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Antimony in paints and enamels of everyday items

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12 **Abstract**

13 Concentrations of antimony have been determined for paints and enamels that are available to

14 the consumer or accessible to the public by x-ray fluorescence spectrometry. The metalloid was

15 only present in consumer paints of a speciality (e.g. artistic) nature, but was common in old

16 household paints as an anti-chalking agent and in brightly-coloured contemporary exterior

17 paints (on roads, street furniture and playground equipment, for example) as a colour fastener

18 with concentrations ranging from a few hundred to about 25,000 μg g^{-1}. Antimony was also

19 found in contemporary container glass and ceramic products as an additive or opacifier and as

20 a colour fastener in enamels at concentrations up to a few thousand μg g^{-1}. Overall, the yellow

21 pigment, lead antimonite, was only evident in two ceramic products, with Sb concentrations

22 exceeding 62,800 μg g^{-1}. Available data in the literature suggests that, while Sb concentrations

23 up to 30 μg g^{-1} are bioaccessible in exterior paints and that concentrations of up to 20 mg L^{-1}

24 are migratable in some ceramicware, no relevant regulations are currently in place. Given our

25 lack of understanding of the health impacts of Sb, more studies on its toxicity and mobility from

26 commonly encountered products are required.

27

28 **Keywords:** antimony; paints; enamels; ceramics; glass; XRF

1. Introduction

Antimony (Sb) has been used as a pigment, and mainly as antimony sulphide and lead antimonite, for several thousand years (Aldersey-Williams, 2011; Schwarz-Schampera, 2014). Currently, Sb has the status of a technologically-critical element that is essential for economic development, and although the principal present uses are in antimonial lead, as a flame retardant synergist in textiles and plastics and as a catalyst (e.g. in the production of polyethylene terephthalate, PET), the element is still used in pigments, ceramic glaze and glass (Orisakwe, 2012; Dupont et al., 2016). The most recent European data available indicate that Sb employed as an additive in glass manufacture represents 1% (250 t) of total use while application in pigments, paints and ceramics constitutes about 4.5% (1100 t); these values compare with 3.9% in PET production and 32.9% in flame retardant synergist manufacture (European Communities, 2008). In addition to these applications, Sb readily and widely contaminants many contemporary consumer plastics at low levels through the recycling of electronic and PET-based products (Turner and Filella, 2017).

While the behaviour of Sb in different environmental compartments is relatively well-known, or at least has been extensively studied (Filella et al., 2009), the toxicological implications of its pervasiveness in everyday products are far less understood. Specifically, the occurrence and fate of Sb as a catalyst or synergist have received recent attention (Haldimann et al., 2013; Snedeker, 2014; Turner and Filella, 2017), with concentrations migratable from the matrix regulated for (PET) food containers (European Commission, 2005) and children's toys (European Parliament and the Council of the European Union, 2009). However, its occurrence, function, mobility and potential impacts in contemporary paints, consumer glass and ceramicware are poorly documented, despite the obvious exposure routes arising from these applications.

In this study, we examined the occurrence of Sb in the paints and enamels of various consumer goods and everyday items that have been purchased new or that are in circulation and in extant applications on structures which the public are exposed to and where the presence and/or mobility of other elements, like Cd, Cr(VI) and Pb, are highly regulated on environmental and health grounds (Sheets, 1999; Turner, 2019). Measurements of Sb were made *in situ* or in the laboratory by energy dispersive x-ray fluorescence (XRF) spectrometry and relevant information in the literature on its migration from any applications was reviewed. The investigation complements an earlier study that determined the occurrence and distribution of Sb as catalytic residue or a flame retardant synergist in consumer plastics using the same protocols and techniques (Turner and Filella, 2017).

2. Materials and methods

About 400 measurements of Sb have been performed on distinct regions (in terms of colour or texture) of 335 samples acquired from or located within south west England. Many of the samples had been measured as part of independent research projects (Turner et al., 2015; Turner et al., 2016; Turner and Solman, 2016; Turner, 2019a) and here published and unpublished data on Sb have been compiled, while other samples were analysed specifically for the purposes of the present study. The painted and enameled samples are described and categorised below.

