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Open Book, Open Source: PCB Usage in Mass-Market Paperback Book Adhesives

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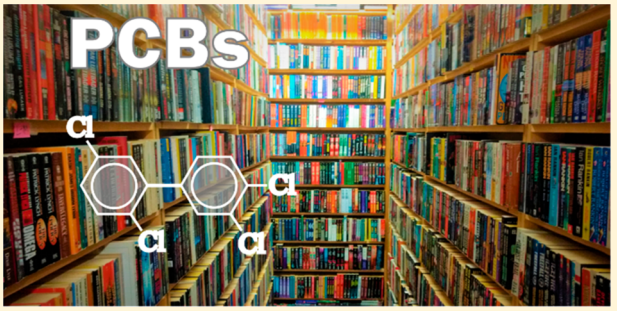
Supporting Information

ABSTRACT: This study documents an unrecognized source of polychlorinated biphenyls (PCBs) to indoor and outdoor environments from mass-market paperback book adhesives. The PCB content of common consumer products like books is less documented than industrial products. Patents describe PCBs as components for hot-melt adhesives used for "perfect" bound paperback bookbinding. However, the PCB formulations, concentrations, and dates of use for these adhesives have not been confirmed by chemical analysis. We tested production-dated paperback books manufactured between 1946 and 1974 from six major publishers. Chemical analysis of the binding adhesive confirms the widespread presence of PCBs in paperback books. PCBs were detected in adhesives from all tested books manufactured between 1948 and 1974 (n = 21). High PCB concentrations (6.1−18 wt %/wt), consistent with use as a plasticizer, were found in half the samples (n = 12). We tentatively identified polychlorinated terphenyls (PCTs) in four samples with lower, ppm-level PCB concentrations. From 1948 to 1974, we estimate that over 6 billion mass-market paperback books were sold domestically, many of which remain in homes, libraries, and stores. Therefore, book adhesives may be a potential PCB source to indoor air and to the environment via paper mill effluent, landfills, and recycled paper.



INTRODUCTION

The scientific community has raised concerns about polychlorinated biphenyls (PCBs) in indoor air,[1−3] sediment, and aquatic ecosystems,[4,5] and other environmental reservoirs globally.[6−8] While PCB compositions within "closed" systems—such as transformers, capacitors, and heat transfer fluids—have been well documented,[9−13] recognition of "open" system end-uses (adhesives, sealants, and surface coatings) remain relatively obscure.[14,9,11] Mass-market paperback books are an underappreciated "open" system where PCBs used in hot-melt adhesives remain in contact with the environment, particularly indoor air.

PCBs were described in patents,[15,16] trade,[14,17] and technical literature[18,19] as components in hot-melt adhesives for bookbinding. Hot-melt adhesives were used for manufacturing adhesive-bound paperbacks, known as "perfect" bound books in the bookbinding trade.[20] However, PCB formulations, concentrations, and the dates of use for these adhesives have not been confirmed by chemical analysis. Erickson and Kaley's review of PCB applications summarizes the problem: "*the bulk of the references to the use of PCBs as adhesives are from patents; there is no evidence how many products were ever in commerce or what PCB volumes they represented.*"[11] To our knowledge, no one has published PCB analytical results of adhesives from mass-market paperback books.

Monsanto Chemical Company (Monsanto), the primary U.S. producer of PCB mixtures between 1929 and 1977,[21] voluntarily ceased sale of PCBs for open uses by 1972.[11] Monsanto marketed PCB mixtures under the Aroclor trademark as liquids (1221, 1232, 1242 (1016), 1248, and 1254) and solids (1260, 1262, 1268, and 1270).[9,11] The Aroclor product line also included polychlorinated terphenyls (PCTs) as solid PCB−PCT mixtures (e.g., 2265, 4465, 6050) and solid PCT mixtures (e.g., 5460, 5441, and 5432).[9,11,14] Monsanto was the primary U.S. producer of PCTs between 1929 and 1972,[22] with PCT imports continuing in the 1970s.[22]

Hot-Melt Adhesive Use in Paperback Books. Hot-melt adhesives were used in "perfect" bound books to bind the pages of the paper block with a flexible adhesive and join it to the cover. The solid hot-melt adhesive was melted, heated to the proper viscosity, and then applied to the paper block usually by rollers. Prior to the development of hot melts, synthetic emulsions and animal glues were used in adhesive-bound books. These other adhesives had long drying times, leading to production bottlenecks.[23,24] Seeking improved speed and lower costs, the book publishing industry saw promise in fast-setting hot melts, first patented in 1944−1946.[23,24,15,16] Despite some dissatisfaction with early hot-melt

