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3 **Lead in exterior paints from the urban and suburban**4 **environs of Plymouth, south west England**

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21

22 **Abstract**

23 The dry weight concentrations of lead in paints on a variety of structures in the urban
24 and suburban environs of a British city (Plymouth, south west England) have been
25 determined in situ and ex situ by field-portable x-ray fluorescence spectrometry. Lead
26 was detected in 221 out of 272 analyses, with overall median and mean concentrations
27 of $4180 \mu\text{g g}^{-1}$ and $29,300 \mu\text{g g}^{-1}$, respectively, and a maximum concentration of
28 $390,000 \mu\text{g g}^{-1}$. The highest concentrations were observed in extant paints on poorly
29 maintained, metallic structures, including railings, gates, telephone kiosks and
30 bridges, in various yellow road line paints, and in paints of varying condition on
31 public playground facilities (ramps, climbing frames, monkey bars). Occupants of
32 households in the vicinity of structures that are shedding leaded paint are at potential
33 risk of exposure from paint particles being tracked in on shoes while children in
34 contact with leaded paints in playgrounds and recreational areas are at potential risk
35 from the direct ingestion of paint flakes. Since the issues highlighted in the present
36 study are neither likely to be restricted to this city, nor to the UK, a greater, general
37 awareness and understanding of the sources and routes of exposure of exterior leaded
38 paint is called for.

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40 **Keywords:** leaded paint; chromium; urban; bridges; telephone kiosks; playgrounds

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43 **1. Introduction**

44 Because of the well-documented toxic effects of lead, and in particular those that
45 impact on the development of young children, the use of leaded products has been
46 restricted or phased out over the past few decades (Mansson et al., 2009; Bierkens et
47 al., 2011). In the domestic setting, a significant vehicle for Pb poisoning has been the
48 inadvertent or deliberate ingestion of household paint particles containing various
49 leaded pigments (Jacobs et al., 2002; Su et al., 2002). Consequently, the Pb content of
50 consumer paints has come under close scrutiny in the scientific literature and by
51 regulators and limits on its concentration in formulations have been progressively
52 revised downwards. In the US, the current limit for Pb in consumer paints has been set
53 at 90 ppm (CPSC, 2011), and although several countries have since adopted similar
54 thresholds, the Global Alliance to Eliminate Lead Paint (GAELP) has a goal of
55 eradicating Pb in paint by 2020 (Kessler, 2014).

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57 An additional source of Pb exposure in the home environment is from contaminated
58 external geosolids, like soils and road dusts, that are airborne or tracked in on shoes
59 and clothing (Alradady et al., 2003; Hunt et al., 2006). External solids are often
60 contaminated with residual Pb associated with its past use in gasoline (Zahran et al.,
61 2013; Datko-Williams et al., 2014), but an additional source that has received less
62 attention is extant leaded paint on exterior structures, and in particular on
63 constructions in poor condition and where paint is readily flaking from the substrate
64 through weathering and deterioration of the binder (Weiss et al., 2006). Flaking paint
65 in the external environment may also pose more direct risks to contractors repairing or
66 repainting structures (Jacobs, 1998), and to children if poorly maintained painted

67 surfaces are encountered within or in the vicinity of recreational areas (Mathee et al.,
68 2009).
69
70 In a previous study, a variety of metals in paint flakes sampled from a number of
71 structures in the centre of Plymouth, a coastal city in south west England, were
72 measured by inductively coupled plasma (ICP) spectrometry following acid digestion
73 (Turner and Sogo, 2012). Despite many leaded pigments having been either banned or
74 voluntarily removed from non-consumer paints in the UK by the 1990s, we
75 discovered concentrations of Pb in many samples that exceeded the US urban
76 abatement action level of 5000 $\mu\text{g g}^{-1}$ (Horner, 2004). This finding has prompted us to
77 hypothesise that leaded paint may be a pervasive problem in this city and in the
78 British urban and suburban environments more generally. Accordingly, the present
79 investigation employs a field portable x-ray fluorescence (FP-XRF) spectrometer,
80 configured in a plastics mode and with a thickness correction algorithm for the
81 analysis of thin layers, to allow a greater throughput of paint analyses on a wider
82 range of structures in Plymouth. Measurements are made both in situ and ex situ and,
83 although we specifically target Pb, we also measure Cr because of its occurrence in
84 many leaded pigments. The results are used to provide an inventory of the types of
85 structures on which leaded paint is likely to be encountered in the city and in the UK,
86 and to address the potential sources of Pb exposure to the general public from paint in
87 the urban and suburban settings.

