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## Assessment of dermal exposure to inorganic lead caused by direct skin contact with lead sheet and moulded PVC profiles

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Lead uptake can occur via inhalation and ingestion exposure, while dermal absorption is thought to be minimal. However, dermal exposure is still important as it can contribute to ingestion exposure due to transfer from the skin to the mouth via the fingers. This study was designed to provide information about i) the potential for dermal lead exposures caused by direct skin contact with lead sheet material, and ii) lead surface levels of polyvinyl chloride (PVC) profiles, as might occur in a consumer or residential environment. This was to enable a health risk assessment to be completed by industry representatives.

Controlled tests were carried out in the laboratory to evaluate the rate of transfer of lead to the skin by varying the number of skin contact events with lead sheet and lead ingots. Contact tests were also carried out at six different sites at two different historic buildings where lead sheet has been used as part of the building fabric or to provide weather or physical protection in keeping with the historic nature of the site. Tests were carried out in the laboratory to determine the presence of lead on the surface of different samples of PVC profiles. These included new (manufactured in 2006) PVC and old (manufactured in 1990) PVC.

A total of 54 dermal samples were collected from six volunteers participating in the lead contact tests in our laboratory. Exposures were low, ranging from less than the limit of detection to 2.24  $\mu$ g/cm<sup>2</sup>. In general tests on lead sheeting and ingots indicated that as the number of contacts with the lead surface increased, the amount transferred to the hands increased. Thirty six samples were collected during the field survey and exposures ranged from 0.07 to 5.05  $\mu$ g/cm<sup>2</sup>. A similar pattern of increasing lead exposure with increasing contact was observed. Twenty samples were collected from the surface of PVC profiles either using wiping or microvacuuming techniques. Micro-vacuuming was inefficient at removing lead and therefore unsuitable. Low levels of lead were removed by wiping from both old and new PVC and exposures ranged from 0.14 to 0.45  $\mu$ g/cm<sup>2</sup>.

Overall, dermal exposures were low and exposure of visitors to historic buildings is likely to be minimal. Levels of lead removed from lead stabilised PVC were low and exposure of residents is likely to be minimal.

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## **1 INTRODUCTION**

This report describes a study undertaken to determine the potential exposure to lead arising from skin contact with lead in the form of lead sheeting and lead ingots, and the surface levels of PVC profiles. This chapter provides some background information about sources of environmental and consumer exposure to lead, the importance of dermal exposure in determining overall levels of exposure to lead and the health effects that may arise following excessive exposure to lead. The following chapters outline the study aims, the methods used and the study results. The implications of the study's findings are discussed in the final chapter.

#### 1.1 BACKGROUND

#### 1.1.1 Sources of exposure to lead

Lead occurs naturally in the earth's crust and has been extracted and used since ancient times. Environmental contamination is mostly a result of man's use of lead and has given rise for concern. The main industrial uses are the lead battery industry, smelting, refining, alloying and casting and work with metallic lead and lead containing alloys. In the past the main sources of exposure for non-occupationally exposed populations were leaded petrol, lead in paint, lead contamination of food in soldered cans and water transported in lead pipes. However, exposure from these sources has been substantially reduced since the reduction in the allowable level of lead in petrol and the introduction of non-leaded fuel. The use of lead is no longer permitted in household paints or in food cans in many countries and most lead water pipes have been removed. Levels of blood lead in the general population have declined since the 1970s and by the 1990s average national levels for blood lead in the EU were mainly below 10  $\mu$ g/dl (Thornton *et al.*, 2001).

Uptake of lead can occur via inhalation and ingestion exposure. For most people who are not exposed occupationally, exposure by the inhalation route will be minimal, with ingestion of food containing trace amounts of lead being the main source of exposure for the general adult population (Thornton *et al.*, 2001). Dermal uptake of lead is thought to be of minimal importance, however, dermal exposure is still important as it can contribute to ingestion exposure due to transfer from the skin to the mouth via the fingers. For example, due to hand-to-mouth activity, young children are likely to be exposed through ingestion of soil or house dust containing lead. Although leaded paint is no longer used, a major source of lead in house dust in the United States comes from leaded paint still present in older houses (Thornton *et al.*, 2001).

