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Lead in painted surfaces and dusts from rented urban properties (Plymouth, UK)[☆]

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ABSTRACT

X-ray fluorescence (XRF) spectrometry has been used to measure lead (Pb) in painted surfaces and indoor dusts from 14 rented properties in the city of Plymouth, UK. Lead was detected in 78 out of 164 surfaces measured (and in 69 out of 150 interior surfaces) with an overall median concentration of 7100 mg kg^{-1} (range from 157 to $139,000 \text{ mg kg}^{-1}$), and was detected in all but two properties that included one building constructed within the last 25 years. Selected measurements on an area basis ($n = 48$) returned Pb concentrations ranging from below 0.01 to 16.1 mg cm^{-2} that were significantly correlated with concentrations on a mass basis but with scatter that was attributed to the degree to which leaded paint had been overpainted. As potential measures of Pb exposure, mean concentrations in samples of lint ($n = 8$) were $<15 \text{ mg kg}^{-1}$ while mean concentrations in hoovered dusts ($n = 14$) were more variable; specifically, mean Pb dust concentrations ranged from ~ 20 to 140 mg kg^{-1} in 13 samples but was $\sim 4500 \text{ mg kg}^{-1}$ in a property that had recently undergone extensive renovation. Although mean concentrations of Pb in lint or dust were not related to median Pb concentrations in paint, a strong correlation between barium (Ba) and Pb in dusts suggests that paint is the main source of dust Pb. This study indicates that, in most cases, leaded paints historically applied to interior surfaces have been over-coated and pose little risk, provided that surface coatings are in good condition. However, inappropriate practices during renovation can result in significant contamination of dusts. Our findings are likely to be more broadly applicable to the urban rented sector in the UK and landlords and tenants should, therefore, be made aware of government guidance on the safe decoration or remodelling of older properties where leaded paint is pervasive.

1. Introduction

Because of its malleability, ductility, low melting point, high corrosion resistance and high density, lead (Pb) has had a wide variety of uses, including in mass marketed consumer products. However, because of its diverse acute and chronic toxicities, and in particular its health impacts on young children, the heavy metal has been phased out, replaced with safer alternatives or restricted in many sectors (Gulson et al., 1995; Lanphear and Roghmann, 1997; Glorennec et al., 2010). Lead was, historically, one of the most important pigments in household paints, making applications more durable, adherent, crack-resistant and colourful (Gooch, 1993; O'Connor et al., 2018). Although many different pigments were used, Lucas et al. (2012) suggest that applications on non-metallic household surfaces are likely to be basic lead carbonate, $2\text{PbCO}_3 \cdot \text{Pb}(\text{OH})_2$ (white lead), while those on metallic surfaces are likely to be lead tetraoxide, Pb_3O_4 (red lead).

In 1963, and with an increasing awareness of the hazards of Pb, the Ministry of Health and the Paintmakers Association of Great Britain, now the British Coatings Federation, voluntarily agreed that paint containing more than $10,000 \text{ mg kg}^{-1}$ of Pb by weight in the dry film should be labelled with a warning that it should not be applied to surfaces which children might chew (Johnson et al., 1984). A subsequent European Community Directive (1977) required all paint containing more than 5000 mg kg^{-1} of Pb to be inaccessible to children. In 1992, the Council of the European Communities (1989) Directive was implemented in the UK that prevented the intentional addition of white lead to consumer household paints. Red lead was not referred to in this legislation but the compound has gradually been replaced by safer alternatives.

Despite these restrictions, older properties, or, strictly, any properties built before 1992, may contain legacy layers of leaded paints that remain exposed or that have been over-coated. Leaded paints are

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generally safe if the top layer is in good condition and older paints have been sealed or encapsulated (O'Connor et al., 2018). However, exposure to harmful levels of Pb may take place if paint is disturbed or removed during repair or renovation practices that generate dust. According to Jaeger et al. (1998), where lead has long been eliminated from petrol and safe, non-leaded plumbing systems have been installed, interior paints may well represent the most important non-occupational source of Pb exposure. This type of exposure is also believed to be accentuated for minority groups and socio-economically disadvantaged adults and children, including those who rent private accommodation.