88

89 **2. Materials and methods**

90 *2.1. Sampling and sample locations*

91 Fifteen urban and suburban regions of Plymouth (population ~ 250, 000) that
92 provided a wide geographical coverage of the city were visited between February and
93 April of 2015 and during periods of dry weather. In each region, as many paints on
94 public structures and facilities and on municipal and commercial buildings (excluding
95 private residences) that were directly accessible from the roadside or pavement were
96 examined using a FP-XRF spectrometer. On site, painted wooden, metallic and
97 tarmacked surfaces, including gates, railings, bridges, roads, buildings, posts,
98 playground facilities, pillar boxes and K6-type telephone kiosks, were photographed,
99 position-fixed and coded, and the colour, condition and degree of layering of paint
100 recorded. Surfaces that were in reasonable condition were measured in situ while
101 those that were visibly flaking were sampled directly or from fragments that had
102 accumulated on the ground using a pair of stainless steel tweezers. Where distinct
103 layers of paint or distinct components or colours were observed on a single structure,
104 multiple measurements were made or multiple samples collected. Samples were
105 stored individually in labelled specimen bags and returned to the laboratory in a
106 sealed polyethylene box.

107

108 2.2. XRF analysis

109 The outer surfaces of the paint fragments and painted structures were analysed for a
110 variety of metals, of which Pb was the focus of the present study but Cr was also
111 considered, by energy dispersive FP-XRF spectrometry using a battery-powered
112 Thermo Scientific Niton XRF analyser (model XL3t 950 He GOLDD+).

113

114 For in situ measurements ($n = 58$), a smooth, regular area of the painted surface was
115 wiped clean and dry using a medical-grade wipe before the XRF nose, including the

116 measurement window and proximity sensor, was positioned firmly against a
117 measurement area of 8 mm in diameter. Surfaces were measured for a period of 200
118 seconds (100 seconds each for the main and low energy ranges) by depressing the
119 trigger mechanism of the instrument. Spectra up to 50 keV were quantified by
120 standardless analysis in ‘plastics’ mode and with a thickness correction of 50 μm to
121 yield metal concentrations in parts per million ($\mu\text{g g}^{-1}$) and with an error of 2σ (95%
122 confidence). Data were subsequently transferred to a laptop computer in the
123 laboratory using Thermo Scientific Niton data transfer (NDT) PC software.

124

125 For the analysis of paint fragments in the laboratory ($n = 224$), the XRF was securely
126 fixed into a bench top accessory unit and connected via USB and a remote trigger to
127 the laptop. Individual samples were carefully placed on to a SpectraCertified® Mylar
128 polyester 3.6 μm film using a pair of tweezers and with the outer face downwards,
129 before the slide was positioned centrally over a 3 mm small-spot collimator above the
130 detector. Measurements were activated through the laptop under conditions identical
131 to those described above and with concurrent transfer of data through the NDT
132 software.

133

134 The Niton XLT3t GOLDD+ series analysers define detection limits that are specific
135 to the characteristics of the sample and the counting time in terms of 3σ ;
136 measurements are reported where concentrations exceed this threshold and detection
137 limits are provided otherwise. Detection limits specific to the paint samples and
138 analytical conditions of the present study were about $8 \mu\text{g g}^{-1}$ and $15 \mu\text{g g}^{-1}$ for Pb and
139 Cr, respectively. No XRF reference paints exist that report Pb concentrations on a
140 w/w basis above $100 \mu\text{g g}^{-1}$. However, analysis of NIST SRM2579a paint films that

