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Research Article

Lead and mercury in historical books and their contribution to dust contamination

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HIGHLIGHTS

- Pb and Hg measured by XRF in different components of 133 books from two collections.
- Pb concentrations above 1000 mg kg^{-1} in books published between 1850 and 1960.
- Hg above 1000 mg kg^{-1} in red panels, illustrations and edgings of Victorian-era books.
- Pb concentrations significantly higher in library dusts than household dusts.
- Historical books could act as a source of indoor Pb exposure.

GRAPHICAL ABSTRACT

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ABSTRACT

Historical products are often a source of toxic substances, like heavy metals, that have since been restricted. In this study, the lead (Pb) and mercury (Hg) content of 133 books housed in two collections in southwest England (a university library and council repository), and published between 1704 and 2018, have been determined on-site by X-ray fluorescence spectrometry. Lead was detected in the front panels, text blocks and interior colour illustrations of most books, with maximum concentration of 15,100 mg kg^{-1}, 8680 mg kg^{-1} and 12,800 mg kg^{-1}, respectively. However, concentrations above 1000 mg kg^{-1} were generally restricted to books published between about 1850 and 1960. Mercury was detected in fewer cases, but concentrations above 5000 mg kg^{-1} were found in the red panels, coloured illustrations and red fore-edging of books published in the Victorian era. Mean concentrations of Pb in dusts from council repository shelves (112 mg kg^{-1}) and library shelves (159–224 mg kg^{-1}) and light casings (71.7 mg kg^{-1}) were significantly higher than mean concentrations in household dusts from buildings constructed over the same period (24.8 mg kg^{-1}). The findings suggest that historical books could be a source of Pb exposure where collections are housed or sold and could also be used to improve evaluations of historical indoor pollution.

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1. Introduction

Many heavy metals (with a density exceeding five times that of water) are highly toxic and are known to induce multiple organ damage, even at relatively low levels of exposure [12,24,9]. Amongst the metals (and metalloids) of greatest concern from a health perspective are arsenic, cadmium, chromium (VI), lead and mercury. Consequently, strict regulations regarding their concentrations and exposure in the environment, workplace, food and consumer goods have emerged and evolved over the past few decades [17,19,3].

Despite contemporary restrictions, legacy contamination of the environment and the exposure to and circulation or recycling of historical products remain problematic. For instance, heavy metal contamination of soil, sediment and water is commonly encountered in the vicinity of abandoned, historical metal mines [1,2], while restricted metals are often reported in old extant paints and plastics [18,30]. Many current regulations and limit values are specific enough to exclude older products, but where they are more general, historical articles cannot necessarily be neglected. This issue was raised when new Pb limitation standards were applied in the US by the Consumer Product Safety Commission (CPSC) to anything used by children, including articles manufactured historically [21]. Amongst the articles referred to, but for which no quantitative information could be provided by the CPSC or concerned respondents, were old books.

Recently, the Poison Book Project began to analyse the bookcloths of Victorian-era books housed in the Winterthur Library collection (Delaware) by X-ray fluorescence (XRF) spectrometry [25,26]. The main focus thus far appears to have been on As, prompted by the ready flaking of the toxic, bright green colourant, emerald green (copper acetoarsenite), from the rustic adornments of several nineteenth century books (and up to concentrations of 43,000 mg As kg^{-1} or 1.42 mg As cm^{-2}). A subsequent study of green, Victorian-era books from Northwestern University Libraries (Illinois) revealed that As-based pigments and their (arsenate) degradation products could be transferred to adjacent books, thereby extending the reach of potential contamination and exposure [31]. While these studies also mention the occurrence and measurement of other heavy metal pigments (based on Cr, Hg and Pb) in the collections, quantitative data have not been reported to date.

In the present study, a wider selection of older books published across a more expanded timeframe and housed in two collections in Plymouth, southwest England, has been analysed by XRF for various heavy metals, but with the focus on Pb and Hg. Measurements are taken of the book covers (front panels), as well as the interior text blocks and any coloured interior illustrations or fore edgings. In order to evaluate the potential of books to act as a source of exposure through inhalation and ingestion, Pb and Hg concentrations are also measured in samples of settled dust. Specifically, concentrations in samples from the vicinity of the collections are compared with concentrations in dusts from areas more remote from older books and in independent households constructed during the same era as the collection buildings.