In spite of the wide, historical use of leaded paints in UK domestic properties, published information on the concentrations and distributions of Pb appears to be limited to four samples analysed nearly 40 years ago (Sturges and Harrison, 1985). Accordingly, we investigate the presence and concentrations of Pb in interior painted surfaces of a range of properties in the city of Plymouth, southwest England, in situ and non-destructively, by hand-held X-ray fluorescence (XRF) spectrometry (Turner et al., 2016; Tighe et al., 2020; Zaharieva et al., 2022). We also measure the concentrations of Pb in some exterior paints, and in household dust and laundry drier lint as proxies for exposure in order to evaluate any relationships between legacy sources and contemporary receptors. We focus on rented accommodation because of the potential for elevated exposures for tenants implied above, and because recent UK guidance for both landlords and tenants directly refers to the risks from poorly maintained leaded paint (UK Government, 2020; Landlordvision, 2021).

2. Methods

2.1. Properties under study

Fourteen private, rented properties within about 2 km of the city centre of Plymouth were selected for study, with relevant information shown in Table 1. Properties ranged from about 20 to 160 years old (median age = 139 years) and amongst them were a grade II listed building (#3), a converted public house (#8) and a converted nursing home (#10). All properties were contained in terraced blocks, and while most were close to the pavement and road and had back yards, #1 and #4 were fronted by a small area of lawn and bare soil. Inside, properties had a combination of carpeted and uncarpeted (mainly vinyl, hard wood, engineered wood and laminate) flooring. The number of occupants ranged from two to ten and while most properties housed university students, some housed additional, longer-term tenants, including young children. Also shown in Table 1 are the general condition of painted surfaces in each property and whether a high efficiency

Table 1

Ages and occupancies of the rented properties, along with painted surface condition and whether a HEPA filter was used for collecting dust. Good = new or clean paint with only minor scuffs; fair = scuffs and limited peeling; poor = more extensive but localised damage and peeling; very poor = extensive damage and peeling.

Property no.	Built	No. occupants	Paint condition	HEPA filter
1	1857	10	good	n
2	1857	7	fair	y
3	1867	6	fair	n
4	1857	10	good	n
5	1862	4	poor	y
6	1862	3	good	y
7	1895	2	very poor	n
8	1862	6	fair	n
9	2001	5	fair	n
10	1895	5	good	y
11	1895	6	fair	y
12	1895	3	fair	y
13	1928	3	good	y
14	1928	5	poor	y

particulate air (HEPA) filter was installed on any dust-collecting device.

2.2. In situ lead measurements

Lead was measured in situ on painted wooden, metallic, plastered and masonry surfaces, including some varnished wood, using a hand-held Thermoscientific Niton (UK) He GOLDD+ XL3t X-ray fluorescence (XRF) spectrometer. Surfaces were measured in at least three rooms in each property and included interior walls, ceilings and ceiling beams, skirting boards, floors, stairs, banisters and stair spindles, fire-places, built-in cupboards, headboards and shelves, picture rails and other decorative features, window sills and frames, pipes, and doors and door frames. In some properties, exterior painted features that could impact on interior dust Pb, like steps, drainage pipes, walls, railings and outbuilding doors, were also measured.

For each surface (164 in total; 150 interior and 14 exterior), the nose of the XRF, housing the measurement window and proximity sensor and, where possible, fitted with a backscatter shield, was positioned firmly against a clean (or cleaned), homogeneous area of about 10 mm in diameter. Measurements were performed by depressing the trigger mechanism of the instrument for a period of about 10–15 s in a standardless “plastics” mode (40 μ A and 50 kV) and with a thickness correction of 50 μ m and a beam collimation of 3 mm. Fluorescent X-ray peaks for Pb L_{α} and Pb L_{β} at 10.5 and 12.6 keV, respectively, and for K and L lines of a variety of lighter elements, were converted to mg kg⁻¹, the unit of measurement for paint most commonly referred to in the UK, and with an error of 2 σ , using Niton fundamental parameters software. For 48 surfaces that returned variable concentrations of Pb, measurements were repeated for 10 s using a “lead paint” mode (40 μ A and 50 kV) that provides concentrations in mg cm⁻², the unit of measurement preferred in the US, along with a depth index (from 1 to 10) that yields a semi-quantitative indication of the amount of non-leaded paint covering the Pb detected.