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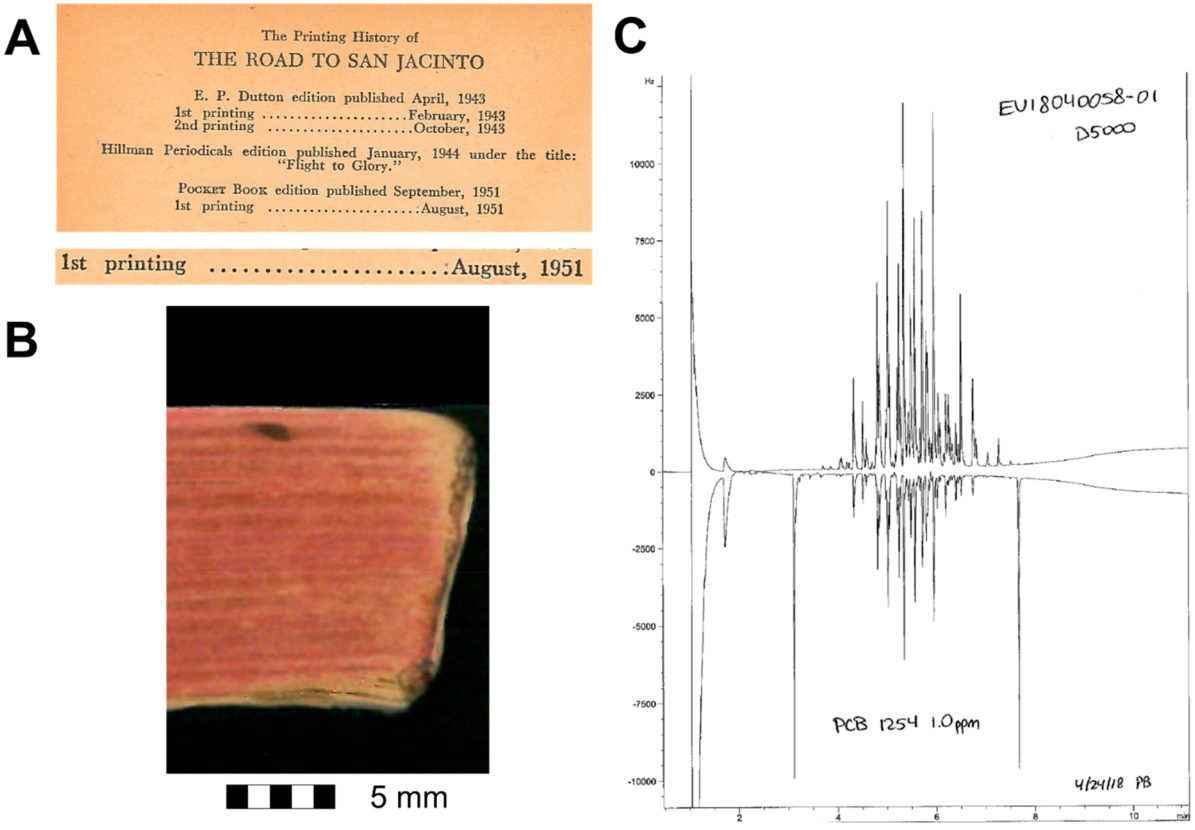

Figure 1. PCB Aroclor analysis of mass-market paperback books. A representative mass-market paperback used in this study. (A) Excerpt of copyright page (top) showing the publishing history and production date: shown enlarged (below). (B) Close-up end-view image of the adhesive binding. The corners of the book are worn and rounded as illustrated by the lack of red-dyed edge. (C) A laboratory-provided electron capture detector (GC-ECD) mirrored chromatogram comparing the adhesive sample result (top) with an Aroclor 1254 standard (bottom) by EPA Method 8082A.

adhesives,[24] "perfect" binding became a standard method for paperback publishers by 1947.[25]

PCBs in Adhesives. As early as 1930, Chester Penning, a Swann research chemist, suggested Aroclor use for adhesives.[26] By 1944, Monsanto had actively expanded the application of Aroclors as versatile plasticizers (softeners).[27,28] With the growth in PCB production capacity during WWII,[11] Monsanto marketed the resinous Aroclors (i.e., PCB−PCT mixtures Aroclor 2565 and 4465) as "*better than anything else discovered to date primarily because of their high compatibility with other resins and their thermo-stability.*"[27] PCBs and PCTs are also compatible with polyvinyl acetate (PVAc),[14,18,29,30] a type of resin used for bookbinding emulsions and hot-melt adhesives.[14,19,31] In general, PCBs are compatible with PVAc resins up to a ratio of 1:2 by weight.[30] Additionally, the Aroclors used as plasticizers and tackifiers in PVAc resins also helped to create sharp melting and congealing points.

