Microplastic Pollution in the Library!
A Collaborative Investigation into the Curious Case of a Crumbling Waterproof Field Guide

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Abstract
Synthetic paper is waterproof and durable, but the discovery of the crumbling synthetic paper pages of a 37-year-old waterproof library monograph raised concern both about its longevity and its potential to contribute to environmental pollution. The page substrate was identified as polypropylene, a polymer known to deteriorate over time and still widely used today. I alerted and surveyed libraries holding waterproof copies of the field guide and determined that over 50% of library copies have deteriorated, but at different rates. Statistically, the number of circulations is not a significant factor in predicting the extent of deterioration, suggesting that ambient environmental conditions and chemical transformations of the polymer are the leading initiators of the deterioration observed. While the specific phenomenon observed with this one book may be a relatively rare occurrence, the collection management implications of plastic materials found within library collections are also discussed.

Keywords: Synthetic paper; waterproof paper; polypropylene; plastic deterioration; microplastic pollution; library materials management

Introduction
It was a day like any other day during routine collection management at the Marine Resources Library, but I observed a curious phenomenon in the stacks. While moving a book upon the bottom shelf, a fine dust became airborne and fell to the carpet along with some larger flakes of material; moments later, I detected a potent plastic odor. After a closer inspection of the item I had moved, it became clear that the fragments had come from an adjacent monograph. Upon retrieval, the origin of the fragments and plastic odor became clear.

I held a copy of the waterproof edition of John E. Randall's *Underwater Guide to Hawaiian Reef Fishes* co-published in 1981 by Harrowood Books, Newtown Square, PA and Treasures of Nature, Kaneohe, HI. Boasting 204 full-color plates over 72 pages, the guide is filled with bold, bright images of reef fishes and is intended for use in the field. In fact, the introduction to the book states, "This book is printed on plastic so it can be handled with wet hands, exposed to rain or salt spray, or even taken underwater by a diver. If wet with sea water, it should first be washed in fresh water before drying. When not in use, store out of direct sunlight." A further note advises, “Rinse and towel-dry after use; store away from sunlight.” The back cover states, “This book is printed on plastic with special ink…”

While the book cover appeared intact, the pages within had deteriorated resulting in fragmented material. Around the edges of the pages, extending inward up to an inch, the ink of the plates was crumbling and flaking from the page. Where the ink had disappeared, a thin white opaque material – the plastic page substrate – was revealed. Turning the pages caused more ink to crumble, demonstrating their precarious condition. Furthermore, in the middle third of the book the edges of the plastic pages within two inches of the binding were, themselves, crumbling. An investigation of circulation activity demonstrated that this book had circulated just once in 1987.

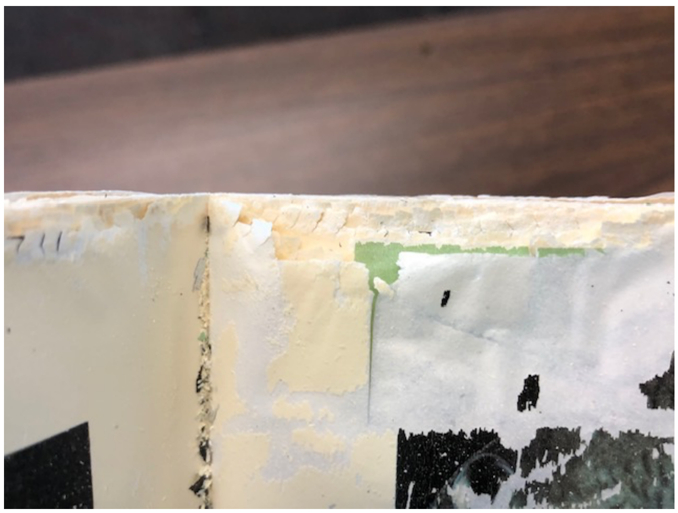

Figure 1. Deteriorating ink and pages in the waterproof field guide
Photo credit: Geoffrey P. Timms

Synthetic Paper

The need for waterproof writing and printing materials has long been recognized due to the poor durability of paper in wet conditions (“Waterproof Writing Ink and Paper,” 1886). Chemical innovations of the twentieth century heralded a new era for the written word: polymer-based synthetic paper. Manufacturing methods of synthetic paper included the creation of synthetic fibers for use with existing paper-production equipment, as well as the development of extruded film from thermoplastic polymers. Printing on extruded film synthetic paper required further innovation because inks used on cellulose-based paper do not adhere well to the surface of the film (Lunk & Stange, 1971).