2.3. Lead measurements of dust and lint

Each household supplied a composite sample of Hoover dust (~30 g) collected from the entire property and, where tumble driers were in use ($n = 8$), a sample of laundry drier lint (~20 g). In the laboratory, samples were cleared of any large, visible pieces of debris and hairs using a pair of tweezers. However, we avoided sieving through a fine mesh as this process may remove significant fractions of paint particles which are important contributors to Pb dust through their ready fragmentation (Hunt et al., 1993). Samples were subsequently compacted into polyethylene XRF sample cups (Chemplex series 1400; 21-mm internal diameter) to a depth of 20 mm and collar-sealed with 3.6 μ m SpectraCertified Mylar polyester film. Each sample was analysed at six different locations by the XRF spectrometer housed in a laboratory test-stand, with counting for 60 s in the plastics mode but without thickness correction.

2.4. Quality assurance

While in the test stand, the performance of the XRF was evaluated by analysing, in sextuplicate, a series of reference materials, with the results summarised in Table 2. Analysis of three lead paint films returned concentrations in mg cm⁻² that were within 6% of reference concentrations and precisions that were better than 10%. These paints also returned depth indices ranging from 1.03 to 1.24. A plastic disc that had been uniformly impregnated with Pb returned a concentration in mg kg⁻¹ that was within 5% of the reference value and with a precision that was better than 1.5% when analysed in plastics mode. A reference soil was also analysed for Pb in plastics mode in order to evaluate the potential impacts of porosity and higher net material density in the dust and lint samples. Because the mean measured concentration was 17.9% lower than the reference value, a corresponding correction factor was

Table 2

Results of the sextuplicate analysis of various reference materials. Errors are one standard deviation about the mean.

Reference material	Mode	reference value	measured value
2579a (yellow paint)	lead paint	$3.58 \pm 0.39 \text{ mg cm}^{-2}$	$3.51 \pm 0.10 \text{ mg cm}^{-2}$
2579a (red paint)	lead paint	$1.04 \pm 0.06 \text{ mg cm}^{-2}$	$1.10 \pm 0.10 \text{ mg cm}^{-2}$
2579a (green paint)	lead paint	$0.307 \pm 0.021 \text{ mg cm}^{-2}$	$0.320 \pm 0.100 \text{ mg cm}^{-2}$
PN 180–554 (polyethylene disc)	plastic	$1002 \pm 40 \text{ mg kg}^{-1}$	$954 \pm 14 \text{ mg kg}^{-1}$
MSH-100 (soil)	plastic	$1070 \pm 5.46 \text{ mg kg}^{-1}$	$878 \pm 39 \text{ mg kg}^{-1}$

applied to measured Pb concentrations in the samples.

Limits of detection, as $2\sigma \times 1.5$, ranged from about 18 to 50 mg kg^{-1} in paints measured in situ, and from about 6 to 12 mg kg^{-1} for dust and lint samples measured in the test stand.

3. Results

The results of the in situ Pb measurements are reported in Table 3. About one-half of all surfaces measured contained detectable Pb, including nine out of fourteen exterior surfaces and one out of three interior varnished surfaces. Overall, detectable concentrations ranged from 157 to 139,000 mg kg^{-1} , with a median concentration of about 7100 mg kg^{-1} ($n = 78$). Concentrations greater than 100,000 mg kg^{-1} were encountered in paints on a banister, two (interior) plastered walls and an exterior down pipe. Two properties contained no detectable Pb on painted (or varnished) surfaces whereas six properties contained detectable Pb on at least 50% of surfaces measured and five properties had a median detectable Pb concentration exceeding 10,000 mg kg^{-1} . For comparison, the median concentration for all interior surfaces where Pb was detected was 7410 mg kg^{-1} ($n = 69$) while the median concentration for all exterior surfaces was 5640 mg kg^{-1} ($n = 9$).