"Typical" or "starting" hot-melt formulations for bookbinding were reported as containing PCBs at 43%, 48%, and 50% in patent literature,[16] 26% and 35% in technical literature,[18,19] 5% and 21% in Monsanto marketing literature,[14] and 20% and 29% in *The Chemical Formulary*,[32] listed here by weight. Bookbinding hot melts are also described in conjunction with PCTs use.[17,33−36] Monsanto internal documents report usage of Aroclor 1248, 1254, and 5460 in 1961[34] and mainly Aroclor 1248 and Aroclor 5460 in 1968.[33,35] While hot-melt adhesives were commonly used for "perfect"-bound softcover books, application to hardcover books—more typically made with sewn bindings—was rarer.

Pocket Books jump started the "paperback revolution" in 1939[37,38] and maintained the dominant market share in the 1940s and 1950s.[20,38,39] Binderies working with Pocket Books were early adopters of hot-melt adhesive binding methods.[20,37] By testing Pocket Books brand paperback books and additional books from five other major publishers, we confirm the continued presence of PCBs and PCTs in the adhesives of these mass-produced paperback books.

MATERIALS AND METHODS

Sample Acquisition. We purchased 22 mass-market paperback books manufactured between 1946 and 1974 from online and local book sellers. This study focused on Pocket Books brand paperbacks (n = 15 and 2 replicates). We also tested books from five other major publishers (n = 7): Bantam (1952, 1962, and 1964), Ballantine (1964), Popular Library (1964), Gold Medal (1964), and Pyramid (1965). A Pocket Books edition published in 1963 was singled out for additional testing. We tested three books from the same edition and split and tested one adhesive sample in triplicate. Figure 1 shows a representative book, copyright page excerpt, end-view of the adhesive, and laboratory-provided chromatogram. Additional details of sample dating and preparation are provided in the Supporting Information Methods section, Figures S1−S22, Table S1, sample information, and Table S3 paperback bibliography.

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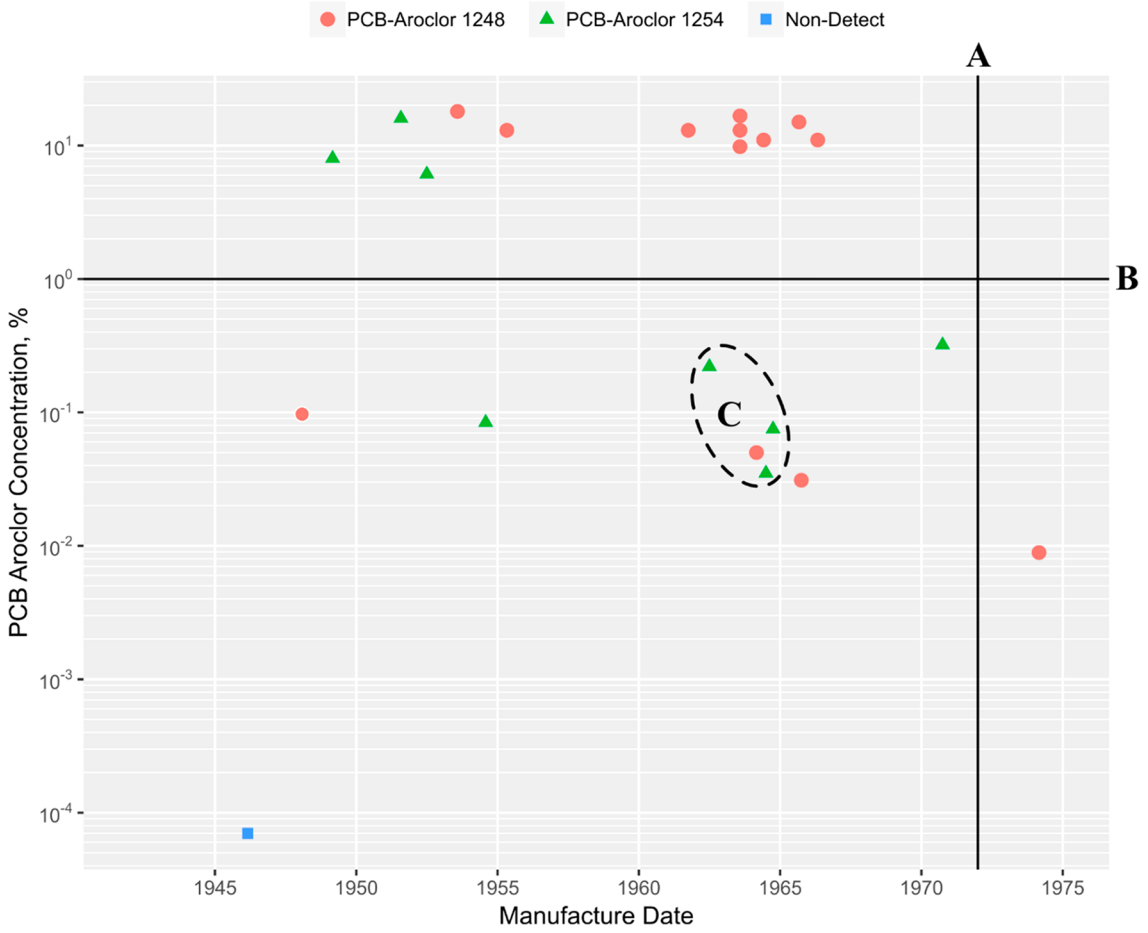

