* coordinate. The optimized ΔE_{00} with LED-MFT was also very small (between 0.0 and 0.3 ΔE_{00}), with only small variances, but there was a systematic negative drift towards blue ($\Delta b^* < 0$).

The practical considerations of our results are the following: The photo-yellowing of paper ($+\Delta b^*$) during a fading experiment might be identified with more accuracy with the xenon-MFT rather than with the LED-MFT. On the other hand, the ΔL^* in the xenon-MFT is characterized by higher unpredictable variations, which makes it less suited for reading the bleaching ($+\Delta L^*$). Despite these considerations, which for the sake of precision and accuracy are important to be noticed, the absolute numbers in discussion are very small for both instruments. Indeed, very unlikely values of $\Delta E_{00} < 0.4–0.3$ will be considered significant when interpreting light sensitivity. Finally, through drift optimization and quantification, we are able to obtain more solid data for interpretation and to include BW4 in our measurements.

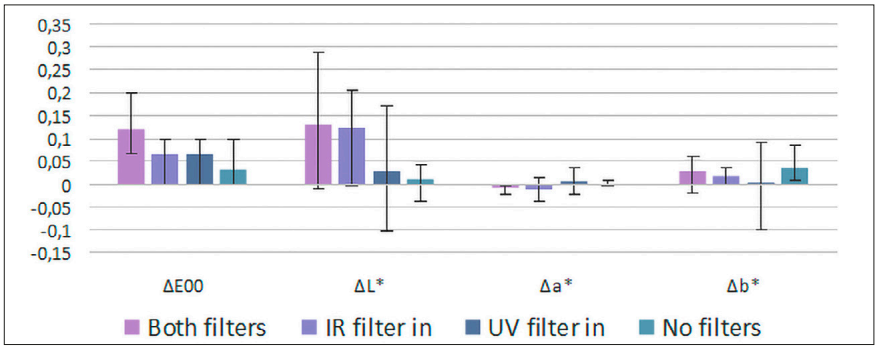

Figure 2. Xenon-MFT lamp drift within 15 min: overall color change (ΔE_{00}) and ΔL^*, Δa^*, Δb^* with different filters and their combinations. Bars indicate the absolute errors associated with the measurements

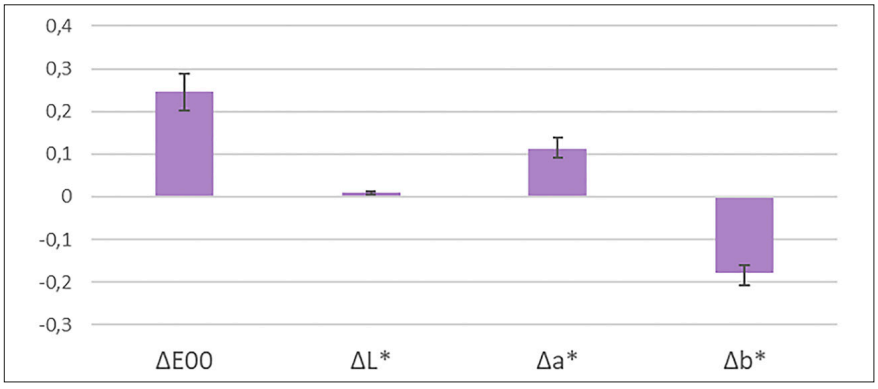

Figure 3. LED-MFT lamp drift within 20 min: overall color change (ΔE_{00}) and ΔL^*, Δa^*, Δb^*. Bars indicate the absolute errors associated with the measurements

VISIBILITY OF MFT-MEASURED SPOTS

When using MFT, it is important to determine a maximum color difference threshold at which irradiation should be terminated, so that the measurement spot on the original object remains invisible. On a macroscopic scale (e.g., observing the whole artwork), the threshold at which color differences become visible (the JND) can lie in the range of $1\Delta E_{00}$ to 3–4 ΔE_{00}, according to Saunders (2020, 58) and supported by the analysis by Pretzel (2008, 762). However, MFT spots are by definition tiny (diameter: ≤ 0.5 mm) and size is a significant factor in the visibility of local discolorations. In the MFT literature, threshold values of $\Delta E_{00} = 3.0$ (Beltran et al. 2021) for microfading spots have been reported, but to the authors' knowledge published data supporting this threshold is lacking.