Concentrations of Pb on painted surfaces that were determined on an area basis are shown in Fig. 1 along with corresponding depth indices. Concentrations range from 0.05 to 16.1 mg cm^{-2} and depth indices range from 1, where lead paint is close to the surface, to 10, where Pb paint is covered by multiple coatings of newer paints. Measures of Pb concentration on a mass basis and on an area basis were significantly related but, because of the variable depths of leaded paint, displayed considerable scatter (Fig. 2).

Concentrations of Pb in household dust and lint samples are shown in Table 4. Mean detectable concentrations in the former range from 19.0 to 4520 mg kg^{-1} , with an overall median concentration of 92.4 mg kg^{-1} . The variability within individual samples reflects the heterogeneity of Pb within the dusts and, as relative standard deviation, ranges from about 10 to 60%. Lead was detected in three out of eight lint samples, with mean concentrations close to the lowest detectable concentration reported for dust and relative standard deviations always below 12%.

Table 3

Number of surfaces analysed in each property, including the number returning positive Pb readings, and distributions and statistical summaries of Pb concentrations on a mass basis. Figures succeeding E and V denote the number of surfaces that were external to the property or that were varnished, respectively.

Property no.	No. analyses	No. Pb detected	90–5000	5000–50000	>50,000	median	min	max
1	16 (E2)	13 (E2)	6	7	0	5080	350	26,700
2	13 (E2, V1)	8 (E1, V1)	8	0	0	485	157	4890
3	9	1	1	0	0	752	752	752
4	11 (E1, V1)	9 (E1)	3	5	1	8070	401	61,000
5	11 (E2)	8 (E1)	0	5	3	31,300	6720	139,000
6	7 (E1, V1)	0	0	0	0			
7	12	5	2	2	1	10,400	301	124,000
8	10	5	0	5	0	17,900	7400	19,800
9	8	0	0	0	0			
10	13	6	1	4	1	19,200	1020	61,000
11	13 (E4)	4 (E4)	1	2	1	28,600	874	133,000
12	10 (E1)	3	1	2	0	8100	752	22,000
13	15 (E1)	6	4	1	1	3150	507	58,500
14	16	11	4	5	2	5600	626	60,400

4. Discussion

According to the EC Directive (1977) and the US Residential Lead-Based Paint Hazard Reduction Act (USEPA, 1995), paint was originally defined as Pb-based or a health concern to children at concentrations exceeding 5000 mg kg^{-1} , with the latter regulation assuming an equivalent limit on an area basis of 1 mg cm^{-2} . A total of 48 out of 79 surfaces in which Pb was detected, and including 44 interior surfaces that encompassed all substrates considered and ten out of fourteen properties tested, exceeded the threshold on a mass basis; on an area basis, 76% of surfaces selected for measurement, but based on detectable Pb on a mass basis, exceeded the corresponding limit. Despite UK government guidance on the dangers of leaded paint (Defra, 2005; HSE, 2014; Landlordvisison, 2021) and a plethora of companies in the UK providing in situ paint surveys, there is very little quantitative data for household paints that are publicly available or reported in the scientific literature. For example, Sturges and Harrison (1985) give Pb concentrations (and up to 30,000 mg kg^{-1}) in four paints retrieved house dusts from the centre of Leeds and Horner (1994) provides Pb concentrations (and up to 23,000 mg kg^{-1}) for six samples from a London nursery constructed in the 1920s. More widely, results are reported for XRF studies of households, and mainly in France and the US, but are often restricted to the numbers or percentages of positive cases or levels exceeding 1 mg cm^{-2} or 5000 mg kg^{-1} (USEPA, 1995; Gasana and Chamorro, 2002; Lucas et al., 2012; Sefchick et al., 2021). However, Lanphear et al. (1998) indicate a geometric mean value of 1.73 mg Pb cm^{-2} for interior painted surfaces of 183 urban houses in the US, with maximum values in individual properties ranging from 0.5 to 58 mg cm^{-2} , and Balasubramanian et al. (2011) report a geometric mean value of 8.17 mg Pb cm^{-2} for interior paints of eleven pre-1950 US houses. By comparison, the geometric mean for all data shown in Fig. 2 is 2.66 mg cm^{-2} ($n = 48$) and when exterior surfaces are excluded is 2.98 mg cm^{-2} ($n = 43$).