Figure 2. PCB Aroclor content in mass-market paperback book adhesive samples by date of book manufacture. (A) Vertical black line (1972) marks the end of U.S. domestic Aroclor sales for open system uses such as adhesives. (B) Horizontal black line at 1% (10,000 ppm) demarcates two groups where PCB concentrations suggest intentional use as a plasticizer (above the line) and nonintentional use (below the line). (C) Samples within the dashed ellipse had chromatograms showing late-eluting non-PCB peaks consistent with an Aroclor 5460 standard, a PCT mixture. PCT mixtures have been historically used for plasticizer applications.

Table 1. Summary of ΣPCB Concentrations (ppm) Quantified as PCB Aroclors[a] in Mass-Market Paperback Book Adhesives (1948−1974) for Two Groups Where PCB Concentrations Suggest Intentional Use as a Plasticizer and Nonintentional Use

Group	Year[b]	Sample size	Min	Mean	95% CI[c]	Max	SD	95% PI[d]
Intentional	1949 to 1966	12	61,000	125,472	[100,000−150,000]	180,000	35654	[50,000− 200,000]
Nonintentional	1948 to 1974	9	89	1023	[240−1800]	3200	1021	–

[a]PCBs were quantified as either Aroclor 1248 or Aroclor 1254 based on laboratory Aroclor identification (see Table S2 for identified Aroclor). [b]Year of mass-market paperback book manufacture. [c]CI refers to confidence interval about the mean calculated assuming a *t*-distribution. [d]PI refers to prediction interval, which is not calculated for the nonintentional use group.

PCB Aroclor Analyses. We mechanically separated the adhesive binding of each book from the pages before an analytical laboratory performed a chemical extraction on the binding. ALS Global in Everett, WA, USA, performed the extraction and analytical chemistry by Aroclor Method EPA 8082A.[40] The ultrasonic extraction was performed by EPA Method 3550C[41] with 1:1 dichloromethane/acetone exchanged with hexane. The sample extracts were analyzed using a Hewlett-Packard (HP) 7890 gas chromatogram (GC) with dual columns and dual injectors paired with HP G2397A electron capture detectors. Two Restek columns, CLP-1 30 m × 0.53 mm × 0.5 μm and CLP-2 30 m × 0.53 mm × 0.42 μm, were used (120 °C, ramping up at 30 °C per minute to 200 °C, then 20 °C per minute until 330 °C, for 2 min). Table S2 includes analytical results. An assessment of data quality is included in the Supporting Information.

PCT Standards. The laboratory analyzed two PCT standards Aroclor 5460 and 5442 by EPA Method 8082A using 35 μg/mL standards in toluene (AccuStandard T-460S and T-442S).