Several strategies have been employed to improve printing upon synthetic paper, involving modifications to both the ink and the paper surface. Specially formulated inks incorporating organic solvents and synthetic resins were developed, while modification of synthetic paper with additives, including calcium carbonate surface coating and clay-based fillers, improved ink adhesion (Hutchinson, 1976; Katz, 2017). An additional strategy to improve print quality was the modification of extruded film surfaces using corona plasma treatment. The resulting oxidation of the surface of the synthetic material creates a roughened texture to which ink more readily adheres (Lunk & Stange, 1971; Sellin & Campos, 2003; Strobel et al., 1989). Such innovations made widespread use of printed synthetic paper feasible, particularly for labeling and packaging.

Synthetic paper has slowly permeated the publishing and banking industries including the production of waterproof maps, species identification cards, books, and currencies. In April 1986, the first hardcover book was printed on extruded polypropylene synthetic paper (Frank, 1986; Monaghan, 1986). Two years later, Australia launched the world's first synthetic banknote. The plastic substrate was formed as a linear low-density polyethylene and polypropylene co-extruded laminate (Prime & Solomon, 2010). By 2016, over 30 countries had polymer currency in circulation. Polyethylene and polypropylene remain in widespread use today.

Polypropylene, a petrochemical-based product known as a polyolefin, is a very widely used plastic polymer with a competitive manufacturing cost (George & Celina, 2000). The first polypropylene-based synthetic paper, Ucar, was produced by Union Carbide Corporation and Mead Paper Company in the 1960s (Kuypers, 2016). By the 1970s, Dupont was manufacturing a polypropylene-based synthetic paper material called Typar, a building wrap product (Lunk & Stange, 1971). Variants of both products continue to be manufactured.

As a durable, waterproof, and feasible alternative to cellulose-based paper, polypropylene-based synthetic paper may exhibit an Achilles heel: polypropylene has been observed to be temporally unstable. This instability, resulting in increased brittleness, is associated with its oxidative tendency (George & Celina, 2000) and increased density with age (Fiebig et al., 1999). Even so, in one accelerated ageing study Yupo Corporation's Yupo extruded polypropylene synthetic paper actually demonstrated superior durability compared to both cellulose and synthetic fiber paper when artificially aged (Karlovits & Gregor-Svetec, 2012). The incorporation of some additives to the polymers used in synthetic paper – to enhance color, brightness, and ink adhesion, for example – may contribute to faster and less predictable rates of chemical deterioration than with a pure polymer alone (Kuypers, 2016). Recent studies have observed, modeled, and predicted degradation rates of polypropylene art in museum lighting conditions (Manfredi et al., 2017) and polypropylene products exposed to natural environment conditions, with particular interest in temperature, rainfall, humidity, and ultraviolet light exposure (Azuma et al., 2009; Kuypers, 2016; Lv et al., 2017; Ni et al., 2015; Song et al., 2014). Despite its short-term qualities, the long-term viability of polypropylene has presented challenges.

Research into the use of stabilizing additives to mitigate thermal, photo, and oxidative degradation of polypropylene has been ongoing for decades. Polypropylene stability may be improved with additives that screen ultraviolet light or inhibit thermal and photo exposure degradation processes (Matta et al., 2017; Zannucci & Lappin, 1974). A negative consequence of reduced degradation rates is the increased persistence of plastics in landfills, which has prompted research into viable organic plant-based stabilizers such as lignin (Gregorová et al., 2005; Ye et al., 2016).

Initial motivators in the development of synthetic paper were durability and waterproofing, but more recently environmental stewardship has emerged to further encourage a transition from tree-based paper to a synthetic product. Some of the largest producers of synthetic paper promote their commitment to environmental protection, declaring that production of synthetic paper does not contribute to deforestation and uses as much as five times less water than traditional paper. Additionally, disposal by recycling or incineration is claimed to avoid negative impacts upon the environment (Polyart n.d.; Yupo n.d.). The synthetic paper market continues to grow, with production estimated to increase by over six percent from 2016-2023 (Katz, 2017), while the development of techniques to both waterproof cellulose paper and improve synthetic paper is ongoing. However, environmental activism may ultimately suppress the synthetic paper industry that it helped to build, as demands for reduced consumption of plastic products become increasingly prevalent.

Research Questions

The physical deterioration of Randall's *Underwater Guide to Hawaiian Reef Fishes* raised several questions from which I have established hypotheses:

1) What is the deteriorating material on which Randall's book is printed?
   - Hypothesis 1: The material is a polymer still widely used today.