Regarding the depth distribution of leaded paint, results returned from the analysis of the reference paints, in which Pb was uniformly distributed throughout a single layer, suggest that depth indices of between about 1 and 1.2 are indicative of Pb exposed at the surface.

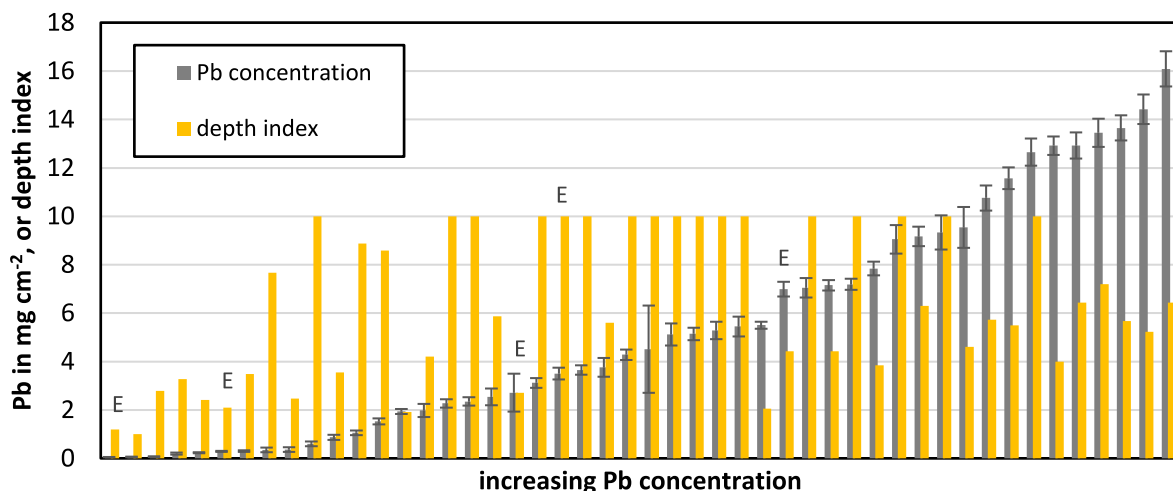


Fig. 1. Concentrations of Pb on painted surfaces in the different properties on an area basis ($n = 48$), shown in ascending order and with 2σ counting errors, and associated measures of depth indices. E denotes exterior surfaces.

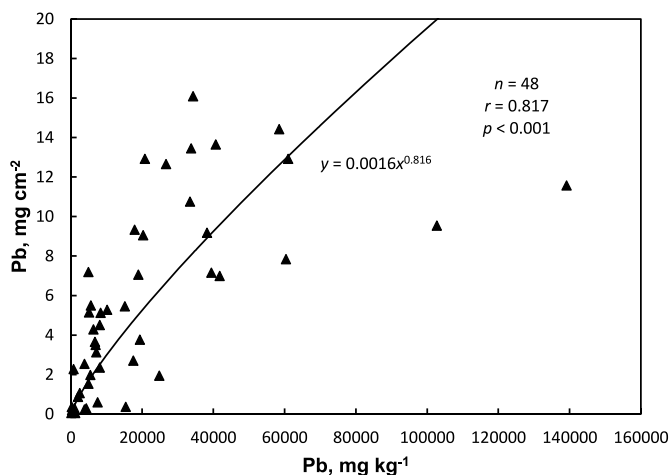


Fig. 2. Concentrations of Pb on an area basis versus concentrations of Pb on a mass basis for the painted surfaces measured in the lead paint and plastics modes of the XRF spectrometer, respectively. Also shown is the Pearson's moment correlation coefficient and the best-fit power line defining the relationship.

Table 4
Concentrations of Pb in dust and lint (in mg kg^{-1}). Errors represent the standard deviation about the mean of six different measurements.