#### 1.1.2 Routes of lead exposure

Apart from inhalation exposure the main route of exposure will be ingestion. The rate of permeation of lead through the skin will be minimal and depends on the form of lead, with higher rates likely for more soluble forms of inorganic lead (Stauber *et al.*, 1994). A number of publications have shown a statistically significant positive association between dermal lead exposure and blood lead levels (Askin and Volkmann, 1997, Sun *et al.*, 2002). However, as dermal uptake of lead is minimal it is likely that these associations are due to ingestion exposure after transfer of lead from the hands to the mouth (Cherrie, 2003, Cherrie *et al.*, 2006).

#### 1.1.3 Health effects associated with lead

The adverse health effects of lead can be widespread and have been shown to affect the central nervous system, kidneys, cardiovascular system, reproduction and causing anaemia. IARC have classified lead as a group 2A carcinogen (IARC, 2006). The health effects vary depending on the level and duration of exposure. Young children are especially vulnerable and there is



evidence that exposure to lead can cause development and behavioural problems (Thornton *et al.*, 2001, Lanphear *et al.*, 2005a).

#### 1.1.4 Sources of lead exposure explored by this study

This study was designed to provide information on the level of dermal lead exposures caused by direct skin contact with lead sheet material and on lead surface levels of polyvinyl chloride (PVC) profiles, as might occur in the general population. This information would subsequently inform a health risk assessment (not part of this report).

Lead sheeting is typically used in the building and construction industry. Its durability and malleability make it ideal for use as flashings and weatherings and it is extensively used in historic buildings. Lead ingots are produced in lead refineries and used as feedstock to produce various lead chemicals. Exposure is confined to occupational use.

Transfer of lead from lead sheet material is an obvious route of possible exposure but in the case of PVC material the source is not quite so obvious. PVC is commonly formulated with leadbased stabilisers including diabasic lead phthalates and diabasic lead sulphates to prevent it degrading rapidly during manufacture or in use. Lead stabilisers are the most cost effective and commonly used and are used in pipes, including water pipes, window frames, guttering and electrical cables. Exposure to heat, sunlight and aging may result in lead migrating to the surface of the PVC.

Concern has been expressed in the popular media that lead dust may migrate to the surface of PVC profiles in the residential setting and thereby become available for exposure, possibly by direct skin contact, mouthing by children, or by dispersion into the air. As part of the Vinyl 2010 Voluntary commitment, updated in 2001, the PVC industry has agreed to phase out the use of lead stabilisers by 2015 (http://www.vinyl2010.org/, accessed December 2006).

#### 1.2 OVERVIEW OF STUDY

Controlled tests were carried out in the laboratory to evaluate the rate of transfer of lead to the skin by varying the number of skin contact events with lead sheet and lead ingots. Handling of lead ingots is limited to industrial situations, but has been included in the study at the request of the sponsor. In addition, tests were carried out to determine the presence of lead on the surface of different samples of PVC profiles. These included new and old PVC samples.

Additional repeat contact tests for metallic lead were collected *in situ* from six different sites at two different historic buildings where lead sheet has been used as part of the building fabric or to provide weather or physical protection in keeping with the historic nature of the site. With one exception, all sites were accessible to the public.



## 2 AIMS

The study was intended to supplement existing scientific knowledge about the potential for dermal lead exposure from incidental contact with lead sheet or ingots or due to contact with lead sheeting in publicly accessible places. In addition the potential for transfer of lead compounds from PVC profiles was assessed.

This aim was achieved by addressing the following objectives:

- I. Measurement of the level of lead deposition onto the skin after hand contacts with new lead sheets and ingots in a controlled laboratory environment;
- II. Measurements of dermal exposure after contact with *in situ* lead sheet in historic buildings/sites where lead was used for preservation purposes;
- III. Measurement of lead surface contamination on samples of PVC window frames (old and new);
- IV. Measurement of lead surface contamination on *in situ* PVC window frames. Due to the unavailability of an appropriate test site, this was not achieved prior to completion of this report.





## 3 METHODS

#### 3.1 GENERAL SAMPLING STRATEGY

The dermal loading after incidental contact with lead material was assessed by carrying out a varying number of repeat hand contact tests with lead sheeting and lead ingots in a controlled environment and with lead sheeting *in situ*. Samples were collected from the skin using moist wipes. Each sample comprised three sequential wipes of the palms and fingers of the hand so that the flat of the hand was sampled. These tests were conducted using volunteers drawn from IOM staff.