2. Experimental section

2.1. Study sites

Two book collections were considered in the present study. Firstly, a collection of about 3000 old and valuable books housed in a small study room (about 35 m^2 in floor area and 3 m in height) at the Charles Seale-Hayne (CSH) library on the main campus of the University of Plymouth. Here, books are stored on shelves in locked but ventilated stainless steel cabinets and are available for consulting on request. The room itself is locked but is available for booking for student study. Secondly, a large book depository (> 1000 m^2 in area) on the second, open-plan floor of a modern building in an industrial park about 5 km outside Plymouth City centre. Here, a range of tens of thousands of books serving Plymouth City Council libraries is stored on steel rolling stack shelving (i.e., shelving mounted on wheeled carriages attached to a rail system).

2.2. Book measurements

A total of 66 books from the CSH library and 67 books from the council repository were measured on site by portable XRF spectrometry using a Niton XL3t GOLDD+ . Books were selected that encompassed a range of ages, cover colours and types (thick cardboard or cardboard covered with cloth, clothette or leather), page numbers, and content (e.g., fiction, non-fiction, local history, children's, poetry, artwork, serials). Each book was photographed (examples are shown in Fig. 1) and the title, author, publisher and place of publication recorded. (See supplementary information for details of all books analysed.).

Concentrations of various toxic elements (As, Cd, Cr, Pb, Hg) were determined on the front cover or panel and through the pages (text block; see Fig. 1d), with coloured, internal artwork or fore edging also measured on selected books. In order to minimise operator exposure to radiation, books were cradled in a radiation apron on a wooden bench, and to eliminate any fluorescent interference from the apron, a new, "clean" book (with no measurable metals of interest) was placed below the sample book. Measurements of the book covers or any artwork or edging were performed for about 20 s in a standardless plastics mode (and about 10 s each in the main and low energy bands) with a 0.1 mm thickness correction applied. Measurements of the text block (> 15 mm) were performed through the centre of the first printed page for about 20 s in plastics mode but without thickness correction. Plastics mode was selected because of the low density, polymeric (cellulose and lignin) makeup of most book materials.

2.3. Dust collection and measurements

In the CSH library, deposited samples of fine dust were taken from the top cover and two lower shelves of one book cabinet and the tops of hanging light casings in the study room, the light casings in the office space directly below the study room, and the empty top shelves in the centre of the main library collection. A thin plastic card (8 cm × 5 cm) was used to scrape and gather material from about 0.5–1 m^2 of smooth and level surfaces and, with the aid of a clean sheet of white paper, resulting accumulations were carefully siphoned into a series of labelled polyethylene XRF sample cups (Chemplex series 1400; 21-mm internal diameter, 20 mm depth). Dust samples were compacted with a glass rod before being collar-sealed with 3.6 µm SpectraCertified Mylar polyester film. In the book depository, a sample of dust from about 1 m^2 of the top casings of the stack shelves in the centre of the collection was gathered and processed likewise.

An additional ten composite samples of dust from vacuum cleaner bags were supplied by residents of houses in Plymouth that had been constructed within the past 40 years (the approximate age of the CSH library). After removing any large fragments or hairs with tweezers, samples were compacted and sealed in XRF cups as above.

At five different locations (CSH library and repository samples) or in the centre (all household dusts), samples were measured through the Mylar film by the portable XRF spectrometer housed in a laboratory test stand for a total counting time was 300 s (comprising 250 s in the main energy band and 50 s in the low energy band) in plastics mode without thickness correction.

2.4. Metals of interest and XRF performance

While in the test stand, the performance (accuracy) of the XRF spectrometer in the plastics mode was evaluated by quintuplicate analyses of two, 13-mm thick polyethylene discs that had been uniformly impregnated with Pb and Hg (Niton PN 180–554: Pb = 1002 ± 40 mg kg^{-1} and Hg = 1000 ± 40 mg kg^{-1}; Niton PN 180–619: Pb = 150 ± 12 mg kg^{-1} and Hg = 101 ± 10 mg kg^{-1}). Mean concentrations returned were within 7% of the respective reference values for both discs

Fig. 1. A selection of the books in the CSH library that were measured by XRF. (a) Various books from the late nineteenth to early twentieth century, exemplifying coloured illustrations and decorated and embossed covers, (b) a cardboard-bound children's annual from 1950, (a) red fore edging on a book published in 1865, and (d) the front page of a book published in 1868 showing, as a red circle, the approximate location where the text block was measured.