RESULTS AND DISCUSSION

PCB Aroclor Identification and Concentration. Either Aroclor 1248 or Aroclor 1254 was identified in all tested adhesives from books manufactured between 1948 and 1974 (n = 21) (Figure 2 and Table S2). The identification of these two Aroclors is consistent with historic documentation for hot-melt adhesives in books.[16,19,33,34] PCBs were not detected (at 0.70 ppm) in the adhesive from a book published in 1946.

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These results are consistent with the accepted chronology of hot-melt adhesive development for bookbinding in the mid to late 1940s.

We divided the samples into two groups based on PCB concentrations: (i) intentional use (greater than 1% PCBs) and (ii) nonintentional use (less than 1%). Percent-level PCB concentrations (6.1% to 18%) were found in half the books (n = 12) published between 1949 and 1966, consistent with intentional use of PCBs as an adhesive plasticizer.[18,19] Nonintentional use levels (89 to 3200 ppm) were found in samples from books (n = 9) published between 1948 and 1974. In all cases, our results provide a lower bound estimate of PCB concentrations in the adhesive at the time of manufacture, since volatilization and plasticizer migration into the paper are possible.

Intentional PCB Use. The estimated mean concentration for the tested books where PCBs were intentionally used is 12.5% (95% confidence interval [10%, 15%]) (Table 1). These samples were manufactured between 1949 and 1966 (Figure 2). However, historical documents and our results suggest a wider time frame for intentional PCB use in adhesive book bindings as early as the late 1940s through 1968 and possibly until the cessation of Aroclor sales for open use in 1972.[11] For instance, the detection of PCBs in a book published in 1948 (970 ppm as Aroclor 1248) suggests that books were being concomitantly bound with PCB adhesives at higher concentrations.

Our results indicate PCB adhesive concentrations likely vary considerably among the extant population of mass-market paperback books (95% prediction interval [5.0%, 20%] for books manufactured intentionally with PCB-plasticized adhesive. The variability is consistent with industry use where hot-melt adhesives formulations may vary through time and by bindery, bookbinding equipment, book width, film thickness, adhesive formulator, and PCB mass-loss by migration to the paper or volatilization to the air. Three books from the same printing edition had PCB concentrations of 9.8%, 13%, and 17% (coefficient of variation 0.26), with one book tested in triplicate 13%, 16%, and 21% (coefficient of variation 0.24), suggesting that differences in extraction efficiency and sample heterogeneity across sample splits also contribute uncertainty to the PCB mass estimates. Although most of the books in this study were published by Pocket Books, the results may be broadly applicable to "perfect"-bound paperbacks and magazines because only a few binderies with specialized high-speed equipment serviced the major mass-market publishing houses.[20,23,38]

Nonintentional PCB Use. Adhesive samples with PCBs detected in part per million concentrations (89 to 3200 ppm) for samples (n = 9) from books published between 1948 and 1974 are not consistent with purposeful PCB plasticizer use. Other plasticizers, such as phthalates and PCTs, may have been used as alternatives to PCBs in some hot-melt adhesive formulations. We compared sample chromatograms to two PCT standards, Aroclor 5442 and 5460, and found a good match to Aroclor 5460 for four samples manufactured between 1962 and 1964 (which contained 350, 500, 750, and 2200 ppm PCB) (Figures S23 and S24). The chromatograms suggest appreciable percent-level PCT concentrations in each of the four samples, but the PCT concentrations are not quantified here.

Adhesive samples with low-detected PCB concentrations (89 to 3200 ppm) could have resulted from cross-contamination during adhesive formulation or from glue pot residue during book manufacture. PCBs can also occur as impurities (0.5% to 1%) in PCT Aroclor mixtures, produced during chlorination of a terphenyl feedstock containing biphenyl.[42,43] Volatilization sufficient to reduce the PCB content in book adhesives from more than 5% to less than 1% (the amount needed here to misclassify intentional versus nonintentional PCB use in this study) cannot be completely ruled out. PCB volatilization is temperature dependent, increasing with temperature as a function of vapor pressure.[44] The storage histories of the individual book samples are unknown.

Scale of PCB Usage in Hot-Melt Adhesives. Mass-market paperback books were common and represent ongoing, open sources. We estimate that approximately 6 billion mass-market paperback books were sold between 1948 and 1972.[25,45−48] We discovered historical information that Aroclors were used in bookbinding hot-melt adhesives at more than 500,000 pounds per year in 1961[34] and at least 460,000 pounds per year in 1968.[33] It turns out, Aroclor used in hot-melt adhesives was one of seven major plasticizer uses in 1961[34] and ranked as the fifth highest plasticizer use in 1968 for Monsanto direct sales.[33] All told, on the order of 10 million pounds of PCBs may have been used in hot-melt adhesives for bookbinding, assuming 1/2 million pounds were used annually for a period of 20 years. Our results confirm the widespread presence of PCBs in the adhesives of these mass-produced paperback books.