141 are defined by a wide range of Pb concentrations on a mg cm^{-2} basis (x) returned
142 results in $\mu\text{g g}^{-1}$ (y) that were directly proportional to certified values ($y = 3.2 \times 10^4 x$; r^2
143 > 0.999 ; $p < 0.01$). For an evaluation of absolute accuracy, a reference plastic
144 manufactured by Niton (PN 180-554, 13 mm thick polyethylene disk impregnated
145 with metals) was analysed in quadruplicate and without thickness correction. The
146 mean measured Pb concentration (\pm one sd) of $944 (\pm 12) \mu\text{g g}^{-1}$ compared with an
147 added Pb concentration of $1002 (\pm 40) \mu\text{g g}^{-1}$, while the mean measured Cr
148 concentration (\pm one sd) of $1090 (\pm 12) \mu\text{g g}^{-1}$ compared with an added concentration
149 of $995 (\pm 40) \mu\text{g g}^{-1}$.

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151 Since Pb and Cr were measured both on painted surfaces in situ and on flakes returned
152 to the laboratory, a comparison between the two approaches was made on a number of
153 surfaces where intact areas were adjacent to visibly flaking areas. Despite possible
154 differences between adjacent areas in terms of thickness of application and degree of
155 corrosion, coupled with any potential confounding effects associated with the
156 underlying substrate itself, good agreement was observed between Pb concentrations
157 measured across a range of surfaces ($[\text{ex situ}] = 1.40 * [\text{in situ}]$; $r = 0.921$, $n = 10$, $p <$
158 0.01); for Cr, a weaker relationship based on fewer analyses was observed that was
159 nevertheless significant ($[\text{ex situ}] = 0.58 * [\text{in situ}]$; $r = 0.630$, $n = 6$, $p = 0.05$). Note
160 that in the cases where painted surfaces were measured by both approaches, only
161 those determined in situ have been reported below.

162

163 **3. Results and Discussion**

164 *3.1. Structure description and classification*

165 A summary of the type, number and colours of the painted structures examined in the
166 present study is given in Table 1. A total of 224 structures were measured and a total
167 of 272 analyses for Pb were performed, with the numerical difference reflecting
168 multiple analyses of different regions or layers of the same structure. The most
169 common categories examined were railings (all metal and including various fences)
170 and posts (mainly metal and encompassing street posts, lamp-posts and traffic posts)
171 which were a variety of colours; likewise, gates (including gate posts) and downpipes
172 were mainly of metal construction and were painted in a number of colours.
173 Playground facilities embraced a variety of brightly coloured installations, such as
174 roundabouts, ramps, climbing frames, monkey bars and the supporting posts of slides
175 and swings, while painted areas of public and commercial buildings of various ages
176 included walls, doors, window frames and window sills. The iconic K6-type cast iron
177 telephone kiosks and metallic pillar boxes of different designs were always red,
178 whereas the painted (road) surfaces of highways, car parks, pavements and manhole
179 covers, were mainly yellow-orange. Bridges were the largest structures examined and
180 consisted of footbridges over roads and railways, road bridges over disused railways
181 and disused rail bridges over roads. Miscellaneous structures not categorised above
182 included fire hydrants, bollards, waste bins and benches.

183

184 In general, and among the structures examined, the surfaces of the telephone kiosks
185 and bridges were observed to be in the poorest condition with what was perceived to
186 be extensive flaking of multiple layers of paint and visible accumulations of paint
187 fragments on the ground. For this reason, measurements of different layers or regions
188 of paint were made on all kiosks and on the majority of bridges. Other constructions

189 on which multiple measurements were performed included several multi-coloured
190 playground facilities, and various buildings, posts and railings.