2) Is the deterioration unique to this copy of the book, or is it more widely observed with other libraries' copies?
   - Hypothesis 2: Copies held by other libraries do exhibit deterioration.

3) If other libraries' copies exhibit deterioration, does deterioration correlate with circulation frequency?
   - Hypothesis 3: The level of deterioration increases with circulation frequency.

Methods

Chemical analysis

Four samples were taken from the book, deposited in clean glass vials, and sent to the College's Department of Chemistry for analysis. The samples consisted of fragments of ink, powdered material found between pages of the book, cut samples from a page with a white background on each side, and cut samples of exposed page substrate. The larger ink fragments exhibited a white side and a black side, having originated as the black backgrounds upon which images of fish were presented. A Fourier-Transform Infrared Spectroscopy analysis was completed upon the cut samples and each side of several ink fragments.

Attempts were then made to dissolve the powder in six organic solvents in preparation for Gas Chromatography–Mass Spectrometry, Matrix Assisted Laser Desorption/Ionization, and Nuclear Magnetic Resonance Spectroscopy analyses. Solvents used included ethanol, acetone tetrahydrofuran, ethyl acetate, methylene chloride, and xylenes – representing a wide range of polarities and solvating powers. None of the solvents dissolved either the white or black material. Further analysis was not attempted and a report of probable composition for the dark ink and page fragments was returned to the library.

Efforts to contact the publisher to learn about the materials used in production of the monograph were unsuccessful.

Survey of Holding Libraries

A brief survey was developed on Google Forms and distributed by email. Brevity was a priority in the survey design to maximize the number of responses. Two multiple-choice questions sought information about the worst levels of deterioration observed for both ink and pages, with descriptions and photographs of our own copy provided as a guide for consistent rating (see Appendix 1). Circulation counts were solicited for each item and respondents were encouraged to communicate further information in a free-text field.

To identify potential survey recipients, I searched WorldCat to determine library holdings of the waterproof edition of the monograph. For the 76 libraries listed, library websites were accessed both to search library catalogs for the item and to identify contacts to whom the survey could be sent. One library website was unavailable, while searches of 14 library catalogs returned no holdings of the item. Contact information for collection managers, or the closest relevant position, was gathered. Several small library websites provided only a generic library email address, while others provided online forms as a way to contact individuals or departments. One recipient for each library was chosen where possible, and generic library or department email addresses or forms were used where necessary.

In early January 2019, I distributed the survey to 61 libraries by email or contact form with an introduction designed to alert recipients to a potential condition problem for an item held by their library. Photographs demonstrating the condition of our own copy were included in the email message to increase recipient engagement. The survey link was provided, along with a link to the library's own catalog record for the item. Recipients were asked to complete the survey once for each copy of the monograph held by their library.

Findings

Chemical Analysis

The Fourier-Transform Infrared Spectroscopy analysis was reported to reveal a spectrum from the white side of ink fragments approximating that of polypropylene, a material popularly used in synthetic paper. The dark side of fragments resulted in a spectrum representing carbon black, a common pigment in dark inks. The spectra for the white page cuttings and the exposed substrate cuttings also indicated a close match for polypropylene. The lack of solubility of the white and black powder in the six solvents was reported as consistent with the materials being predominantly polypropylene and carbon black respectively. The analysis identified no conclusive evidence of a surface coating on the polypropylene substrate, suggesting that the declaration of the use of "special ink" on the book's back cover represents the printer's solution to the challenge of ink adhesion upon polypropylene.

Survey of Holding Libraries

Fifty-three form submissions were received and representatives of three additional libraries sent emails stating that their copies of the monograph, while listed in their catalogs, could not be located. One form submission acknowledged that the 1981 waterproof edition was missing but that a 1985 paperback edition appeared to have been substituted without correcting the catalog record. This response was excluded from the results. The Marine Resources Library's own copy was evaluated and added to the results.

Of the 53 valid responses, 37 (69.8%) reported no deterioration in ink. Based upon the form narrative section, however, four responses were adjusted to reflect "discoloration" and the five-point rating scale was adjusted to account for this sixth category. Similarly, 31 (58.5%) reported no deterioration in pages. Of these, four were adjusted to reflect "very slight damage including rippling" based upon the descriptions provided and a sixth category was added to the rating scale. Some level of deterioration in ink or pages was observed in 28 copies (52.8%).