(1) "Interior extant paint" on interior walls, doors, frames, skirting boards and floors of various private households in the city of Plymouth ranging in age from 50 to 100 years;

(2) "Exterior extant paint" on the exteriors of these and other private households and including walls, guttering and timber;

(3) "Municipal extant paint" on external wooden, tarmacked and metallic structures and street furniture accessible to the public in the city of Plymouth, including roads, post boxes, phone boxes, benches, gates and playground equipment;

(4) "Extant boat paint" applied to the hull, decking and cabin of pleasure craft or small commercial vessels located on the coastal foreshores in the vicinity of Plymouth and that were either abandoned or in use;

(5) "Consumer paint" for internal and external decoration or for artwork and that had been purchased within the past five years;

(6) "Painted toys/equipment": the paint on wooden and metallic toys and other consumer items (e.g. tools, storage tins, artefacts) around the household;

(7) "Consumer bottle enamel": the decorated enamels on glass for the containment and sale of wine, beer and food products;

(8) "Drinking glass enamel"; the enamels on glassware for drinks and other food-contact items purchased new or secondhand;

(9) "Ceramicware enamel": the enameled decorations and glaze of (mainly food-contact) ceramicware in circulation and purchased new or secondhand or loaned from colleagues.

For each sample, the condition and principal colour (or colours) were noted, along with any distinguishing signage (e.g. place of manufacture). For intact extant paints, and to avoid disturbance or damage to the surface, Sb and other elements that included Br, Cl, Ni, Pb, Ti and Zn, were measured *in situ* and on each distinctive colour and, where possible, the undecorated substrate of structures using a battery-operated Niton XL3t 950 He GOLDD+ portable XRF spectrometer. Here, the 8-mm diameter detector window of the instrument, containing the 200 µA-50 kV x-ray source and large area drift detector, was pointed directly over the area to be measured with the aid of real-time video footage projected on to the touchscreen display through a charged-couple device camera. Counting was activated by the trigger mechanism of the XRF spectrometer for a period of between 30 and 60 seconds in a "plastics" mode, whose performance has been verified by independent measurements of acid digests by inductively

coupled plasma spectrometry (Turner, 2019b). Successive measurements were made at 50 kV-

40 μA and 20 kV-100 μA and employing a thickness correction of between 0.05 and 0.1 mm.

For flakes of paint carefully retrieved from structures, new paints purchased in hardware stores

that had been applied by brush to a series of glass or cardboard slides and air-dried for 24 to 48

h, and new and secondhand ceramic and glass products of less than 200 mm in length, the XRF

was operated under the same conditions but in the laboratory while housed nose-upwards in an

accessory stand and activated remotely via USB. To discriminate Sb present in any décor of

these products from Sb in the glass or ceramic glaze, readings were also taken on undecorated

regions of the substrate, where possible. For products greater than 200 mm in length the surface

to be measured was cradled in a radiation apron on a stainless steel table and the XRF operated

likewise but handheld.

Elemental concentrations (in μg g^{-1}) were derived from secondary (fluorescent) x-ray spectral

peaks using standardless fundamental parameters software. Polyethylene Niton reference discs

impregnated with various elements (including Sb) were analysed at regular intervals throughout

each measurement session and returned concentrations that were within 15% of certified values.

The limit of detection for Sb, defined as three standard counting errors of background intensity

(and derived from errors in samples that did not return a signal for Sb) varied according to the

precise matrix and thickness but was generally in the range 40 to 100 μg g^{-1}. Precision, derived

from quintuplicate measurements of the same location on a range of samples, was always better

than 20%.

3. Results and discussion

3.1. Occurrence, sources and concentrations of Sb

125   Table 1 shows the number of samples considered in each category and the number of cases in

126   which Sb was detected by XRF spectrometry, along with summary statistics of Sb

127   concentrations. Note that where Sb was detected at multiple locations of a sample (for example,

128   on distinctly different colours), the average detectable concentration and principal colour have

129   been recorded. Also shown in Table 1 for each category are the colours of the samples that

130   returned the highest Sb concentrations and the most common colours that returned detectable

131   Sb, as well as the number of cases in which Sb was detected with Pb at a concentration above

132   100 $\mu g\ g^{-1}$.