191

192 *3.2. Lead concentrations and distributions*

193 Table 2 summarises the concentrations of Pb in paints on the different types of
194 structures. Overall, Pb was detected in 221 cases (or 81% of all analyses), with
195 concentrations spanning more than four orders magnitude (ranging from 20 $\mu\text{g g}^{-1}$ to
196 about 390,000 $\mu\text{g g}^{-1}$) and exceeding the US urban abatement action level of 5000 $\mu\text{g g}^{-1}$
197 in 104 instances (or 38% of all analyses). The median, grand mean and standard
198 error for all measurements were 4,180 $\mu\text{g g}^{-1}$, 29,300 $\mu\text{g g}^{-1}$ and 3,560 $\mu\text{g g}^{-1}$,
199 respectively, but where median concentrations were used to summarise multiple
200 measurements of the same structure (and $n = 175$), respective values were 2,700 $\mu\text{g g}^{-1}$,
201 20,500 $\mu\text{g g}^{-1}$ and 2,770 $\mu\text{g g}^{-1}$. Skewness and excess kurtosis for the entire dataset
202 were +3.03 and 12.0, respectively (or +2.52 and 6.70, respectively, for data based on
203 the number of structures measured), indicating a highly skewed, leptokurtic data set
204 whose variability is due to a relatively small number of extreme differences.

205

206 For a given type of structure and for all measurements made on such, concentrations
207 of Pb were highly variable and in most cases distributions were positively skewed,
208 with median concentrations of a few thousand $\mu\text{g g}^{-1}$ for most constructions but an
209 order of magnitude greater for bridges and telephone kiosks. Individual concentrations
210 of Pb greater than 100,000 $\mu\text{g g}^{-1}$ were determined in 21 cases. Namely, the lowest
211 painted layers of a number of telephone kiosks (that may have been base paint
212 primers); paints of various colours on the parapets of two road bridges over a disused
213 railway and a disused railway bridge over a road; two yellow-orange ‘no parking’

214 lines; a green downpipe on the exterior wall of a small industrial estate; the yellow
215 railing of a ramp in a playground; the lowest painted layer of a red pillar box; two
216 yellow car park posts; and black and green flakes sampled from cast iron railings
217 surrounding a hospital and cemetery, respectively

218 Table 1: Classification and colour coding of the structures examined in the study.

	black	blue	brown	green	grey/silver	red/pink	white	yellow/orange	multiple	no. structures	no. analyses
bridges				1					5	6	20
buildings	1	1		2	2		1	2	2	11	16
downpipes	1	2	1	4	1		2			11	11
gates	3		2	4		6	1	4	3	23	27
pillar boxes						14				14	16
playgrounds		3		5		7	5	6		26	26
posts	9			1	12		5	6	2	35	38
railings	12	5	1	16	7	5	2		2	50	52
roads				1		1	2	27		31	31
telephone kiosks						9				9	26
miscellaneous	1			2	2		1	1	1	8	9
total	27	11	4	36	24	42	19	46	15	224	272

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224 Table 2: Distribution and summary statistics for Pb concentrations ($\mu\text{g g}^{-1}$) among the different structures measured.

	no. undetected	$< 10^2$	10^2-10^3	10^3-10^4	10^4-10^5	$>10^5$	min.	max.	median
bridges	2	1	5	1	6	5	94	223,000	30,100
buildings	2	3	6	3	2		33	9,850	573
downpipes				7	3	1	1,810	104,000	4,480
gates	6		7	9	5		166	28,800	1,650
pillar boxes	1	1	4	5	4	1	42	171,000	2,820
playgrounds	6		10	3	6	1	116	115,000	1,170
posts	13	3	7	9	4	2	20	175,000	2,200
railings	7	6	13	9	12	5	32	198,000	4,280
roads	12	2		8	7	2	50	142,000	5,020
telephone kiosks			1	1	20	4	510	389,000	43,200
miscellaneous	2	3	1		3		74	97,200	982
total	51	19	54	55	72	21	20	389,000	4,180

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229 Table 3 summarises the Pb results according to colour classification. (Note that
230 samples and surfaces from the multi-coloured structures listed in Table 1 have now
231 been divided into their component colours.) Lead was measured in all colour
232 categories, and the percentage of cases in which Pb was detectable ranged from less
233 than 65 for grey/silver and white surfaces to more than 90 for blue and red/pink
234 surfaces. Median concentrations ranged from about $400 \mu\text{g g}^{-1}$ for grey/silver paints to
235 more than $25,000 \mu\text{g g}^{-1}$ for red paints, and with the exception of blue paints, colour
236 categories were not normally distributed with positive skewness and positive excess
237 kurtosis.