(Pb = 1018 ± 4.1 mg kg^{-1} and Hg = 936 ± 8.5 mg kg^{-1}; Pb = 149 ± 4.0 mg kg^{-1} and Hg = 99.1 ± 1.7 mg kg^{-1}).

Detection limits, as three counting errors, varied for the books measured handheld on site depending on the book thickness and whether thickness correction had been applied or not. For Pb, values ranged from 7.9 to 246 mg kg^{-1} with a median of 11.4 mg kg^{-1}, while for Hg, values ranged from 7.8 to 387 mg kg^{-1} with a median of 14.9 mg kg^{-1}. For the dust samples measured for a longer period in the test stand, mean detection limits for Pb and Hg were 5.0 and 12.2 mg kg^{-1}, respectively.

2.5. Statistical analysis

Differences in concentrations of Pb or Hg between collections and between panels, text blocks and dust samples were established in Minitab *v*19 after undertaking Anderson-Darling tests for normality. An α value of 0.05 was used as the criterion for statistical significance.

3. Results

3.1. Book details

A total of 133 books were measured by XRF from the two collections, with 124 hard-backed (nine paper-backed), fourteen containing colour illustrations, and six coloured on the fore edges (five in red, one in gold).

The age distribution of the books is shown in Fig. 2. The period of publication spans three centuries, with a median year of publication at the beginning of the twentieth century (and 1890 for the CSH library and 1916 for the council repository). Where stated, the majority of books were published in London (n = 97), but other places of publication included Barnsley, Cardiff, Edinburgh, Exeter, Glasgow, Loughborough, Oxford and Plymouth (UK), New York, Paris and Philadelphia. All but three books were published in English.

The distribution of the main colours of the front panels and that were measured by XRF is shown in Fig. 3. Red, brown and green were the most common colours and comprised > 60% of all books considered.

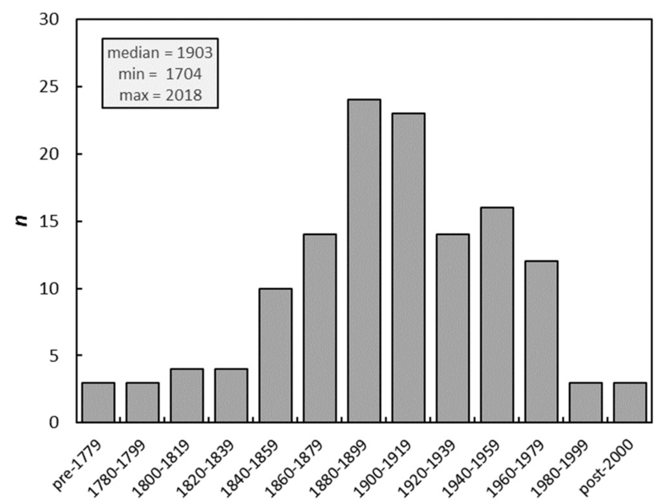

Fig. 2. A histogram illustrating the age distribution of the books measured.

3.2. Pb and Hg concentrations in book components

Table 1 shows the number of front panels and text blocks in which Pb and Hg were detected in books from the CSH library and council repository, along with statistical summaries and distributions of metal concentrations in each case. (The full data set for these metals, including counting errors, can be found in the supplementary information.) According to a signed rank Wilcoxon test, median Pb concentrations were significantly higher in the panels than the blocks in both collections, while according to Mann-Whitney U tests, median Pb concentrations were higher in the panels and blocks of books from the CSH library than in the respective components of books from the council repository. For Hg, there were only four books where the metal was detected in both the panel and block but Mann-Whitney U tests revealed significantly higher median Hg concentrations in the panels and blocks from CSH library than in the respective components from the council repository.

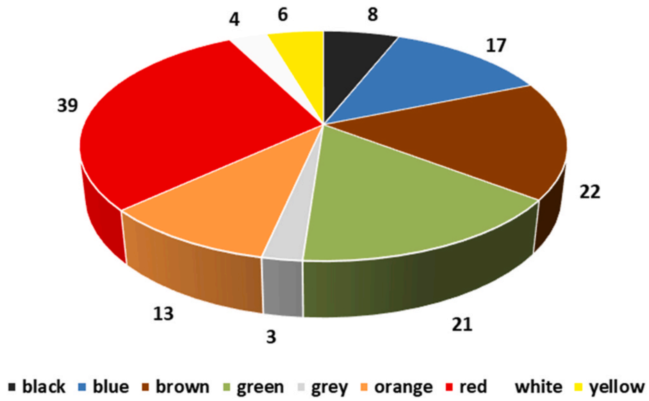

Fig. 3. Number of books according to the main (and measured) colour of the front panel.