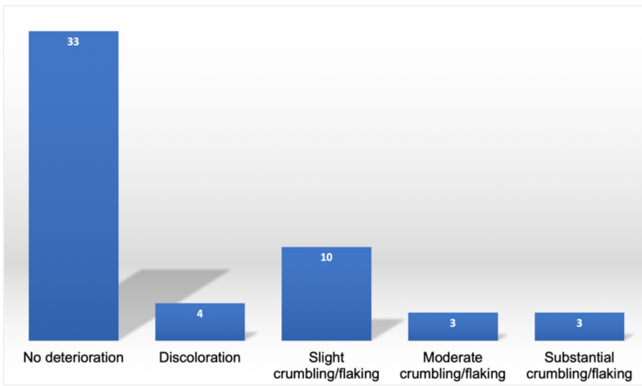

Figure 2. Observed ink deterioration by category (n=53)

The "slight crumbling" category was the most reported level of deterioration in both ink and pages, at 10 copies (18.9%) respectively (Figures 2 and 3). "Slight crumbling" represents crumbling or flaking of ink in areas of up to one square inch or crumbling of page edges up to one quarter of an inch into the page. No catastrophic crumbling of ink was reported, but one incidence of catastrophic page damage was described.

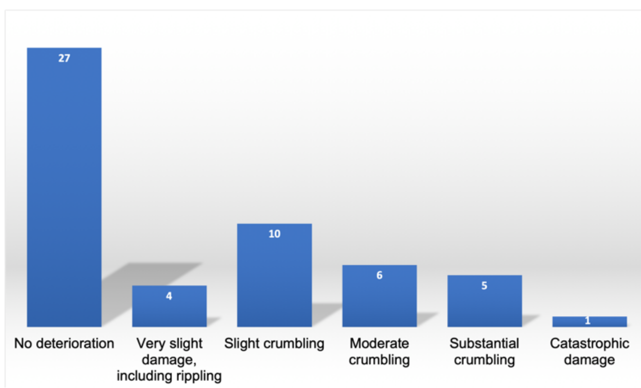

Figure 3. Observed page deterioration by category (n=53).

Circulation rates were reported on 49 survey responses. Of these, six circulation counts were acknowledged to be partial; i.e., complete circulation data had not been preserved during one or more library system migrations. These six responses were excluded from further analysis. Using the 43 remaining circulation rates, a mode of zero and a median of one demonstrate a frequently very low circulation rate for this title (Table 1). Copies with either zero or one circulation account for 23 (53.5%) of the responses. The mean circulation rate of 5.74 is strongly influenced by four particularly high values ranging from 18 to 57. With these outliers removed, a mean circulation rate of 2.67 is observed.

Statistical Test for Correlation

To determine if correlation exists between page or ink condition and circulation count, the data were first evaluated for central tendency. The data were not normally distributed, demonstrating positive skewedness (Figures 3 & 4). Spearman's correlation tests were used to evaluate the correlation between ink condition and circulation count ranks, and page condition and circulation count ranks. The ordinal condition ratings and the circulation data were ranked for the 43 circulation rates not reported as incomplete and Spearman’s correlations were executed.

Table 1. Frequency of observed circulation rates

Circulation Rate	Frequency
0	13
1	10
3	3
4	2
5	2
6	3
7	2
8	2
9	1
10	1
18	1
20	1
48	1
57	1
Total	**43**

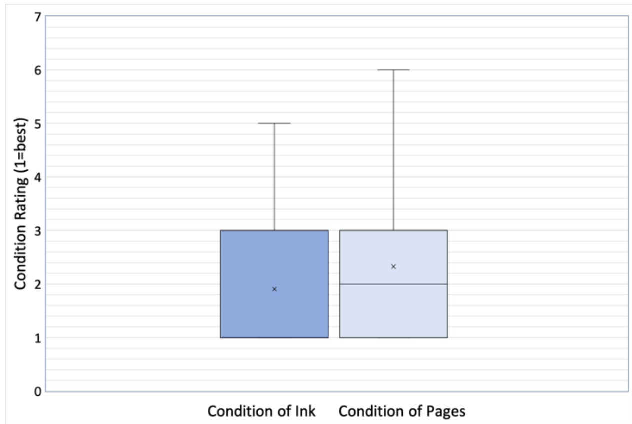

Figure 4. Distributions of ink and page condition ratings.