Samples were also collected from the surface of different samples of PVC mouldings in order to determine the presence of lead contamination. This was done to develop a test method suitable for *in situ* testing in commercial or residential premises. Unfortunately, at the time of writing this report, the sampling of *in situ* PVC had not been carried out due to unavailability of sites with traceable PVC frames.

#### 3.1.1 Laboratory contact tests for lead sheet and lead ingots

It was thought that the level of dermal deposition to the skin from incidental contact with lead sheet would be dependent on the applied pressure and the lateral force applied to the material. The test procedure was therefore designed to incorporate application of a controlled pressure over a set period of time together with a procedure designed to simulate rubbing or grasping of a moving surface.

Three different repeat contact tests were carried out in the IOM laboratory using samples of newly purchased and unused lead sheeting obtained from a local builder's merchant and lead ingots supplied by industry. The laboratory tests were designed to evaluate the amount of lead deposited onto the skin by one, five and ten controlled skin contacts. All tests were carried out by six subjects and included a range of skin types ranging from fair to deeply pigmented. Both male and female subjects were included in these tests. The skin condition of each subject was assessed by measuring the trans-epidermal water-loss (TEWL) on the back of the hand using a Dermalab TEWL meter. The measurements were made prior to the first, second and third set of contact tests and on completion of all tests.

In order to ensure that a consistent level of pressure is applied by each individual the test procedure was arranged in the following manner.

Firstly, a sample of new lead sheet was laid down onto a set of scales and the subject was asked to press down onto the lead sheet to a weight of 10 kg for 10 s using the left hand.

Secondly, a lead sheet sample was rolled around a 50 mm diameter plastic tube to form a 'handle' and attached to a 75 cm length of wire tied to a 2 kg weight. The subject was asked to grasp the lead-clad tube handle with their right hand and raise and lower the weight between two indicated points.

Thirdly, the subject lifted a lead ingot weighing approximately 5 kg from one level of a workstation to another and back again using both hands.

Three wipe samples were collected from the palm of the hand and bulked together into one container. Where both hands were exposed, three wipes were taken from each hand. Each test procedure was carried out with 1, 5 and 10 contacts. Prior to the start of the first test a



background sample of both hands was taken by wiping each hand with three wipes. These were then bulked together into one container. A blank sample was also collected for each volunteer.

The order in which the experiments were carried out was arranged as shown in Table 1.

	Subject A	Subject B	Subject C	Subject D	Subject E	Subject F
Background swab	1	1	1	1	1	1
Sheet 1 contact	2	4	3	8	7	6
5 contacts	3	2	4	9	5	7
10 contacts	4	3	2	10	6	5
Pipe 1 contact	5	7	6	5	10	9
5 contacts	6	5	7	6	8	10
10 contacts	7	6	5	7	9	8
Ingot 1 contact	8	10	9	2	4	3
5 contacts	9	8	10	3	2	4
10contacts	10	9	8	4	3	2

Table 1 Test order for dermal lead exposure

After each contact test subjects thoroughly washed their hands using warm water and soap and dried them with clean paper towels. The dermal exposure level for each volunteer was calculated as the surface loading of lead on the hand divided by the area of the palm which was determined as outlined below.

#### Surface area of hands

The area of the palmar surface of the left and right hands of each volunteer was determined. A stand was constructed so that a digital camera could be positioned at a fixed point above a surface. A photograph was taken and the resulting digital image imported into Corel Draw version 13. The area of the visible surface was outlined and filled in with yellow using drawing tools. The resulting image was then imported into Corel Photo-Paint version 13. Using the histogram facility, a histogram detailing the brightness of different sections of an image with values ranging from 0 (black) to 255 (white) was drawn. This facility also gives the number of pixels corresponding to each level of brightness. The level of brightness for the yellow coloured region was 226 and the number of pixels corresponding to this level and hence the area visible through the camera was 1 994 713. The area of the surface visible through the camera lens was calculated to be 540 cm<sup>2</sup> and it was therefore established that 1 pixel corresponds to 2.71 x  $10^{-4}$  cm<sup>2</sup>. The surface areas of the hands of the subjects participating are given in Appendix 1.