238

239 *3.3. Leaded pigments and the occurrence of Cr*

240 The wide dispersion of Pb concentrations among the different colours and ages of
241 structures suggests that a number of leaded compounds are encountered in the paints
242 analysed. Before being phased out or restricted in use, leaded pigments were used in
243 paints for colour, opacity and corrosion inhibition, and, in combination with other
244 metal compounds, as driers (Abel, 2000). Some commonly employed leaded pigments
245 include the primers, red lead, $2\text{PbO}\cdot\text{PbO}_2$, and basic lead silicochromate,
246 $\text{PbSiO}_3\cdot 3\text{PbO}\cdot\text{PbCrO}_4\cdot\text{PbO}$, white lead, $2\text{PbCO}_3\cdot\text{Pb}(\text{OH})_2$, a pigment that turns black
247 on exposure to sulphurous gases in the atmosphere, lead chromate, a series of yellow
248 and orange pigments that can be pure PbCrO_4 or mixed phase $\text{PbCrO}_4/\text{PbSO}_4$,
249 molybdate red, $\text{Pb}(\text{Cr,S,Mo})\text{O}_4$, and chrome green, $\text{PbCrO}_4/\text{PbSO}_4\cdot\text{FeNH}_4\text{Fe}(\text{CN})_6$, a
250 co-precipitate of lead chromate and Prussian blue.

251

252 It is clear that many of these Pb-based pigments also contain hexavalent Cr, itself a
253 cytotoxin and genotoxin (Wise et al., 2006). Chromium was detected in 106 paints,

254 with median, minimum and maximum concentrations of $443 \mu\text{g g}^{-1}$, $24 \mu\text{g g}^{-1}$ and
255 $33,800 \mu\text{g g}^{-1}$, respectively, and a distribution that displayed positive skewness and
256 excess kurtosis. Chromium was detected in 99 cases in which Pb was detected and
257 these results are plotted as a scatter chart in Figure 1. Here, data have been coded
258 according to colour, although no clear trends or groupings are evident on this basis.
259 Also shown in the figure are ratios of Pb to Cr (on a w/w basis) for the pigments with
260 the lowest and highest ratios of these metals (namely, about 4 for pure PbCrO_4 to
261 about 24 for basic lead silicochromate). Many of the data lie within these two end-
262 points, consistent with the use of a variety of Pb-Cr pigments in the paints. Data with
263 ratios of Pb:Cr exceeding 24, including those containing significant quantities of Pb
264 and no detectable Cr (e.g. many bridge paints; data not shown), suggest the use of
265 leaded pigments that do not contain Cr, like white lead and the primer, red lead, while
266 data with ratios of Pb:Cr below 4 suggest the existence of additional, unleaded but
267 Cr(III)-bearing pigments (e.g. chromium oxide, Cr_2O_3 , and cobalt blue,
268 $\text{Co}(\text{Al,Cr})_2\text{O}_4$). Relatively low ratios of Pb:Cr and the general dispersion among the
269 data may also be attributed to the heterogeneous but preferential weathering and
270 dissolution of Pb over Cr from PbCrO_4 -based pigments (White et al., 2014).
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272 Figure 1: Concentrations of Pb versus concentrations of Cr in the different coloured paint samples. The
273 lines represent the Pb to Cr mass ratios in lead chromate (4:1) and basic lead silicochromate (24:1).

