



Lead and other toxic metals in playground paints from South West England

Andrew Turner ^{*}, Emily R. Kearl, Kevin R. Solman

School of Geography, Earth and Environmental Sciences, Plymouth University, Drake Circus, Plymouth PL4 8AA, UK



HIGHLIGHTS

- Metals in paints on playground structures have been measured by FP-XRF.
- Lead was detected at dry wt concentrations above 10% in many cases.
- Association of Pb with Cr suggests a prevalence of lead chromates.
- Oral bioaccessibilities of Pb in paint range from ~0.1% to 16.6%.
- High concentrations of Pb, Cr, Cd and Sb in playground paints are cause for concern.

GRAPHICAL ABSTRACT



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ABSTRACT

Paints on surfaces of public playground structures in South West England have been analysed for Pb, Cr, Cd and Sb by field-portable, energy-dispersive XRF. Lead was detected ($>8 \mu\text{g g}^{-1}$) in 102 out of 242 cases, with concentrations ranging from 10 to $152,000 \mu\text{g g}^{-1}$ (median = $451 \mu\text{g g}^{-1}$). Chromium was detected ($>25 \mu\text{g g}^{-1}$) in 48 cases, and concentrations ranged from 26 to $24,800 \mu\text{g g}^{-1}$ (median = $1040 \mu\text{g g}^{-1}$) and exhibited a significant positive correlation with Pb concentrations. Antimony concentrations ranged from 273 to $16,000 \mu\text{g g}^{-1}$ (median = $2180 \mu\text{g g}^{-1}$) in 56 detectable cases, and Cd was detected in eight paints and up to a concentration of $771 \mu\text{g g}^{-1}$ (median = $252 \mu\text{g g}^{-1}$). The highest concentrations of Pb, Cr and Sb generally occurred in yellow or red paints but were encountered on a variety of structures and equipment (e.g. gates, flooring lines, railings and handles of climbing frames and seesaws, and the interior of a model train) and were observed in both flaking, extant paint and in formulations that appeared to have been recently applied. Maximum bioaccessible concentrations of Pb, Cr and Sb in a range of paints, evaluated in selected samples by ICP analysis following pepsin-dilute HCl extraction, were 2710, 205 and $23.6 \mu\text{g g}^{-1}$, respectively, or 16.6, 0.82 and 0.56% of the respective total concentrations. Total and bioaccessible concentrations of toxic metals in playground paints that exceed various contemporary and historical standards (and in many cases for Pb, by orders of magnitude) are likely to be a more widespread and pervasive issue that needs addressing by the relevant authorities.

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1. Introduction

Many non-essential metals and metalloids (hereafter referred to as toxic metals) are of concern because of their adverse effects on human

^{*} Corresponding author.

E-mail address: aturner@plymouth.ac.uk (A. Turner).

health coupled with increasing anthropogenic emissions to and dispersion throughout the environment. The toxicity of a metal may be effected by mimicking an essential element or through interference with some metabolic process, and is largely dependent upon concentration (or, strictly, its soluble or accessible concentration). However, chronic exposure to relatively low levels of many metals can also result in bioaccumulation and a variety of diseases, disorders, impairments and organ malfunctions (Ayres and Hellier, 1998).

One important route of exposure of metals and cause of intoxication among humans, and in particular young children, is the inhalation and ingestion of paint dusts (Mielke et al., 2001; Su et al., 2002; Turner and Sogo, 2012). Toxic metals and their compounds have had (and in many cases continue to have) a variety of uses in many paints as, for example, drying agents, preservatives, fire retardants, corrosion inhibitors and pigments for colour or opacity (Abel, 2000). While undisturbed and intact, coatings and their chemical components are relatively safe. However, once the film begins to deteriorate through abrasion or as the binder and pigments degrade via exposure to UV light and moisture, the paint begins to crack, flake and chalk and metal-bearing particulates are mobilised into the environment. The principal metals of concern in this respect, and that have undergone restriction in many paints (including consumer formulations) over the past few decades, are lead, chromium, arsenic, cadmium, mercury and antimony. The former in particular has been used extensively in paints and is often combined with hexavalent Cr, itself a genotoxic carcinogen, as a series of lead chromate pigments. Lead concentrations in the dry films of various paints have often been reported to be greater than 10% (Gottesfeld et al., 2014; Turner et al., 2014) and in some cases exceed 30% (Martinez, 2000; Mielke and Gonzales, 2008).