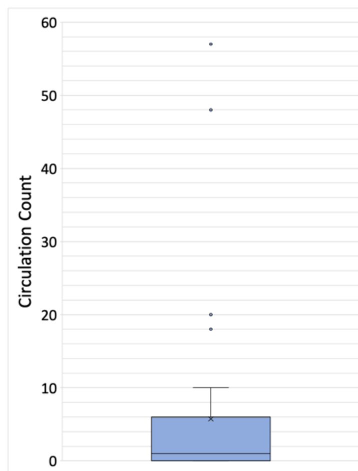

Figure 5. Distribution of circulation count.

No statistically significant correlation was observed between ink condition and circulation count (r_s = 0.19, n = 43, p > .05), and no statistically significant correlation was observed between page condition and circulation count (r_s = -0.07, n = 43, p > .05). With the four highest-value outliers removed from the dataset, Spearman's correlation tests also showed no statistically significant correlation between ink condition and circulation rate (r_s = 0.07, n = 39, p > .05) or between page condition and circulation rate (r_s = -0.18, n = 39, p > .05). Thus, in each case we fail to reject the statistical null hypothesis of no association between page or ink condition and circulation rate.

Limitations

While six survey responses acknowledged that their circulation data were not complete, I have assumed that all other circulation data reported represent all circulations of the monograph for the duration of its presence within the reporting library. Any data that do not represent actual circulations reduce the validity of the correlation analysis.

The circulation data simply tell us how often an item was circulated. We have no way of knowing the conditions experienced by copies of the monograph during circulation. Exposure to UV light, high temperatures (such as those observed in parked cars), humidity, and various natural and synthetic products, like seawater and sunscreen, may impact the condition of a polypropylene-based synthetic paper product. There are, therefore, numerous factors that we cannot measure, and which may have contributed to the deterioration observed in copies of this book.

Similarly, it is not possible to account for the ambient temperature, humidity, and UV light exposure experienced in the library stacks over the life of each book. Some copies with moderate to substantial deterioration of pages or ink were reported not to have circulated at all. Any environmental variables affecting the condition of these books must primarily be a function of the library's internal environment.

Discussion

I have confirmed Hypothesis 1, that the substrate of the deteriorating monograph is a widely used polymer, polypropylene, still in use today. Furthermore, I have observed that the ink comprising the plate backgrounds contains carbon black pigment. I have also confirmed Hypothesis 2, that deterioration of this monograph is not unique to our copy, with evidence that some deterioration in the ink or pages has occurred in 52.8% of the 53 copies evaluated. I have rejected Hypothesis 3, that there is a correlation between use and deterioration by a statistical analysis of data for 43 copies. Thus, we are left to consider which factors may have contributed to the deterioration.

It is unfortunate that data are not available about the duration of each circulation of the book. It could be informative to evaluate the relationship between the cumulative circulation time and the level of deterioration, to determine if books circulated for longer periods are more likely to show the deterioration observed in this study. It is possible that conditions experienced during circulation tend to initiate chemical changes in the page substrate, potentially resulting in a significant positive correlation between the number of days the item has circulated and the observed level of damage. The note printed in the book about protecting it from prolonged sunlight exposure suggests that the effect of UV light was an anticipated problem.

Similarly, if exposure to environmental conditions including UV light, high temperatures, and salt water initiate chemical changes that result in deterioration, then the date of initial exposure may influence the impact-per-circulation of future use of the book. Circulations occurring substantially later when fragility is increased are more likely to cause damage than circulations occurring closer to initial circulation dates. Thus, the chronology of circulations may contribute to explaining the level of deterioration manifest in the present day. Such data are not typically preserved by libraries and cannot be investigated.

It is noteworthy that the four highest circulated items in the current study were all reported to be in good condition. If the exposure of these copies to various environmental conditions during circulation has not visibly impacted their physical condition, it is, perhaps, an indicator that the environment in which the book is located for the long term – the stacks – may be a greater determinant of temporal degradation than environmental conditions during circulation.

The presence of polypropylene on the reverse (white) side of black ink fragments suggests that delamination of the page substrate may have occurred. That is, the surface layer of the extruded polypropylene has separated from the remaining substrate. This could also be indicative of a long-term consequence of corona treatment of the surface of the page, particularly due to the documented relationship between oxidation and deterioration in polypropylene (George & Celina, 2000).