133   Overall, Sb was detected in at least one region of about a third of all samples tested and detection

134   frequency exceeded 30% for extant paints (with the exception of those on boats), the enamels

135   on drinking glasses and ceramicware products. Detection frequency was lowest on painted toys

136   and household equipment and the enamels of consumer bottles. Antimony was detected in all

137   colours, and including the colourless or lightly coloured glass of some products, but most

138   commonly occurred in articles coloured yellow, red (including pink) or shades of white.

139   Significantly, Pb was present at concentrations above 100 $\mu g\ g^{-1}$ in nearly 90% of samples

140   where Sb was detected and across all sample categories and among all colours.

141   The highest concentrations of Sb (above 50,000 $\mu g\ g^{-1}$) were encountered with concentrations

142   of Pb above 100,000 $\mu g\ g^{-1}$ in two bright yellow ceramic items (a tea cup and plate, with the

143   XRF spectra of the former illustrated in Figure 1a). These are the only clear cases in which the

144   pigment, lead antimonite or Naples yellow (whose theoretical molecular formula is $Pb_2Sb_2O_7$),

145   appears to have been used, and where excess Pb can be attributed to its additional use in the

146   glazing of these products. The pigment may have been used in some older interior and exterior

147   paints (where Sb concentrations exceeded 20,000 $\mu g\ g^{-1}$) but here concentrations of Pb were

148   lower than the required stoichiometry of the compound and results and any relationships with

149   colour may have been confounded by the effects of multiple layers of different paint

150   formulations measured *in situ*.

151   Regarding paints available to the contemporary consumer, Sb was present at concentrations of

152   a few thousand $\mu g\ g^{-1}$ in two artists paints containing the bright red pigment, cadmium

153   sulphoselenide (CI Pigment Red 108), and exemplified by the XRF spectrum in Figure 1b, and

154   a speciality leaded paint coloured by lead chromate molybdate sulphate red (CI Pigment Red

155   104), but was not detected in any paints for internal or external decoration. This suggests that

156   Sb detected in extant paints of private properties arises from its presence in older applications,

157   and a co-association with Pb may simply reflect the historical use of leaded paints in the same

158   era. Antimony trioxide (CI Pigment White 11) was added to anatase titanium dioxide (CI

159   Pigment White 6) to reduce chalking of the primary pigment before inherently antichalking

160   formulations were available (Abel, 2000) and this may explain the presence of Sb in white

161   extant paints in association with high concentrations of Ti (see XRF spectrum in Figure 1c).

162   Antimony trioxide is, however, unlikely to be present (additionally) as a flame suppression

163   synergist because concentrations of Cl and Br (indicators of halogenated flame retardants) were

164   always too low to provide any form of fire retardancy (typically a few percent w/w; National

165   Materials Advisory Board, 1970).

166   In extant municipal paints and boat paints, Sb was usually present at concentrations ranging

167   from 1000 to 10,000 $\mu g\ g^{-1}$ in formulations that were brightly coloured, and in particular in reds

168   and yellows. In most cases, the XRF spectrum suggested that the primary pigment for colour

169   was lead chromate (CI Pigment Yellow 34), lead chromate molybdate sulphate or bismuth

170   vanadate (CI Pigment Yellow 184) (Figure 1d). Clearly, Sb is not used in these cases for colour

171   itself but appears to be used as a fastener where colour protection from UV radiation is required

172   (USAC, 2017).

Among the glass containers and drinking vessels, including newly-purchased articles, Sb was present in the glass itself in three products, presumably as a fining component or decolourising agent (Grund et al., 2010), and appeared to be used as a colour fastener on the brightly coloured enamels of ten products, with maximum Sb concentrations in the glass and enamel of about 4000 μg g^{-1} and 8000 μg g^{-1}, respectively. Multiple measurements of ceramicware in circulation suggested that Sb was present in the glaze and uncoloured enamels of 12 products and at concentrations ranging from about 1000 to 3000 μg g^{-1}, possibly as an opacifier in articles fired at low temperatures (Demont et al., 2012), and in the coloured enamels of 15 products. As above, Sb was present as a pigment (lead antimonite) in two cases but appeared to be more commonly encountered in significantly lower concentrations (1000 to 4500 μg g^{-1}), presumably as a colour fastener for other pigments.