The effects of Pb on human health, including those that impact on the neurological development of children, are well-documented with regard to paint exposure in the urban and domestic settings (Farfel et al., 2005; Schwab et al., 2006; Le Bot et al., 2011; Laidlaw et al., 2014). Very little attention has, however, been paid to the routes and degrees of Pb (and other metal) exposure in children arising from paints in public playgrounds (Takaoka et al., 2006; Mathee et al., 2009). This is, perhaps, surprising, since young children are in regular contact with painted surfaces of playground equipment through climbing, crawling, gripping and swinging. In the present study, therefore, we measure the concentrations of Pb, Cr, Cd and Sb in paints on a wide range of structures and equipment in different public playgrounds in South West England. We use a field-portable X-ray fluorescence (FP-XRF) spectrometer on painted surfaces *in situ* and on paint flake samples returned to the laboratory, with the latter also being subjected to a physiological extraction and independent analysis in order to evaluate the oral bioaccessibilities of the metals.

2. Materials and methods

2.1. Sampling and sample locations

A total of 47 playgrounds within five administrative regions (County or District Councils) in South West England were visited between September 2014 and June 2015. Specifically, 20 playgrounds were visited in the city of Plymouth (population ~250,000), 14 in the South Hams district of Devon, six in the county of Cornwall, five in the Sedgemoor district of Somerset, and two in the New Forest district of Hampshire. In each playground, painted structures (entrance gates, benches, shelters and tarmacked flooring) and play equipment (including swings, slides, roundabouts, ramps, climbing frames, see-saws, rockers and goal posts) were inspected, photographed, colour-coded and categorised. XRF analysis was performed *in situ* or in the laboratory depending on the condition of the painted surface. Thus, where paint was visibly flaking, samples, with one dimension of at least 3 mm, were taken using a pair of stainless steel tweezers and stored individually in zip-lock specimen bags in a polyethylene box and returned to the laboratory. Paint was either taken from

the structure itself, taking care not to disturb the integrity of remaining paint, or from fragments that had accumulated at the base of the installation. Where paint had been newly applied or was not possible to dislodge from its substrate with tweezers, the surface was measured with the XRF on site.

2.2. XRF analysis

The outer surfaces of the paint samples and painted structures were analysed for Pb, Cr, Cd and Sb by energy dispersive FP-XRF using a battery-powered Niton XRF analyser (model XL3t 950 He GOLDD+) in 'plastics' mode. Although the XRF has 'lead-in-paint' and 'thin film' modes that are often used to analyse paints, the mode for plastics is capable of handling thin sections through a sample thickness correction algorithm and has the advantage of providing concentrations of Pb and other elements on a dry weight basis rather than on an areal (mg cm^{-2}) basis.

For *in situ* measurements ($n = 102$), a smooth area of the painted surface was wiped clean and dry using a medical-grade wipe. The XRF nose, including the measurement window and proximity sensor, was then positioned firmly against an area of 8 mm in diameter and the surface analysed for a period of 200 s (100 s each for the main and low energy ranges) by depressing the trigger mechanism. Spectra up to 50 keV were quantified by standardless analysis after applying a thickness correction of 50 μm to yield metal concentrations in the dry film 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 Niton data transfer (NDT) PC software.

For the analysis of paint samples in the laboratory ($n = 140$), the XRF was secured into a bench top accessory stand and connected to a laptop via USB and a remote trigger. Samples were placed on to a SpectraCertified Mylar polyester 3.6 μm 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 XRF 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 detection limits of the XRF are dependent on a number of factors, including the element, mode of application, measurement time, composition and thickness of material, effective diameter of the detector and presence and nature of a substrate. Furl et al. (2012) suggest that a measurement detection limit of the Niton XL3t can be approximated by multiplying the counting error by 1.5, and on this basis, and for the lowest three values of 2σ reported for samples that were detectable, we estimate detection limits for paint samples and painted surfaces (in $\mu\text{g g}^{-1}$) of around 8, 25, 40 and 70 for Pb, Cr, Cd and Sb, respectively. It is worth noting that Hg was not detected in the samples and that while As often returned a result, inspection of the spectra usually revealed significant overlap of its principal peak ($K_{\alpha 1}$ at 10.544 keV) with one of the main Pb peaks ($L_{\alpha 1}$ at 10.551 keV).

As a calibration-accuracy check, two Niton reference polyethylene discs, 31 mm in diameter and 13 mm thick and that had been impregnated with various elements, were analysed in quintuplicate without thickness correction. Thus, measured concentrations (in $\mu\text{g g}^{-1}$) of Pb, Cr and Cd in PN 180-554 (batch SN PE-071-N) were 944 ± 12 , 1090 ± 12 and 139 ± 5 , compared with respective reference values of 1002 ± 40 , 995 ± 40 and 150 ± 6 , while measured concentrations (in $\mu\text{g g}^{-1}$) of Pb, Cr, Cd and Sb in PN 180-619 (LOT#T-18) were 143 ± 6 , 124 ± 3 , 262 ± 7 and 88 ± 11 , compared with respective reference values of 155 ± 12 , 106 ± 10 , 292 ± 20 and 94 ± 10 .

Since metals were measured both on painted surfaces *in situ* and on samples 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 regions in terms of thickness of paint application, exposure of underlying paint layers and degree of corrosion, coupled with any

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