#### 3.1.2 Laboratory testing of PVC profiles

Five samples of new PVC (BS EN ISO 12608:2003, Pb stabiliser), manufactured in November 2006 and five samples of PVC profiles, manufactured in 1990 (stabiliser "probably Pb/Ba/Cd" (personal communication Julian Cubitt, November 2006) were obtained from industry for evaluation. The inner and outer surfaces of the five samples of PVC from 2006 were covered with polyethylene tape, this tape was removed prior to sampling and stored in labelled containers. Since it was thought possible that lead may have been removed with this tape, one tape sample was analysed for lead.

Three methods of sampling for lead were proposed: surface wiping, adhesive tapes and microvacuuming. Initial tests with adhesive tape (Hypafix, 2.5 cm wide) suggested that recovery of



lead from this medium was low (around 75%). Further, since it is expected that level of lead on PVC will be low, the surface area sampled needs to be maximised and the use of adhesive tape will present practical difficulties associated with handling large strips of tape. It was therefore decided to sample the PVC frames using the wipe and micro-vacuum techniques only.

An area of  $108 \text{ cm}^2$  (5.4 cm x 20 cm) was selected for sampling for both wiping and microvacuuming. A wipe sample and micro-vacuum sample was taken from each of the 10 pieces of PVC. In each case the inside of the window frame was sampled, since this will be the surface that residents are most likely to come into contact with.

#### Wiping

This was based on MDHS 97 (HSE, 2002). Three consecutive wipes were used. A consistent wipe pattern was applied whereby the area was wiped once round the edge of the sample region in an anti-clockwise direction and then wiped across a number of times in parallel. The wipe was then folded and the region wiped again using a motion consisting of 10 consecutive circles. Constant pressure was applied throughout sampling. The three wipes were bulked together into one container.

#### Micro-vacuuming

Samples of dust were collected onto a cellulose ester filter (25 mm diameter, 1.0  $\mu$ m pore size) held in electrically conducting cowl. A 5 cm section of Tygon tubing was attached to the sampling inlet, which had a 45° angle cut into the sampling end. The sampling head was attached to a high flow sampling pump calibrated to 10 l/min. A sample was collected over a period of two minutes from the surface, in a consistent pattern, with the area being vacuumed lengthwise and breadthwise twice. Three consecutive samples were taken from the same area. All three filters were placed in one container after sampling.

#### 3.2 FIELD MEASUREMENTS LEAD SHEETING (HISTORIC SCOTLAND)

Samples were collected at two locations owned by Historic Scotland that are open to the public. Six test items were included in the study from two different sites selected by Historic Scotland. The six locations selected were:

- 1. A horizontal lead sheet surface on a lead gazebo roof both hands
- 2. Leaded windows both hands
- 3. Sloping lead pieces on wooden steps both hands
- 4. A non painted lead cap (polished) on a hand rail in the great hall right hand
- 5. New non painted lead cap on a hand rail left hand
- 6. Non painted lead cap (non polished) on a hand rail- right hand

With the exception of the gazebo roof, all sampling locations were accessible to the public.

Two IOM researchers (MvT and AS) were involved with the sample collection procedure. For each test one person acted as the test subject and the other collected the dermal sample. Each test was repeated with the roles reversed. The tests were conducted by applying the flat of one or both hands to the lead product under study. For tests 1, 2 and 3, the subject applied the full weight of his/her body to the test item for ten seconds. This comprised one contact. For tests 4, 5 and 6, one contact test comprised of rubbing the hand over the lead cap for 10 seconds prior to colleting a sample. This test was repeated for 50 and 100 seconds. In each case, subject A carried out the contact tests first.

The order in which the experiments were carried as shown in Table 2. The test order was the same for both subjects.



	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Background swab	1	1	1	1	1	1
1 contact	2	4	3	4	3	2
5 contacts	3	2	4	3	2	3
10 contacts	4	3	2	2	4	4

Table 2 Test order for dermal lead exposure field study

Note, for tests 4, 5 and 6, one contact comprised rubbing for 10 seconds, five contacts comprised 50 seconds rubbing and 10 contacts comprised 100 seconds rubbing

In between each sample, the hands were washed in a bucket of warm, soapy water and rinsed in bucket with clean water. Whenever feasible, the water was refreshed after completion of all samples at a location. Unfortunately, due to the distance between the sample location and facilities, it was not possible to refresh the water between tests 2 and 3, and between tests 4, 5 and 6.