To test the visibility of MFT spots, samples of different materials and colorants were produced and then microfaded using a xenon-MFT with spot sizes of 0.2 and 0.4 mm. On each sample, a defined color change up to the following color differences was induced: $\Delta E_{00} = 0.3$, 0.5, 1.0, 1.5, 2.0, 2.5, 3.5, 5.0, 7.0, and 10.0. Additional samples without MFT spots were included in the sets. Thirty-one observers (ages: 16–58) who had passed the Ishihara test for the detection of red-green color deficiencies, to ensure that they had normal color vision, were then asked to judge whether the spot was visible. The observers were instructed to locate the MFT spot on the samples on a 3 × 3 grid and were also told that in some samples there might be no spot at all.

The preliminary results indicate that, beyond a color change of $\Delta E_{00} > 5.0$, larger spots (0.4 mm) were visible for > 80% of the observers, while smaller spots (0.2 mm) with $\Delta E_{00} > 5.0$ were detected correctly by > 70%. Accordingly, a termination threshold of $\Delta E_{00} = 3.0$ for MFT on original objects seems a reasonable value, as it enables a cautionary approach that avoids causing visible change.

MEASURING OPTICALLY BRIGHTENED PAPER (XENON-MFT)

Samples belonging to the group of contemporary papers with or without OBAs (Table 3) were faded using two different instrumental setups: with filters (UV & IR = about 6.5 Mlx with 20 µW/lum residual UV) and with the UV filter removed (8 Mlx with 100 µW/lum UV), resulting in a maximum UV value close to the maximum value accepted by most museums but an exposure significantly lower than that produced by unfiltered fluorescent or daylight.

The total color change (ΔE_{00}) and the changes in the three-color coordinates (ΔL^*, Δa^*, and Δb^*) are shown in Figures 4 and 5. With the UV/IR filtered setup (Figure 4), the fading response of the papers was low; all samples were more stable than BWS 4, with C1, C7, and C9 being the most sensitive. Without the UV filter (Figure 5), the samples were more damaged, with C7, C8, and C9 as the most responsive samples, reaching BWS 3–4. In particular, the OBA-rich samples displayed yellowing ($+\Delta b^*$), which is a typical color change resulting from the photodegradation of OBAs (Mustalish 2000).

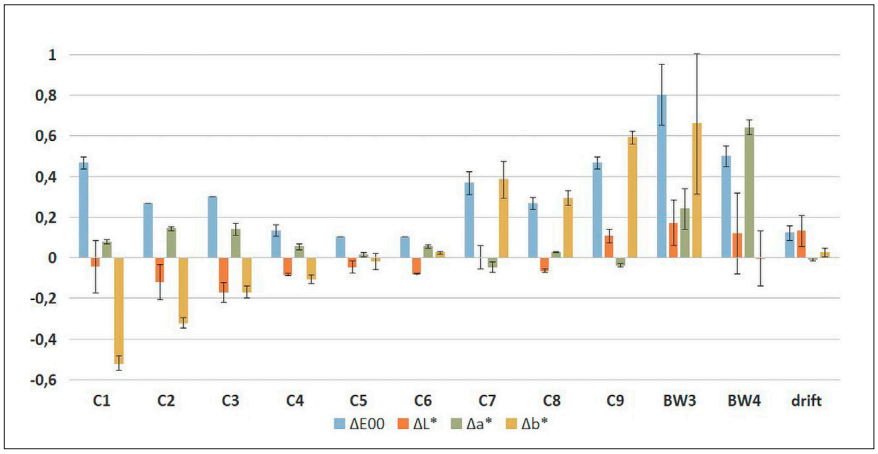

Figure 4. Xenon-MFT (with UV and IR filters) color changes of contemporary paper samples, BWS 3 and BWS 4 (15 min measuring time) and lamp drift