Table 1
Number of detects (*n*) and summary statistics and distributions of Pb and Hg concentrations in the front panels and text blocks of the books from the CSH library and council repository.

	[Pb]-panel	[Pb]-block	[Hg]-panel	[Hg]-block
CSH library				
n	61	65	13	7
median, mg kg^{-1}	427	180	54.5	121
min, mg kg^{-1}	18.5	13.0	12.2	17.2
max, mg kg^{-1}	15,100	8680	24,500	994
$n < 100$ mg kg^{-1}	8	24	8	2
100 mg kg^{-1} $< n < 1000$ mg kg^{-1}	38	33	2	5
1000 mg kg^{-1} $< n < 10,000$ mg kg^{-1}	14	8	1	0
$n > 10,000$ mg kg^{-1}	1	0	2	0
Council repository				
n	60	61	4	5
median, mg kg^{-1}	133	74.0	27.4	22.6
min, mg kg^{-1}	10.2	12.8	9.1	11.5
max, mg kg^{-1}	5290	1040	627	201
$n < 100$ mg kg^{-1}	24	41	3	4
100 mg kg^{-1} $< n < 1000$ mg kg^{-1}	21	19	1	1
1000 mg kg^{-1} $< n < 10,000$ mg kg^{-1}	15	1	0	0
$n > 10,000$ mg kg^{-1}	0	0	0	0

Fig. 4 shows the concentrations of Pb and Hg in the front panels and text blocks of books from both collections as a function of the year of publication. In the panels, concentrations of Pb exceeded 1000 mg kg^{-1}, the threshold according to the current Restriction of Hazardous Substances (RoHS) Directive [6], in the oldest book (published 1704; red), and in books of various colours but usually green, red, yellow or orange, published between 1828 and 1961. In the blocks, Pb concentrations exceeded 1000 mg kg^{-1} in books published between 1847 and 1950.

Mercury concentrations exceeded 1000 mg kg^{-1}, its threshold according to the RoHS Directive [6], in two red panels of books published in 1868 and 1890, and in the block of one book published in 1878. Closer inspection of the text block results revealed that many of the highest metal concentrations were associated with colour illustrations and/or coloured text in parts of or throughout the book (and as indicated in Fig. 4). Measurements of the coloured parts of individual pages returned concentrations of Pb up to 20,000 mg kg^{-1} and Hg up to 6400 mg kg^{-1}, with multiple measurements of the same page often revealing different concentrations according to colour. In all cases where one or more concentration of Pb or Hg in coloured illustrations or text exceeded 1000 mg kg^{-1}, books had been published between about 1850 and 1950.

Of the six edgings measured, two coloured red returned Hg concentrations of 6200 mg kg^{-1} and 7500 mg kg^{-1}, and one in gold returned a Pb concentration of 435 mg kg^{-1}. All of these books were published in the 1860s

In books published since the mid-1960s, concentrations of Pb did not exceed 100 mg kg^{-1} in any component measured, while in books published since the mid-1950s, Hg was never detected.

3.3. *Pb and Hg concentrations in dust samples*

Table 2 summarises the concentrations of Pb and Hg in the dust samples from the CSH library, council repository and Plymouth households. Mean concentrations of Pb ranged from about 20 mg kg^{-1} in the office area directly below the study room of the CSH library to over 220 mg kg^{-1} on the bottom shelf of one of the bookcases in the study room. According to one-way ANOVA and Tukey's post hoc test, mean Pb concentrations were significantly higher in all samples from the study room than other parts of the CSH library and the composite household dust samples, and mean Pb concentration was significantly higher in the council book repository than in the household dusts.

Mean concentrations of Hg in dusts ranged from 13 mg kg^{-1} in Plymouth households to 56.7 mg kg^{-1} on the bottom shelf of a book case in the study room. According to one-way ANOVA, the mean concentration of Hg in household dusts was significantly lower than concentrations in all other dust samples.