3.2. Migration and potential environmental and health impacts of Sb

Given concerns about the toxicity of Sb (IARC, 1989; NTP, 2018), the lack of regulations that directly apply (or that have been applied) to the sample types considered herein and the data presented in Table 1 is surprising. The most relevant regulation concerns material like paint, that can be scraped off children's toys, where a maximum migratable limit (mobilised by 0.07 M HCl) is 560 μg g^{-1} according to the amended EU Toy Safety Directive (European Parliament and the Council of the European Union, 2009). However, the results reported in Table 1 suggest that the type of paint used on children's toys and other household wooden and metallic artefacts rarely contains detectable Sb. Whether this reflects attempts to avoid Sb in such products or simply results from no requirement for the metalloid in these applications is unclear. Perhaps of greater concern to human health in this perspective is the use of Sb as a fastener in brightly coloured paints used on public playground equipment. The highest concentration of Sb returned for such equipment was about 6000 μg g^{-1}, with an earlier study revealing that up to 0.56 % of Sb in these formulations was accessible to dilute HCl (Turner et al., 2016). This suggests about

198   30 μg g^{-1} of Sb in playground paint could be accessible to children through inadvertent

199   ingestion. While this concentration is well below the limit stipulated by the Toy Safety

200   Directive, it should be noted that, in the majority of cases considered here, Sb was co-associated

201   with high concentrations of lead and that any synergistic effects of these elements on human

202   health are unknown.

203   Much recent attention has focussed on the migration of Sb from single-use PET food-contact

204   items like water bottles and ready meal trays. Regardless of storage conditions, measured

205   concentrations of Sb in bottled water have not exceeded the European Commission limit for

206   natural mineral water of 5 μg L^{-1} or the US Environmental Protection Agency maximum

207   contaminant level of 6 μg L^{-1} (Shotyk et al., 2006; Greifenstein et al., 2013; Payan et al., 2017;

208   Roje and Sutalo, 2019). While concentrations of Sb reported in food contained and cooked in

209   PET packaging sometimes exceed the 40 μg kg^{-1} limit specified by European Union Regulation

210   10/2011, typical food consumption results in intakes of Sb that are significantly below the

211   World Health Organization (WHO) tolerable daily intake (TDI) of 6 μg kg^{-1} (Haldimann et al.,

212   2007; Whitt et al., 2016). Antimony concentrations in contemporary PET as residual catalyst

213   (typically 200 to 300 μg g^{-1}; Westerhoff et al., 2008) are, however, an order of magnitude lower

214   than concentrations in many articles of ceramicware and glassware reported in the present study.

215   Moreover, the latter are subject to sustained or long-term use and to foodstuffs that may be

216   relatively acidic.

217   Despite the occurrence of Sb in historical and contemporary food-contact ware, the degree of

218   its migration from such articles is poorly documented. Burns (1935) studied the migration of

219   Sb compounds from the enamel (and glaze) of a ceramic jug that contained 9800 μg g^{-1} Sb

220   using various preparations of 0.5% citric acid solution. Antimony concentrations of up to 5.6

221   mg L^{-1} were detected colorimetrically in successive 18-hour extracts of the original article in

222   which the acid was introduced while boiling, but extraction rose to 20 mg L^{-1} when the jug had

been scoured. Although the author was unable to definitively state that such concentrations were toxic, the prohibition of Sb in enamelware was called for. More recently, Demont et al. (2012) studied the mobilisation of Sb and other elements from customised ceramicware that had been treated with various pigments (including Sb_2O_3 as and opacifier, but of unspecified concentration) and glazed. Extraction after 24 h was greatest, and up to about 0.2 mg L^{-1}, for 4% acetic acid but tests were not performed after disturbing the glaze to simulate sustained usage and washing. Given the high levels of Sb in some of the ceramicware and enamels studied herein and regulations limiting both the concentrations and migration of lead and cadmium from ceramicware and the lip area (within 2 cm of the rim) of decorated drinking glasses (Council of the European Communities, 1984; Rebeniak et al., 2014) it would seem prudent to investigate the rates and mechanisms of Sb migration from food-contact articles more systematically. The potential risks arising from migration should also be evaluated with respect to the WHO 6 $\mu g \ kg^{-1}$ TDI for the metalloid.