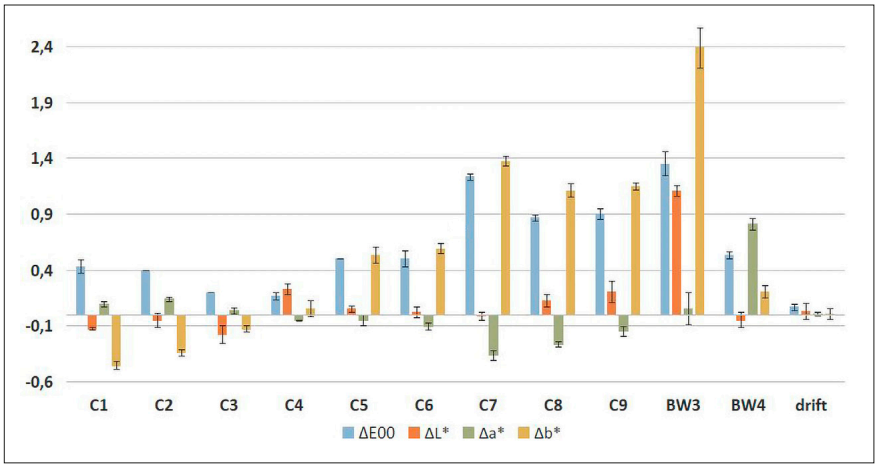

Figure 5. Xenon-MFT (UV filter removed, IR filter inserted) color changes of contemporary paper samples, BWS 3 and BWS 4 (15 min measuring time) and lamp drift

However, the same OBA papers (C2–C9) showed significantly greater responses in the museum foyer than to either of the xenon-MFT setups, even though the papers were displayed behind UV-filtering glass (Figure 6). Although the relative responsivity of the papers to the xenon-MFT without UV filter and to the natural light exposure in the museum foyer (BWS 3–4 vs. BWS 2–3) was similar, with C7–C9 as the most sensitive samples, MFT at best underestimated the overall sensitivity of the OBA papers by one BWS unit. These results demonstrate that the light sensitivity of optically brightened papers is not adequately reproduced by microfading using the xenon-MFT, even if some UV light is introduced by removing the UV filter.

A major issue in microfading tests of OBA papers, and one that most likely leads to these discrepancies, is the MFT-light source. Optically brightened papers are generally problematic in colorimetric measurements because their color impression is a mixture of reflected light and UV-induced fluorescence in the blue end of the visible spectrum. For adequate measurements, a light source with a relative UV portion calibrated to a standardized value and spectral range, such as the ISO 13655:2017 M1 standard, is required to avoid strong metameric effects (Andersson and Norberg 2007, Cheydleur and O'Connor 2011). The xenon-MFT light source cannot currently be "UV-calibrated" in a similar manner and at

Figure 7. Color change caused by photodegradation of OBAs in paper (C8 and C9, samples as listed in Table 3) when observed with a UV-filtered vs. a UV-rich illuminant (750 µW/L); the former represents the color change as "seen" by the xenon-MFT with the UV filter inserted

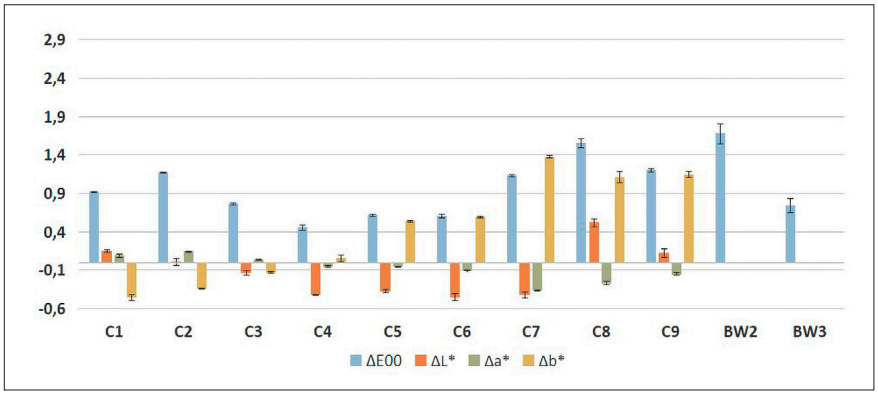

Figure 6. Color changes of contemporary paper samples after ca. 1 Mlx·h exposure behind UV-filtering glass in the museum foyer, BWS 2 and BWS 3 (X-Rite, ISO 13655:2009 M1 measurement)

