



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



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HIGHLIGHTS

- Because of toxic effects, the use of leaded products has been restricted or phased out.
- Here, exterior paints in a British city were analysed for Pb (and Cr) by FP-XRF.
- Pb was detected in 221 out of 272 analyses with a median concentration of 4180 $\mu\text{g g}^{-1}$.
- Pb concentrations were highest in extant paints on poorly maintained structures.
- Leaded paints used on public playground equipment are of greatest concern.

GRAPHICAL ABSTRACT



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ABSTRACT

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

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1. Introduction

Because of the well-documented toxic effects of lead, and in particular those that impact development of young children, the use of leaded products has been restricted or phased out over the past few decades

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(Mansson et al., 2009; Bierkens et al., 2011). In the domestic setting, a significant vehicle for Pb poisoning has been the inadvertent or deliberate ingestion of household paint particles containing various leaded pigments (Jacobs et al., 2002; Su et al., 2002). Consequently, the Pb content of consumer paints has come under close scrutiny in the scientific literature and by regulators, and limits on concentration in formulations have been progressively revised downwards. In the US, the current limit for Pb in consumer paints has been set at 90 ppm (CPSC, 2011), and although several countries have since adopted similar thresholds, the Global Alliance to Eliminate Lead Paint (GAELP) has a goal of eradicating Pb in paint by 2020 (Kessler, 2014).

An additional source of Pb exposure in the home environment is from contaminated external geosolids, like soils and road dust that are airborne, or tracked in on shoes and clothing (Alradady et al., 1994; Hunt et al., 2006). External solids are often contaminated with residual Pb associated with past use in gasoline (Zahran et al., 2013; Datko-Williams et al., 2014) but an additional source that has received less attention is extant leaded paint on exterior structures, and in particular on constructions in poor condition and where paint is readily flaking from the substrate through weathering and deterioration of the binder (Weiss et al., 2006). Flaking paint in the external environment may also pose more direct risks to contractors repairing or repainting structures (Jacobs, 1998) and to children if poorly maintained painted surfaces are encountered within or in the vicinity of recreational areas (Mathee et al., 2009).

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

2. Materials and methods

2.1. Sampling and sample locations

Fifteen urban and suburban regions of Plymouth (population ~ 250,000) that provided a wide geographical coverage of the city were visited between February and April of 2015 during periods of dry weather. In each region, as many paints on public structures and facilities and on municipal and commercial buildings (excluding private residences) that were directly accessible from the roadside or pavement were examined using a FP-XRF spectrometer. On site, painted wooden, metallic and tarmacked surfaces, including gates, railings, bridges, roads, buildings, posts, playground facilities, pillar boxes and K6-type telephone kiosks, were photographed, position-fixed and coded, and the colour, condition and degree of layering of paint recorded. Surfaces that were in reasonable condition were measured in situ while those that were visibly flaking were sampled directly or from fragments that had accumulated on the ground using a pair of stainless steel tweezers. Where distinct layers of paint or distinct components or colours were

observed on a single structure, multiple measurements were made or multiple samples collected. Samples were stored individually in labelled specimen bags and returned to the laboratory in an airtight polyethylene box.

2.2. XRF analysis

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

For in situ measurements ($n = 58$), a smooth, regular area of the painted surface was wiped clean and dry using a medical-grade wipe before the XRF nose, including the measurement window and proximity sensor, was positioned firmly against an area of 8 mm in diameter. Surfaces were measured for a period of 200 s (100 s each for the main and low energy ranges) by depressing the trigger mechanism of the instrument. Spectra up to 50 keV were quantified by standardless analysis in 'plastics' mode and with a thickness correction of $50 \mu\text{m}$ to yield metal concentrations in parts per million ($\mu\text{g g}^{-1}$) and with an error of 2σ (95% confidence). Data were subsequently transferred to a laptop computer in the laboratory using Thermo Scientific Niton data transfer (NDT) PC software.

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

The Niton XLT3t GOLDD+ series analysers define detection limits that are specific to the characteristics of the sample and the counting time in terms of 3σ ; measurements are reported where concentrations exceed this threshold and detection limits are provided otherwise. Detection limits specific to the paint samples and analytical conditions of the present study were about $8 \mu\text{g g}^{-1}$ and $15 \mu\text{g g}^{-1}$ for Pb and Cr, respectively. No XRF reference paints exist that report Pb concentrations on a w/w basis above $100 \mu\text{g g}^{-1}$. However, analysis of NIST SRM2579a paint films that are defined by a wide range of Pb concentrations on a mg cm^{-2} basis (x) returned results in $\mu\text{g g}^{-1}$ (y) that were directly proportional to certified values ($y = 3.2 \times 10^4 x$; $r^2 > 0.999$; $p < 0.01$). For an evaluation of absolute accuracy, a reference plastic manufactured by Niton (PN 180-554, 13 mm thick polyethylene disk impregnated with metals) was analysed in quadruplicate and without thickness correction. The mean measured Pb concentration (\pm one sd) of $944 (\pm 12) \mu\text{g g}^{-1}$ compared with an added Pb concentration of $1002 (\pm 40) \mu\text{g g}^{-1}$, while the mean measured Cr concentration (\pm one sd) of $1090 (\pm 12) \mu\text{g g}^{-1}$ compared with an added concentration of $995 (\pm 40) \mu\text{g g}^{-1}$.

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

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