Potential for Human Exposure. The concentrations of PCBs in paperback book adhesives raise human exposure considerations from inhalation of contaminated indoor air, ingestion of dust, and possibly direct dermal contact. Indoor air PCB concentrations are typically more than an order of magnitude higher than corresponding outdoor levels[1,2,49,50] because of emissions from PCB-containing indoor sources. Indoor PCB patterns are reported to resemble Aroclor 1242 in houses,[51] Aroclor 1248 in schools and houses,[52] and Aroclor 1016, 1242, and 1254 in schools.[49] Books are typically stored and used indoors where they are a possible indoor air PCB source.

Joint sealants (caulk), light ballasts, capacitors, paints, and other building materials are widely recognized as important Aroclor-containing sources of PCBs to indoor air in residential, occupational, and school settings.[1,2,49,52−54] Here, we evaluate the potential for indoor air impacts from paperback books using the screening-level PCB emission model of Guo et al. developed from laboratory chamber emission rate test results for caulk[44] by following the approach of Thomas et al.[53] (see Supporting Information, Methods section). Given the many model assumptions, a modest number of PCB-containing books could potentially lead to measurable indoor air concentrations. We estimate between 1 to 150 PCB-containing books could result in a bedroom indoor air concentration of 8 ng/m^3, while 6 to 1900 books result in a 100 ng/m^3 concentration.

Mass-market paperbacks were not manufactured for durability as were library editions,[31] and the current stock is likely much diminished from the estimated 6 billion paperback books produced between 1948 and 1972. However, we had little difficulty finding mass-market paperbacks manufactured during that era at bookstores, in many public library catalogues, and the stacks at the Library of Congress' paperback collection. Furthermore, the impact of a primary

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source can persist in secondary sources even after the primary source has been removed.[55] Accordingly, the potential for books to be a PCB source to indoor environments should be not be disregarded; the indoor air, ingestion of dust, and direct dermal contact pathways warrant further investigation.

Other Exposure Implications. The conventional view is that the major source of PCBs in the pulp and paper industry was from recycled carbonless copy paper containing 2.8% to 3.6% Aroclor 1242 and manufactured between the mid-1950s and 1972.[9] Like carbonless copy paper, appreciable quantities of book trim and books with PCB-containing adhesives could have been papermill pulp furnish. Although the earliest bookbinding hot-melt adhesives were not soluble in the alkali solutions used in pulping, alkali-dispersible hot-melt adhesives were available as early as 1950.[23,24] PCBs dispersed in alkali solutions could have transferred to the pulp itself, sludge, and effluent.

This study presents empirical evidence that percent-level PCBs were intentionally added to hot-melt adhesives used to manufacture mass-market paperback books. Although the sample size in this study is modest (n = 22), PCBs were detected in all the samples from books manufactured between 1948 and 1974, with intentional percent-level use of PCBs for years spanning 1949 and 1966. PCBs were not detected above the limit of detection (0.70 ppm) in one book from 1946. During the intentional use years, we estimate more than 6 billion paperback books were sold domestically. Those books still in use represent a potential open source of PCBs to humans and the environment. These findings confirm the presence of PCBs and PCTs in the adhesives of these mass-produced paperback books and demonstrate the need to consider book adhesives as an open PCB source to indoor air and the environment.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.9b00489.

Mass-market paperback sample information (Table S1), PCB Aroclor results (Table S2), bibliography of books sampled (Table S3), figures showing book covers, book end, copyright page, and GC-ECD chromatograms (Figures S1−S22), figures comparing chromatograms of PCT Aroclor standards to four samples (Figures S23−S24), and supporting methods including sample dating, preparation, analysis, and methods used to estimate the number of books to impact indoor air concentration with supporting tables (Tables S4 and S5). (PDF)

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Notes

The authors declare no competing financial interest.

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■ ABBREVIATIONS

PCB, Polychlorinated biphenyl; PCT, Polychlorinated terphenyl; ppm, parts per million; wt, weight; ng/m^3, nanograms per cubic meter

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