most (when the UV filter is removed) includes only a small relative amount of UV radiation (100 µW/lum spot average), rendering OBA fluorescent properties mostly inactive during the measurement. Therefore, not only is the MFT poorly suited for correct colorimetric measurements of OBA papers, it also will not detect the portion of the overall color change that is due to the loss of OBA fluorescence, which makes up a significant part of the photo-yellowing of OBA papers (Figure 7).

CONCLUSION

Our findings suggest a broad light responsivity range for white papers, ranging from the mid "sensitive" to the "intermediate" category of current light policies. The preliminary results obtained in the museum foyer exposure showed that both naturally aged rag paper and optically brightened contemporary paper can be as prone to light-induced color changes as ligneous papers. For several reasons, MFT results are accompanied by a certain degree of noise, which translates into a colorimetric shift that is not related to the material under investigation. This noise can be significantly lowered by optimizing the operational practice of the MFT instrumental setup, but its effect must be considered when interpreting MFT results.

MFT measuring spots are visible only when testing exceeds a relatively high color difference of ΔE_{00} >5; testing in accordance with current best practices will not cause visible change in originals. The introduction of a certain amount of UV in MFT seems promising, especially for papers that contain UV-sensitive materials. However, the use of MFT to predict color changes in contemporary papers with OBAs remains problematic, and the light response of such papers may not be adequately represented by MFT results in general.

Forthcoming studies will evaluate other modes of museum exposure, accelerated light aging, xenon-MFT, and LED-MFT. The long-term goal is to formulate specific guidelines and precautions concerning the use of MFT on white paper, thus improving existing recommendations regarding MFT use in paper-based collections. We also hope to provide data that can be applied to more precisely distinguish the light sensitivities of white papers of different composition, thus differentiating their conservation requirements in terms of light exposure and exhibition planning and thereby allowing the amendment of current exhibition policies.

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MATERIALS LIST

Chromophores
2,5-dihydroxy-1,4-benzoquinone
2,5-dihydroxyacetophenone
5,8-dihydroxy-1,4-naphthoquininone
Sigma-Aldrich Chemie GmbH
Eschenstr. 5
82024 Taufkirchen, Germany
Tel.: +49 89 65130
gecustomerrelations@merckgroup.com
https://www.sigmaaldrich.com/

Filter paper
MN 260, 90 g/m²
Macherey-Nagel
52355 Düren, Germany
Tel.: +49 2421 969 0
sales@mn-net.com
https://www.mn-net.com/

Hardwood sulfate pulp
bleached/unbleached
Lenzing Papier GmbH

Werkstraße 2
A-4860 Lenzing, Austria
Phone: +43 7672 701 - 0
Telefax: +43 7672 701-2231
office@lenzingpapier.com

Historic papers
- Whatman rag paper, ca. 1801, from book quire, ca. 137–157 g/m²
Collection of the paper conservation department
Kupferstichkabinett Berlin
- Low-quality machine paper, ca. 1980, 80 g/m²
Study collection of the SABK Stuttgart, Paper conservation course
- NYT newsprint, ca. 1990s, blank page, ca. 45 g/m²
Personal collection of Irene Brückle

Rag pulp
From unwashed historic linen (ca. 1926) from the collection of the SABK Stuttgart, Paper conservation course
Custom processed by Gangolf Ulbrichts Werkstatt für Papier
Mariannenplatz 2
Kunstquartier Bethanien
10997 Berlin, Germany
https://papiergangolfulbricht.de

Rosin size solution
Fennosize RS KN 12 35A
Colophony, treated with fumaric acid, aluminum sulfate, mixed 5-Chlor-2-methyl-2H-isothiazol-3-on, and 2-Methyl-2H-isothiazol-3-on
Kemira Oyj
Energiakatu 4
P.O. Box 330
00101 Helsinki, Finland
Tel. +358 10 8611
Fax. +358 10 862 1119
https://www.kemira.com

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