Unlike Sb-containing electronic plastics and PET food packaging, and, with the exception of container glassware, the products and applications described herein are not generally recycled. Thus, the use of Sb in ceramics, enamels and paints requires its disposal through landfill or incineration, both of which are challenging because of regulatory or environmental constraints on the metalloid in the waste stream. For example, in Europe the limit value for Sb leaching from granular solid waste (by deionised water and according to standard batch test EN 12457) is among the lowest for all elements reported, and above 5 $\mu g \ Sb \ g^{-1}$ waste is classified as hazardous (Environment Agency, 2013). With regard to incineration, and despite the volatility of many antimony compounds, about 50% of the metalloid remains in bottom ashes due to interactions with other chemicals in the fuel bed and the consequent formation of thermally stable antimonates (Paoletti et al., 2001). Because of the leaching of Sb from incinerator bottom

247   ashes, the EU considers the element to be the most problematic in terms of the reuse and

248   recovery potential of waste material (European Communities, 2008).

249

250   **4. Conclusions**

251   Antimony has a variety of applications in paints, glass and ceramicware that are available to

252   the consumer and accessible to the public. It is commonly encountered in old domestic paints

253   as an anti-chalking agent but appears to be limited to contemporary consumer paints of a

254   speciality nature where the metalloid is used as a fastener for brightly coloured, and often toxic,

255   primary pigments. It is more likely to be found in contemporary paints applied to exterior

256   municipal structures, like roads, boats, playground equipment and telephone kiosks, where it

257   serves as a fastener to reduce the deterioration of bright colours. Antimony is present in glass

258   and glazed ceramics and appears to be added as a fastener for pigments in enamels but clear

259   evidence of its occurrence as the primary pigment, lead antimonite, was restricted to two bright

260   yellow articles of ceramicware. Little information exists on the migration or accessibility of Sb

261   in paints, glass and ceramicware but given concerns about its heath risks and that regulations

262   exist for other metals in such products, more systematic research into Sb mobility is called for.

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Table 1: Occurrence, distribution and concentrations of Sb in paints, ceramicware and container glass. Categories are shown with the number of samples analysed, n (column 2) is the number of cases in which Sb was detected, mean, median, minimum and maximum concentrations are in $\mu g\ g^{-1}$, $colour_{max}$ is the colour of the article returning the highest Sb concentration, $colour_{com}$ is the colour where Sb was most commonly detected, and Sb-Pb denotes the number of cases in which Sb co-existed with Pb above a concentration of 100 $\mu g\ g^{-1}$.

	n	mean	median	min	max	$colour_{max}$	$colour_{com}$	Sb-Pb
Interior extant paint (n = 25)	10	8670	7630	2610	24500	yellow	white (n = 6)	10
Exterior extant paint (n = 25)	9	2530	1020	260	8540	brown	white (n = 3)	9
Municipal extant paint (n = 93)	38	2220	1190	147	27500	grey	yellow (n = 18)	32
Extant boat paint (n = 21)	6	3000	993	176	8080	red	red (n = 3)	4
Consumer paints (n = 18)	3	2070	2670	520	3020	red	red (n = 3)	3
Painted toys/equipment (n = 33)	2	318	318	215	420	red	red (n = 2)	1
Consumer bottle enamel (n = 20)	3	1290	1610	629	1640	green	various (n = 1)	2
Drinking glass enamel (n = 31)	10	2310	1510	507	7790	yellow	green (n = 2)	8
Ceramicware enamel (n = 69)	23	6430	933	308	62800	yellow	yellow (n = 6)	22
Total (n = 335)	104	3360	1180	147	62800	yellow	yellow (n = 29)	91

Figure 1: XRF spectra of four samples and shown as counts per second versus energy from 0 to 35 keV. (a) A second hand teacup pigmented with lead antimonate and glazed with lead oxide and tin oxide; (b) an artists' paint pigmented with cadmium sulphoselenide and where Sb is present, presumably, as a fastener; (c) the exterior paint of a private household where Sb is likely present as an anti-chalking agent with titanium dioxide and lead; (d) a galvanised steel playground gate coloured by bismuth vanadate and where Sb is present as a colour fastener.