Having identified this single occurrence of deteriorating plastic material in the library collection, the question arises of how much other plastic exists in the collection. Plastic may be found within library collections in numerous forms. In addition to synthetic paper, library materials may be laminated with plastic, spiral or comb bound with transparent plastic covers, or bound using plastic combs. Individual journal issues can be shelved in plastic file cases. Libraries with three-dimensional collections and learning objects may house a variety of plastic materials. Various audiovisual media also incorporate plastic casings, laminates, and films. Plastic, then, is likely to be present in the collections of most libraries.

Walking the stacks is one way to identify and evaluate plastic materials, particularly comb bindings which are easily identified and tested for condition. Comb bindings can become brittle with age and lose their "teeth." This is easier to address than a book which is crumbling, because a plastic comb can be replaced relatively easily. Identification of plastic materials that are incorporated into covers, cover laminates, pages, or inserts of books are harder to identify without handling each item. Monographs would need to be removed from the shelf and inspected externally and internally, according to several criteria. Such handling would demand substantial time to work through even a small part of a collection and would likely be considered an unfeasible investment of staff time. At the Marine Resources Library, brittle and damaged plastic comb bindings and plastic report covers have been observed in the collection when handling materials and plastic fragments have been found on the floor among the shelf ranges. A 2021 collection shift increased awareness of the extent of plastic comb binding degradation.

If bibliographic records were to consistently identify the use of plastics in synthetic paper, bindings, or covers, then collection managers could selectively evaluate items based upon the results of a catalog search. The inclusion of notes about plastic materials in bibliographic records that are increasingly generated by third parties would demand additional effort, also making this an unrealistic proposal for libraries with limited staff resources. If notation of plastic content were to become a cataloging standard, however, then occasional evaluation of the collection would be better facilitated. Indicators of synthetic material would be particularly helpful for items of historical and cultural importance that become archived for long-term preservation.

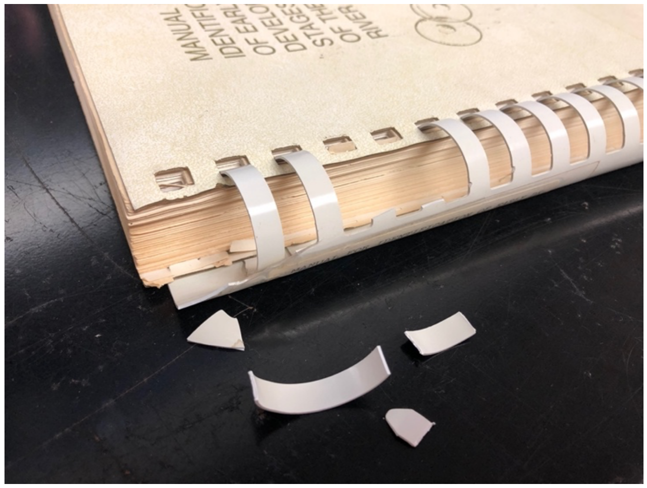

Figure 6. Brittle and fragmenting comb binding.

Library collections themselves are not the only sources of plastic degradation and potential pollution in the library. In the past year at the Marine Resources Library, chair wheels in the study area of the library have been observed to be deteriorating, with a surface layer of soft plastic crumbling into fragments and further fragmenting as the chair wheels run over them. The same chairs in the main campus library exhibit the same problem. Furthermore, plastic sheets stored in plastic boxes for use during impending hurricanes are deteriorating. The sheets are losing flexibility and large fragments detach when the sheets are unfolded for use.

Figure 7. Plastic fragments from library chair wheels.

Lastly, the disposal of degraded plastic, of any size or form, is problematic. If it is vacuumed from floors or deposited in trash cans, it will likely find its way to a landfill where it can potentially be ingested by animals or reach water courses and eventually the ocean. Degraded plastic, however, cannot be recycled with traditional recycling methods. Chemical recycling, which can process degraded plastic, offers hope but is still in its commercial infancy (Tullo, 2019). Perhaps the best current way to keep deteriorated plastic that originated in a library collection away from marine and aquatic ecosystems is to store it safely until recycling technologies advance such that it can be processed in an environmentally responsible way.

Conclusions

We have observed that the deterioration of a monograph printed on polypropylene synthetic paper has occurred unpredictably, and at different rates and severities. If the use of polymer-based paper increases in the publishing industry, there are implications for the longevity of library materials. While the value of synthetic paper for applications with anticipated short-term lifespans is well established, more research needs to be undertaken to understand the consequences of the use of polymers in both publications and bindings anticipated to be used in the long term. The presence of plastic in today's library collections warrants awareness of plastic degradation as well as mitigation efforts to ensure that the library does not become a pathway to environmental plastic pollution.

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