#### 3.3 ANALYTICAL METHODS

All samples were analysed for lead by inductively coupled plasma atomic emission spectroscopy (ICP/AES). The documented in-house method, based on OSHA method 121 (OSHA, 1991) is accredited by the United Kingdom Accreditation Service (UKAS) under UKAS accreditation number 0374. The samples were desorbed by heating with nitric acid. Each sample was cooled, transferred to a volumetric flask, rinsed and made up to a standard volume using deionised water. The acid digests were then analysed by ICP/AES.

Calibration standards were prepared using known weights of analytical grade reagents and the sample masses were determined with reference to these calibration standards. The analytical accuracy for this method was  $\pm 3\%$ .

The quantity of inorganic lead in each sample was expressed as a measure of dermal surface loading in terms of mass per unit area ( $\mu g/cm^2$ ) taking into account the mass of lead in the sample and the palmar surface area of the test subject, or in the case of the PVC, the measured surface area that was wiped or vacuumed.

Samples of blank moist wipes and blank filters were analysed to determine the background lead content of the test media and to establish the analytical detection limit. These blank levels were used to correct the test samples when they were analysed. The limit of detection for this method was  $0.1 \mu g$ .

#### 3.4 EVALUATION OF SAMPLING METHODS

Since the moist wipe sampling procedure has been thoroughly evaluated for lead in previous work (Hughson, 2005) no additional testing was considered to be necessary. The analytical recovery from cellulose ester filters was determined by preparing a small number of spike samples. These spike samples were prepared by adding known amounts of lead directly to the filters. A range of spike masses from 10  $\mu$ g to 100  $\mu$ g were chosen.

#### 3.4.1 Sampling efficiency

Tests were carried out to investigate the removal efficiency of wipe sampling and microvacuuming from PVC. A litharge provided by Penox Gmbh, labelled Litharge L9 was used for



these tests. According to the technical data sheet which accompanied the sample, the material comprised a minimum of 99.8% lead (II) oxide with a maximum metallic lead content of 0.01%.

It is anticipated that level of lead recovered from PVC will be low. Ideally, pre-weighed quantities of litharge should be applied to a defined area of PVC. However, it was not possible to accurately weigh out litharge samples of less than 0.3 mg and low level spike samples were therefore prepared using a 25  $\mu$ g/ml stock solution using litharge in 10% nitric acid.

A PVC sample from 1996 (Pb stabiliser, day 263 1990 (Year 06)) was chosen as the test surface. This was thoroughly washed prior to use. Two sections of 50 cm<sup>2</sup> were marked off on the surface, one for wipe sampling and one for micro-vacuuming. Spike levels of 1 ml, 2 ml and 4 ml were applied using a pipette to the surface and dried using a hot air gun. This corresponds to levels of  $0.5 \,\mu\text{g/cm}^2$ ,  $1 \,\mu\text{g/cm}^2$  and  $2 \,\mu\text{g/cm}^2$ , for a 50 cm<sup>2</sup> surface area. Prior to each loading, three consecutive wipes were applied to the test area, in order to check for any residual lead on the surface.

#### 3.5 DATA ANALYSIS

The dermal exposure data was summarised in terms of maximum and minimum values, median and the upper 90<sup>th</sup> percentile level. This is the summary data normally required for EU regulatory risk assessments (ECB, 2003).

For sample results below the limit of detection, the exposure value was set to a level of half the limit of detection, in accordance with the approach suggested by Rajan-Sithamparanadarajah *et al.*, 2004.

The association between surface loading on the hands and transepidermal water loss was investigated using Pearson's correlation coefficient. Analysis of variance (ANOVA) with linear trend analysis was carried out to determine whether the number of contacts was associated with dermal exposure using Genstat v9. Differences between lead recovery from 1990 PVC and 2006 PVC were investigated using the two sample t-test. All data were log transformed before statistical analyses.