4. Discussion

Interest in the chemical makeup of book covers has thus far focussed on the Victorian era. This is because until the end of the nineteenth century, bleached cotton bookcloths were backfilled with various materials that often included a brightly coloured pigment, whose chemical makeup was never disclosed [27]. For many bright green books of this period, XRF and Raman analyses have revealed that the toxic pigment, copper acetoarsenite, was employed [25]. In our study, however, and once overlap of the As-Kβ fluorescent line by the Lα line of Pb had been accounted for [8], As was only detected above 100 mg kg^{-1} in the presence of Cu in the green binding of a serial published in 1852 (As = 410 mg kg^{-1}; Cu = 312 mg kg^{-1}).

More important in the covers we analysed were Pb and Hg. Significantly, Pb appeared to have been employed well beyond the Victorian era (and from about 1830 to the 1960 s), and on panels that were covered in a range of materials as well as being constructed solely of thick cardboard. Books containing these metals were both collectible and more widely available and included those intended for children. Lead and Hg were also found in the text blocks, and in particular where coloured illustrations or text was used, and in coloured fore edging.

In many cases where panels or internal illustrations were brightly coloured, high concentrations of Pb were associated with high concentrations of Cr. In Fig. 5, there appear to be two distinctive and statistically significant relationships between Pb and Cr (where concentrations of both metals exceeded 200 mg kg^{-1}), with gradients of about 20 and 1.7. Given the stoichiometry of Pb:Cr on a mass basis in lead chromate is 4.0, this suggests that mixed phase chromates (e.g., $PbCrO_4 \cdot PbSO_4$ and $PbCrO_4 \cdot PbO$) or additional Pb- or Cr-based compounds may have been used in the colourants. We also noted an association of high concentrations of Pb and Sn in many panels of different colours (with or without Cr), suggesting the use of lead-tin yellow (Pb_2SnO_4 or $Pb(Sn,Si)O_3$), although a lack of detectable Sb suggests that Naples yellow ($Pb_2Sb_2O_7$) was not employed. Where Pb was detected in panels or illustrations in the absence of other metals, red lead (Pb_3O_4) may have been employed as a pigment, or other compounds (e.g., litharge, PbO, white lead, $2PbCO_3 \cdot Pb(OH)_2$, or lead acetate, $Pb(CH_3COO)_2$) could have been used as siccatives, dye fixatives or opacifying agents [4,26,11].

It is also possible that the Pb detected through the text blocks of many books results from lead compounds employed as ink driers or