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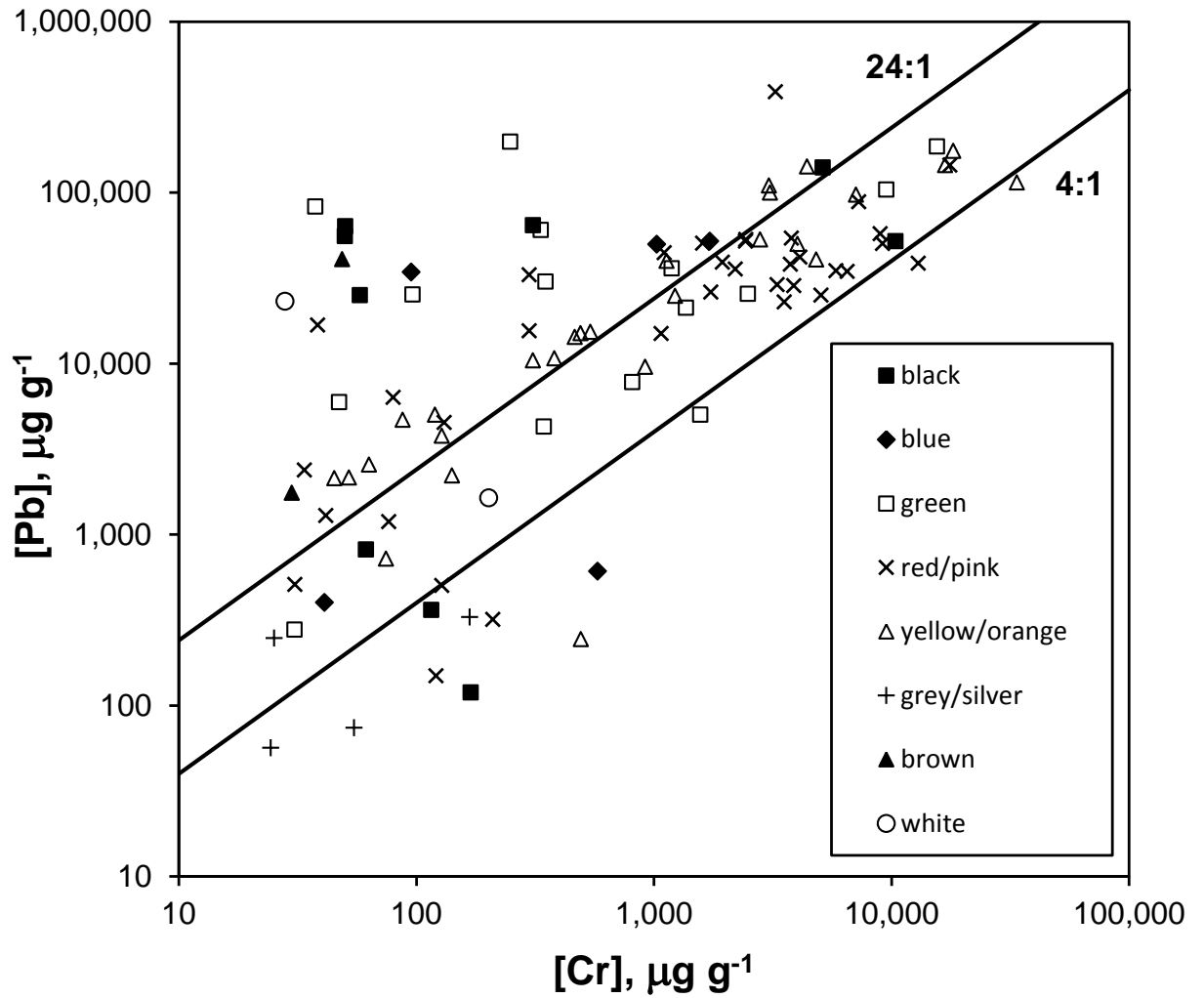


Table 3: Distribution and summary statistics for Pb concentrations ($\mu\text{g g}^{-1}$) among the different colours of paint measured.

	no. undetected	$< 10^2$	10^2-10^3	10^3-10^4	10^4-10^5	$>10^5$	min.	max.	median
black	4		16	5	8	2	119	158,000	982
blue	1	2	4	3	6		32	66,100	5,860
brown	3			2	1	1	1,750	184,000	21,600
green	9	5	6	13	9	3	54	198,000	2,500
grey/silver	9	6	6	3	4		20	18,600	398
red/pink	5	1	11	11	32	7	42	389,000	25,600
white	7	3	3	5	3	1	30	202,000	2,780
yellow/orange	13	2	8	12	12	5	50	175,000	4,680