## 4 **RESULTS**

#### 4.1 LABORATORY CONTACT TESTS

A total of 54 dermal samples (excluding field blanks) from six volunteers were collected for lead analysis. All samples were corrected using the appropriate field blanks. The limit of detection (LOD) was 0.1  $\mu$ g. Using the areas of the palms of the hands (Appendix 1, Table A1), the sample LOD was approximately 0.001  $\mu$ g/cm<sup>2</sup>. Background levels varied from < LOD to 0.12  $\mu$ g/cm<sup>2</sup>, with a median of 0.02  $\mu$ g/cm<sup>2</sup>. The results are detailed in Tables 3 to 5 and Figures 4.1 to 4.3 for lead sheeting on scales, lead sheeting wrapped round a pipe and lead ingots respectively.

The results indicate that, in general, the loading on the hand increases with the number of contacts.

Subject	Surface loading (μg/cm <sup>2</sup> )					
	One contact	Five Contacts	Ten contacts			
А	0.09	0.40	1.52			
В	0.03	0.14	0.20			
С	< LOD	0.10	0.16			
D	0.09	0.32	0.32			
Е	0.20	2.06	1.96			
F	0.21	0.71	1.73			
Minimum	<lod< td=""><td>0.10</td><td>0.16</td></lod<>	0.10	0.16			
Maximum	0.21	2.06	1.96			
Median	0.09	0.36	0.92			
GM	0.04	0.37	0.61			
90 <sup>th</sup> percentile	0.21	1.39	1.84			

# Table 3 Surface loading of lead on the skin after repeated contacts with lead sheeting placed on scales

 Table 4 Surface loading of lead on the skin after repeated contacts with lead sheeting rolled around a plastic pipe

Subject		Surface loading (µg/cm <sup>2</sup>	·)
Subject	One contact	Five Contacts	Ten contacts
А	0.25	0.16	0.34
В	0.08	0.52	0.33
С	1.84	0.93	2.24
D	0.18	0.45	0.49
E	0.46	0.67	0.42
F	0.46	1.48	2.14
Minimum	0.08	0.16	0.33
Maximum	1.84	1.48	2.24
Median	0.35	0.60	0.45
GM	0.33	0.57	0.69
90 <sup>th</sup> percentile	1.15	1.20	2.19



Subject	Surface loading (μg/cm²)					
	One contact	Five Contacts	Ten contacts			
А	0.25	0.63	0.45			
В	0.15	0.45	0.54			
С	0.24	1.59	1.38			
D	0.90	1.03	0.94			
Е	0.47	0.85	0.97			
F	0.29	0.91	0.65			
Minimum	0.15	0.45	0.45			
Maximum	0.90	1.59	1.38			
Median	0.27	0.88	0.79			
GM	0.32	0.84	0.76			
90 <sup>th</sup> percentile	0.69	1.31	1.18			

Table 5 Surface loading of lead on the skin after repeated contacts with lead ingots



Figure 4.1 Surface lead loadings for contact with lead sheeting placed on scales





Figure 4.2 Surface lead loadings for contact with lead sheeting wrapped round a pipe



Figure 4.3 Surface lead loadings for contact with lead ingots

The results of the ANOVA with linear trend analyses are shown in Tables 6 to 8 for lead sheeting on scales, lead sheeting wrapped round a pipe and lead ingots respectively. For all three scenarios there were statistically significant increasing linear trends in the surface loading of the hands with number of contacts. Further, the differences between number of contacts and surface loadings were also statistically significant for lead sheeting on scales and lead ingots (p=0.002 and p=0.001 respectively), but not for lead sheeting wrapped around pipe (p=0.102).



Source of variation	Degrees of freedom	Sums of squares	Mean squares	Variance ratio	p-value
Subject	5	31.04	6.21	5.94	
No. of contacts	2	25.30	12.65	12.11	0.002
Linear trend	1	21.17	21.17	20.27	0.001
Residual	10	10.45	1.04		
Total	17	66.78			

**Table 6** Analysis of variance table for exposure to lead in relation to the number of contacts with lead sheeting on scales

**Table 7** Analysis of variance table for exposure to lead in relation to the number of contacts with lead sheeting wrapped around pipe

Source of variation	Degrees of freedom	Sums of squares	Mean squares	Variance ratio	p-value
Subject	5	9.63	1.93	6.45	
No. of contacts	2	1.72	0.86	2.89	0.102
Linear	1	1.53	1.53	5.12	0.047
Residual	10	2.98	0.30		
Total	17	14.33			

**Table 8** Analysis of variance table for exposure to lead in relation to the number of contacts with lead ingots

Source of variation	Degrees of freedom	Sums of squares	Mean squares	Variance ratio	p-value
Subject	5	2.51	0.50	4.12	
No. of contacts	2	3.32	1.66	13.64	0.001
Linear	1	2.01	2.01	16.52	0.002
Residual	10	1.22	0.12		
Total	17	7.05			

The relationship between TEWL and surface loading with lead is shown in Figure 4.4 below. No association between TEWL and surface loading was discerned (r=0.36, p=0.087).