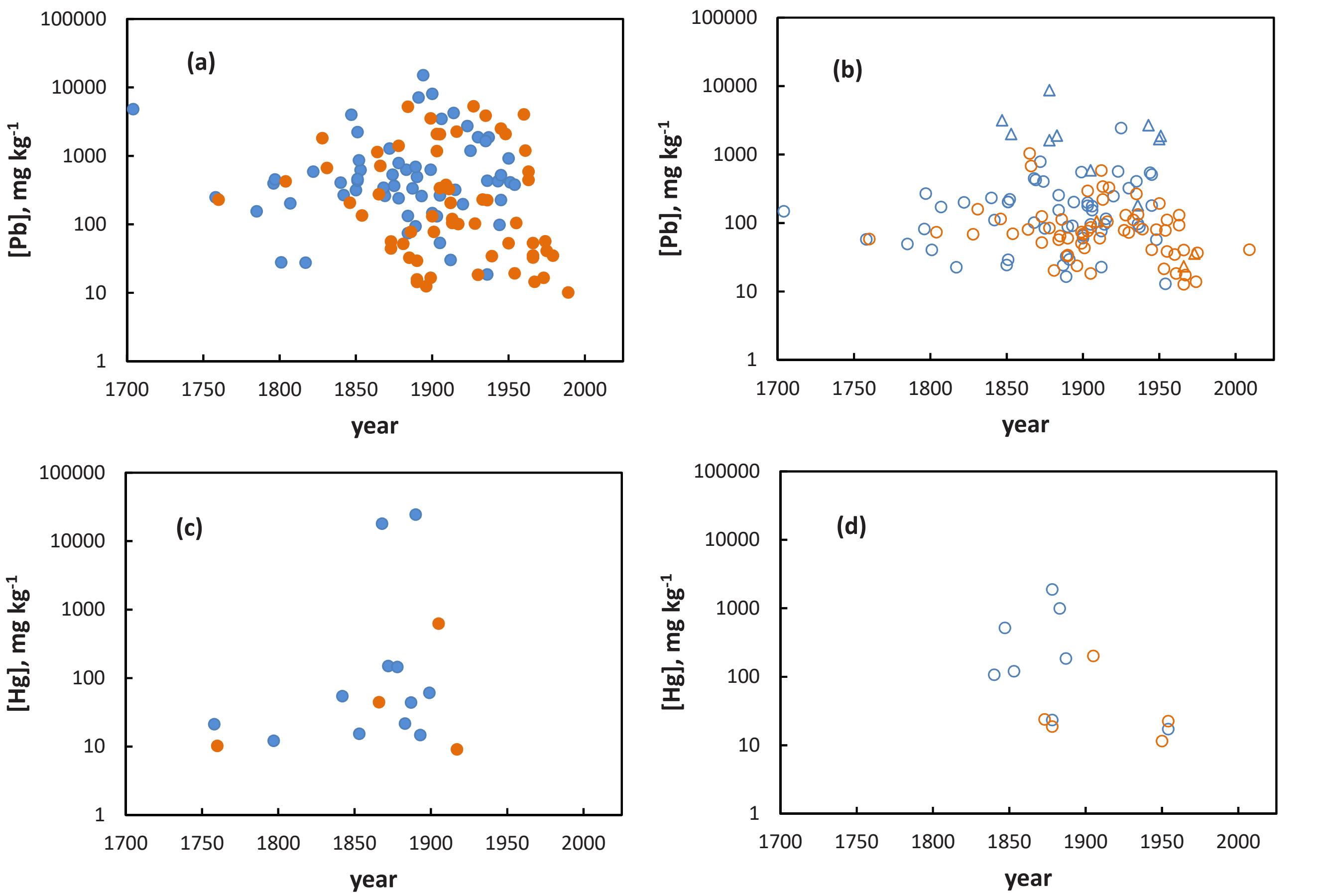

Fig. 4. Concentrations of Pb in (a) front panels and (b) text blocks, and Hg in (c) front panels and (d) text blocks, as a function of year of book publication. Blue symbols denote books from the CSH library and orange symbols denote books from the council repository; triangles indicate books that contained coloured illustrations in part or throughout the text block.

Table 2
Mean (± one standard deviation) of Pb and Hg concentrations in samples of dust from the CSH library, books repository and Plymouth households.

Location	Pb, mg kg^{-1}	Hg, mg kg^{-1}
CSH library study room		
light casings ($n = 5$)	71.7 ± 3.3	30.4 ± 7.0
bottom shelf of book case I ($n = 5$)	224.2 ± 10.8	56.7 ± 18.6
bottom shelf of book case II ($n = 5$)	158.9 ± 67.2	27.8 ± 5.4
CSH office space		
light casings ($n = 5$)	19.8 ± 4.6	19.5 ± 3.5
CSH main library		
top shelving ($n = 5$)	40.2 ± 12.9	22.7 ± 5.7
Council repository		
top casings ($n = 5$)	112.2 ± 16.9	25.8 ± 6.6
Plymouth households		
hoovered composites ($n = 10$)	24.8 ± 14.3	13.0 ± 1.6[a]

[a] Hg detected in five out of ten samples.

fixatives [5] or from contamination of the printing press [14]. In many cases, however, we also detected similar quantities of Pb through the margins of text blocks where there was no text, suggesting that the paper itself may also be subject to contamination during its manufacture.

Regarding Hg, concentrations above 500 mg kg^{-1} were encountered in the red covers of two books published in 1868 and 1890, in the red or brown internal illustrations of four books published between 1847 and 1883, and in the red edging of two books published in 1865 and 1868. These observations reflect the common usage of the bright red pigment, vermilion (HgS), during the Victorian era [26]. Subsequently, and possibly because of costs and poor lightfastness of some varieties,