3.4. Implications for exposure and the local environment

This study has revealed a wide range of Pb concentrations in painted surfaces of a variety of both contemporary and historical structures in the urban and suburban environs of a British city. The highest concentrations (in excess of 100,000 $\mu\text{g g}^{-1}$) were often associated with relatively old, metallic (cast iron and steel) structures that were observed to be in a poor state of repair and that often had distinct layers of flaking paint resulting from aging, weathering and abrasion of the surface. In these cases, Pb concentrations were usually highest in the lowest, and presumably oldest, layers, an effect exemplified by the distribution of Pb concentrations among the different coloured or shaded surfaces of a road bridge in Figure S1 and a telephone kiosk in Figure S2. This effect may be attributed to the reduction in the Pb content of progressively newer paints (including the use of leaded primers as the oldest basecoat), or to an increase in the degree of weathering of non-leaded components of the paint with increasing age. A more surprising observation, however, was the high incidence of concentrations that exceeded the urban abatement action level of 5000 $\mu\text{g g}^{-1}$ (Horner, 2004) on painted surfaces that were largely intact and that appeared to have been applied relatively recently. Examples of the latter included a range of facilities in children's playgrounds (one installation was date-stamped as March 2009), posts and bollards used to demark the entrances of car parks, several small, sprung entrance gates to recreational facilities, and bright yellow paint on the footway of a pedestrian bridge.

From an environmental standpoint, flaking of leaded paint will result in both the direct (particulate) and indirect (through dissolution-readsorption) contamination of local soils, road-dusts and pavement dusts. Contamination of surface waters may

occur indirectly through the gradual dissolution of Pb from paint flakes into rainwater, an effect accentuated by low pH and the presence of natural and anthropogenic complexants, and the subsequent transport of aqueous Pb by storm-water drains (Davis and Burns, 1999). Human exposure associated with flaking leaded paint may be a concern for residents and schools in the immediate vicinity of structures in a poor state of repair (Weiss et al., 2006). In the present study, private houses were observed within 20 m of many of the structures in the poorest condition and whose painted surfaces were associated with the highest concentrations of Pb; we also noted that one of the railings shedding paint and containing Pb at a concentration in excess of 25,000 $\mu\text{g g}^{-1}$ served to enclose a school play area. In these cases, Pb may enter the indoor environment as fine, airborne dust and through paint flakes and contaminated soil tracked indoors on shoes and clothing. Track-in and soil resuspension appear to be the main vehicles for the introduction of external Pb into the contemporary household (Laidlaw et al., 2014) and flaking paint from external structures (and in particular, metallic railings and elevated steel structures like bridges) has been identified as a major contributor to these routes (Weiss et al., 2006; Lucas et al., 2014). The potential for Pb to be tracked into the household is also a concern for families of contractors involved in the repair, maintenance or restoration of structures containing extant leaded paint (Virji et al., 2009) and for members of the public who use telephone kiosks that are in a poor state of repair and that have significant accumulations of paint flakes on the floor.

Perhaps the greatest concerns arising from our research are the wide occurrence and high concentrations of Pb in paints on public playground facilities. Here, the risks are greatest for peeling or chalking paint on structures that children are in direct contact

with, like poles, rails, climbing frames and monkey bars, and to infants under 72 months old because of their tendency to mouth foreign objects and the relatively high absorption of Pb in their gastrointestinal tract (Kennedy et al., 2014). Lead poisoning in a 5-year old child resulting from the biting and ingestion of paint chips from playground equipment in Montreal was described by Health Canada (1994), with the recommendation that painted surfaces be carefully stripped using solvent and repainted using lead-free paint. Leaded paint has since been reported on equipment in contemporary public play-parks in Japan (Takaoka et al., 2006) and South Africa (Mathee et al., 2009) but the present study appears to be the first to document its occurrence in Europe.

From the discussion above, it is clear that the general findings of the present study are unlikely to be specific to the city of Plymouth or to the UK. Thus, since the phasing out of leaded gasoline, exterior paint, in itself or as a component of contaminated soil dust, may represent one of the most significant routes of Pb exposure in the urban and suburban settings. Despite guidance and regulations on the use and removal of paint containing Pb being incorporated into governmental policy (e.g. Health and Safety Executive, 2002), a greater awareness of the pervasiveness of exterior leaded paint is required, both for the public and for employers dealing with the renovation, repair, maintenance or decoration of a wide variety of painted facilities. In particular, means of reducing the inherent risks and managing and regulating Pb in paint on public structures and facilities need to be more clearly and robustly addressed by the relevant authorities and stakeholders.

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