#### 4.2 FIELD MEASUREMENTS LEAD SHEETING (HISTORIC SCOTLAND)

A total of 36 dermal samples (excluding field blanks) were collected for lead analysis (Table 9). All samples were corrected using the appropriate field blanks. The LOD was 0.1  $\mu$ g. Using the areas of the palms of the hands (Appendix 1, Table A1), the sample LOD was approximately 0.001  $\mu$ g/cm<sup>2</sup>. Background levels varied from <LOD to 0.26  $\mu$ g/cm<sup>2</sup>, with a median of 0.01 $\mu$ g/cm<sup>2</sup>.

Location	Subject	One contact	Five contacts	Ten contacts
Gazebo	А	0.41	2.90	2.77
	В	0.41	1.00	2.69
Leaded windows, Lord's hall	А	0.29	1.05	0.90
	В	0.15	0.20	0.07
Wooden steps	А	0.18	0.33	0.74
	В	0.11	0.17	0.21
Old polished hand rail, Great hall $^*$	А	0.15	0.89	2.48
	В	0.43	0.50	1.52
New unpolished hand rail, Great hall <sup>*</sup>	А	1.38	3.48	1.98
	В	0.45	1.39	1.56
Old unpolished rail, Mid tower <sup>*</sup>	А	4.96	4.44	0.20
	В	1.85	3.83	5.05
Minimum		0.11	0.17	0.07
Maximum		4.96	4.44	5.05
Median		0.41	1.03	1.54
GM		0.44	1.02	0.99
90 <sup>th</sup> percentile		1.80	3.80	2.76

#### Table 9 Field results

\* Note, for these tests one contact comprised rubbing for 10 seconds, five contacts comprised 50 seconds rubbing and 10 contacts comprised 100 seconds rubbing

Generally, the surface loadings for subject A were greater than those of subject B, which may be due to the fact that A was always the first test candidate. In general, as the number of contacts increases so does the loading on the hands. One notable exception to this is the results for subject A obtained from the unpolished rail in the Mid tower, where the trend was reversed with the highest loading being obtained for one contact and the lowest loading for 10 contacts. This rail was in an area not likely to be accessed by the public and had clearly not been touched in a long time, being very dirty. Therefore, it is likely that the first touch would have removed a lot of loose surface debris and perhaps lowered the subsequent availability of lead.

The results also suggest that rubbing removes more lead from the surface than simply placing the hand on the surface.

#### 4.3 LABORATORY SURFACE TESTS - PVC WINDOW FRAMES

A total of 20 samples (excluding blanks) were collected, 10 from 1990 PVC and 10 from 2006 PVC. Of the 10 samples from each type of PVC, five were wipe samples and five micro-vacuum samples. All samples were corrected using the appropriate field blanks. The LOD was 0.1  $\mu$ g. Since the area sampled was 108 cm<sup>2</sup>, the sample LOD was approximately 0.001  $\mu$ g/cm<sup>2</sup>.

The results are presented in Tables 10 and 11 respectively. It is clear that wiping is a more effective technique than micro-vacuuming for removal of lead from the surface of PVC. Removal of lead from 1990 PVC ranged from 0.14 and 0.21  $\mu$ g/cm<sup>2</sup>, with a geometric mean of 0.19  $\mu$ g/cm<sup>2</sup>. For the 2006 PVC, removal ranged from 0.17 and 0.45  $\mu$ g/cm<sup>2</sup>, with a geometric mean of 0.31  $\mu$ g/cm<sup>2</sup>.