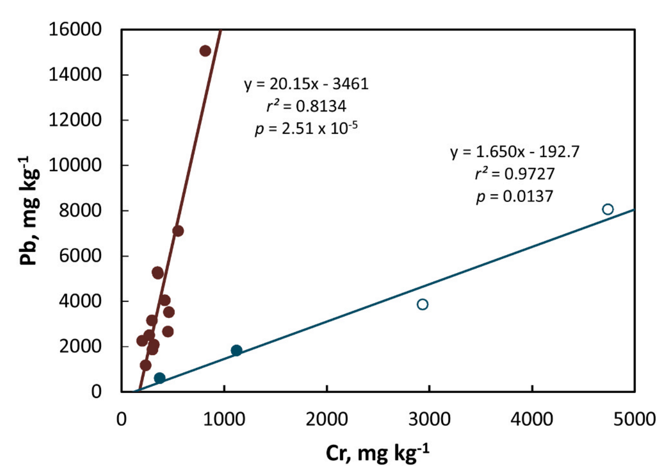

Fig. 5. Concentrations of Pb (> 200 mg kg^{-1}) versus concentrations of Cr (> 200 mg kg^{-1}) in the front panels and (open symbols) coloured illustrations of books from both collections. Regression equations define two groups of data (coloured differently).

vermilion was substituted by red lead, lead chromate and cadmium sulphoselenide pigments [10], although no evidence for the use of Cd in books was found in the present study.

From a health perspective, the greatest risk is from Pb and Hg in the pages and covers of books intended for children (e.g., Fig. 1b), and in particular those in poor condition. This concern was hypothesised when new Pb (but not Hg) limitation standards were applied in the US to anything used by children, including articles manufactured historically [21]. The Consumer Product Safety Commission (CPSC) seemed to suggest that retailers or resellers of books need only act where there was likely to be a high Pb content. However, they also suggested, without any evidence, that Pb concentrations in older books was typically around 300 mg kg^{-1}. Moreover, a compilation of comments and objections made to the CPSC, including from the Association of American Publishers, failed to provide any quantitative information on Pb in historical books [7]. To this end, therefore, the current study is significant in providing a concentration range for Pb in panels and blocks of circulating books (including those used by children) and a publication timespan of greatest concern.

It is unclear whether the observations pertaining to Pb in the present study are more generally applicable to books published globally and in different languages or to a wider range of printed media. For example, an analysis of newspaper and magazine cuttings from the late 1960s and early 1970s found levels of Pb up to 35 mg kg^{-1} in black and white pages (similar to the concentrations found for this period in this present study) but concentrations up to 4000 mg kg^{-1} in coloured pages. This indicates that colourful, lead-based pigments were not completely eliminated from reading material until later than the results above suggest.

A more general source of exposure to Pb and Hg from books, and from both inhalation and ingestion, is via contaminated dust. The covers of older books, and especially those covered in cloth, clothette or leather, become friable over time and abrasion can lead to the formation of particulate matter [25]. On many shelves housing older books, and particularly in the CSH library, coloured debris derived from covers was clearly visible. Depending on storage and environmental conditions, the text blocks are also subject to deterioration through mould growth and damage from insects that feed on cellulosic fibres or starch-based adhesives [20]. The results of our dust analysis indicate an increase in Pb content on the shelving in the study room of the CSH library by up to an order of magnitude and in the council repository by a factor of over four compared with household dusts from residential buildings constructed during the same era. Dust contaminated with Pb was also evident on the local light casings in the study room of the CSH library and, to a lesser but measurable extent, in open areas where more general book collections are housed. With respect to Hg, all dust concentrations determined are at the high end of values reported in the literature for urban households [15] but, unlike Pb, the performance of the XRF has not been assessed at such levels in this type of matrix [29]. Nevertheless, and at least qualitatively, the results suggest moderate Hg contamination in the study room of the CSH library compared with remaining locations. Contaminated dust could arise directly from the abrasion and dispersion of Hg-bearing particles, or indirectly through the off gassing of Hg vapour and its subsequent adsorption to ambient particulate matter [22].

Contaminated dust is predicted to pose the greatest risk where large public or private collections of old books are housed or in shops that specialise in second-hand books, and here, therefore, regular cleaning to minimise dust exposure is recommended. More widely, smaller collections are likely to contribute to the Pb burden of household dust, along with other, well-documented sources such as old interior paint and contaminated-tracked in soil [13,28]. To this end, the findings of the study may also assist in assessing historical indoor air pollution from and exposure to Pb and Hg [16,23].

CRediT authorship contribution statement

Andrew Turner: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Writing – review & editing.

Environmental Implication

Historical books contain compounds of lead (Pb) and mercury (Hg) in the text blocks, covers, interior illustrations and coloured edgings. On abrasion and aging, material can contaminate indoor dusts and act as a source of indoor air pollution and human exposure. Contamination may be a problem in some contemporary settings, but can also be used to evaluate historical indoor air pollution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

Acknowledgements

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.131981.

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