Property	Dust	Lint
1	106.0 ± 10.9	15.0 ± 1.0
2	19.0 ± 4.2	<9.7
3	92.3 ± 5.2	<10.3
4	140 ± 34.2	<8.6
5	84.8 ± 41.8	
6	30.2 ± 6.2	
7	26.9 ± 6.0	
8	4520 ± 2220	<11.0
9	21.1 ± 5.9	13.7 ± 1.6
10	92.5 ± 30.4	
11	82.2 ± 15.6	11.5 ± 1.0
12	96.6 ± 58.7	
13	137.0 ± 54.0	
14	105.0 ± 49.5	<8.8

Amongst the painted surfaces in this study, only two returned a depth index within this range (one interior and one exterior) and both had Pb concentrations below 0.06 mg cm^{-2} and 160 mg kg^{-1} . That is, any paint at the surface does not qualify as being lead-based according to the criteria stated above. Presumably, the variable depth indices for the indoor surfaces measured in the study reflect different (but unknown) decoration histories that appear to involve over-painting.

The figures in this study suggest that, historically, significant quantities of leaded paint have been employed in households, and especially older properties that precede any guidance or legislation on Pb in paint. In most cases, Pb in interior paints was detected on original features, like picture rails, doors and door frames and skirting boards, but was absent on surfaces that appeared to be newer or had been modified (e.g., replastered) or that were part of building extensions and replacement windows and doors. In older properties where Pb was absent from original features (#3 and #6), however, it is surmised that older paints had, at some stage, been removed and surfaces repainted with more modern formulations.

The median concentration of (mean) Pb in dust in the present study (92.4 mg kg^{-1}) is close to the median value reported for various Plymouth household dusts that had been sieved through $63 \mu\text{m}$ and digested in acid before analysis by ICP (87.5 mg kg^{-1} ; Turner and Ip, 2007). The maximum concentrations reported in the two studies are, however, very different (4520 mg kg^{-1} and 195 mg kg^{-1} , respectively). The concentrations of mean Pb in dust in the present study were not correlated with the median concentration of Pb in indoor paint, nor with this measure normalised for frequency of Pb detection. However, likely sources of Pb in dust were inferred from relationships with other elements in the samples. Specifically, mean or individual dust Pb concentrations were not related to the corresponding concentrations of dust Fe (median = 598 mg kg^{-1} ; min = 116 mg kg^{-1} ; max = 2510 mg kg^{-1}) as a measure of external contamination (from road dust and soil, for example; Yoshinga et al., 2014). However, mean and individual Pb dust concentrations exhibited significant correlations with corresponding measures of barium (Ba) in dust (Fig. 3). Barium and Pb were not correlated in the painted surfaces but the former was a major constituent of most paints, presumably as the inert filler, BaSO_4 (Gysau, 2017), and the Pb–Ba mass concentration ratio in the paint that returned the highest Pb concentration (0.67) was close to the slope defining the best fit line for the dust samples (0.48) and as annotated on Fig. 3.

The latter observation, coupled with the lack of soils in the immediate vicinity of most properties and spatial variability of Pb within

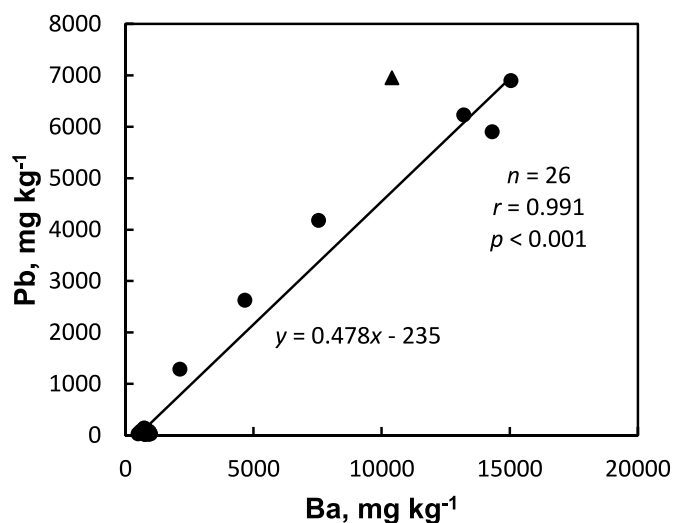


Fig. 3. Concentrations of Pb versus concentrations of Ba arising from individual dust measurements (filled circles) and the Pearson's moment correlation coefficient and best-fit line defining the relationship. Also shown is the concentration of Pb and Ba in the painted surface returning the highest Pb concentration (filled triangle).

individual dust samples, suggests that heterogeneously dispersed leaded paint particles are the principal forms of Pb in the household dusts. While both internal and exterior paints are possible sources in dust, we noted that Cr was often present with Pb in paints on exterior railings and doors, presumably as the pigment, PbCrO_4 (Turner et al., 2016), but was never detected in either indoor paints ($<100 \text{ mg kg}^{-1}$) or indoor dusts ($<20 \text{ mg kg}^{-1}$). This points to an indoor paint source of Pb contamination, with the depth indices returned by the XRF indicating that interior leaded-paints have been over-coated with variable quantities or layers of more modern, unleaded formulations. Where surfaces are visibly damaged, migration of leaded-paint particles is possible, although the painted surfaces in the poorest condition did not necessarily result in the highest dust loading of Pb. It is also possible that surfaces that appear to be in good condition have microscopic damage that allows the migration of leaded-paint particles (Balasubramanian et al., 2011). Alternatively, Pb in dust samples may reflect the historical and improper or unsafe disturbance of older paints during maintenance or renovation (Gooch, 1993). Specifically, the property returning the highest Pb concentrations in dust had undergone extensive renovation as part of the conversion from a public house in 2010.

Despite the range in detection frequency of leaded paint amongst the different properties and the variations in concentrations of Pb in hoovered dust samples, the Pb content of lint samples, as a complementary measure of household Pb contamination, was always close to or below the detection limit of about 10 mg kg^{-1} . The levels reported here are comparable with those given by Mahaffy et al. (1998) for households in Edmonton (Alberta) that were unaffected by occupational exposure to Pb (geometric mean = 20.4 mg kg^{-1}) and by Turner (2019) for households over a wider area of southwest England (median = 15.1 mg kg^{-1}). This suggests a more general and uniform contamination of lint by Pb and that this material by itself may not be a suitable proxy for interior exposure to the metal.

Although this is a relatively small study, it appears to be the most comprehensive, contemporary investigation of Pb in paints in the UK household. Moreover, given the common characteristics of leaded paints, interior designs and renovation and decorating practices, it would be reasonable to assume that the findings of the present study are more generally applicable for older, urban properties in the UK and for properties that are most commonly encountered in the private renting sector (Landlordvision, 2021). Thus, overall, and where original interior structures have been decorated with lead-based paint, overpainting with

more modern, lead-free formulations appears to have been the preferred option; provided that surface coatings are in reasonably good condition, concentrations in dust on a weight-weight basis are generally at the low end of values reported in previous UK studies (Davies and Thornton, 1987; Hunt et al., 1993; Turner and Simmonds, 2006) and there is little immediate risk. However, one dust sample revealed that Pb concentrations can be significantly higher, presumably if renovation, either inside or outside, had not been practiced safely or appropriately (Jacobs et al., 2003).

Current, official UK Government guidance for tenants and landlords (UK Government, 2020) provides a list of hazards that includes Pb in paint, but detail is referred back to existing, older documentation (Defra, 2005). Thus, if landlords or tenants are planning on work that involves removal or disturbance of paint, they should be aware of the original guidance and conduct practices accordingly. This includes removal or protection of local surfaces (e.g. carpets) and furnishings, sanding with on-tool extraction, wet-sanding, chemical stripping or heat stripping below $450 \text{ }^\circ\text{C}$ to remove paint, and use of waterproof abrasive paper for keying intact paint surfaces before over-coating.

5. Conclusions

This study indicates that leaded paints are abundant and pervasive in UK rented properties constructed before restrictions on the use of Pb were introduced. In most cases, leaded paints historically applied to interior surfaces have been over-coated with safer formulations and pose little risk, provided that surface layers are intact. However, inappropriate practices during renovation can result in significant contamination of household dusts that act as a vehicle for Pb exposure. Landlords and tenants in the UK should be made fully aware of government guidance on the safe decoration, maintenance or remodelling of older properties in order to minimise this exposure.

Credit statement

AT: conceptualization; methodology; investigation; formal analysis; writing – original draft; writing – review and editing; project management, ST: conceptualization; methodology; formal analysis; writing – review and editing; AS: conceptualization; project management.

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.

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