Sampla	Sampling method				
Sample	Wiping (μg/cm²)	Micro-vacuuming (μg/cm²)			
1	0.20	0.01			
2	0.21	0.02			
3	0.21	0.004			
4	0.14	0.004			
5	0.21	0.002			
Minimum	0.14	0.002			
Maximum	0.21	0.02			
Median	0.21	0.004			
GM	0.19	0.01			
90 <sup>th</sup> percentile	0.21	0.01			

**Table 10** Surface lead (µg/cm<sup>2</sup>) detected on PVC manufactured in 1990

Table 11 Surface Lead levels ( $\mu$ g/cm<sup>2</sup>) detected on PVC manufactured in 2006

Sampla	Sampling method				
Sample	Wiping (μg/cm²)	Micro-vacuuming (μg/cm <sup>2</sup> )			
1	0.37	0.001			
2	0.45	<lod< td=""></lod<>			
3	0.40	0.001			
4	0.25	0.001			
5	0.17	<lod< td=""></lod<>			
Minimum	0.17	<lod< td=""></lod<>			
Maximum	0.45	0.001			
Median	0.37	0.001			
GM	0.31	<lod< td=""></lod<>			
90 <sup>th</sup> percentile	0.43	0.001			

The recovery from the 2006 PVC was statistically significantly higher than for the 1990 PVC sample (two sample t-test statistic, p=0.021). However, as only very small amounts were removed from both types of PVC, it is unlikely that this difference is of practical significance.

A sample of the tape covering one of the 2006 PVC samples was also analysed for lead and a level of 0.42  $\mu$ g/cm<sup>2</sup> was obtained (LOD=0.1  $\mu$ g, sample LOD=0.0002  $\mu$ g/cm<sup>2</sup>), which is comparable with the amounts of lead removed from the 2006 PVC samples by wiping. This may indicate that the total lead from the surfaces of the 2006 PVC frame was higher than shown in Table 11.

#### 4.3.1 Evaluation of sampling methods

Initial spike recovery tests from cellulose ester filters were carried out and the results are reported in Table 12.

Spiked mass lead (μg)	Mass lead recovered (μg)	Percentage Recovery
10	10.6	106
50	45.9	91.8
100	98.4	98.4

Table 12 Initial tests of recovery from cellulose ester filters



These preliminary results indicate that there was good recovery at all spike levels.

#### Sampling efficiency

Preliminary recovery tests for the wiping and micro-vacuuming sampling methods were carried out by spiking a piece of washed PVC with three different level of lead litharge solution. The results are presented in Table 13. All samples were corrected using the appropriate field blanks. The LOD was 0.1  $\mu$ g. Since the area sampled was 50 cm<sup>2</sup>, the sample LOD was 0.002  $\mu$ g/cm<sup>2</sup>. It is apparent that recovery was poor for both methods, particularly for micro-vacuuming where the amounts recovered were less than the limit of detection for the two lower spike levels.

	Spike	Wiping		Micro-vacuuming	
Sample	mass Pb (ug)	Actual mass recovered (µg)	% Recovery efficiency	Actual mass recovered (μα)	% Recovery efficiency
1	25	8.9	35.6	<lod< td=""><td>-</td></lod<>	-
2	50	3.9	7.8	<lod< td=""><td>-</td></lod<>	-
3	100	17.6	17.6	0.1	0.1

#### Table 13 PVC recovery tests

Prior to spiking, the surfaces to which the known amounts of lead litharge solution were to be applied were wiped to ensure that all lead was removed. However, wipe sampling prior to spiking indicated that there were still residual amounts  $(1.2-2.6 \ \mu g)$  of lead on the surface of the PVC frame after thorough cleansing.

The results show that recovery from spikes prepared using lead litharge solution is not successful. However, we believe that in the case of the wiping methods, this is most likely to be a problem of spiking due to small quantities and use of a hot air drier rather than a problem of the sampling and analytical methods. However, it does provide further evidence that microvacuuming is not a suitable method.





## 5 DISCUSSION AND CONCLUSIONS

This study investigated i) the potential exposure to lead arising from skin contact with lead sheeting and lead ingots; and ii) surface lead levels of PVC profiles. Controlled tests were carried out in the laboratory to evaluate the rate of transfer of lead to the skin by varying the number of skin contact events with lead sheet and lead ingots. In addition, contact tests for metallic lead were also collected *in situ* from six different sites at two different historic buildings where lead sheet was used as part of the building fabric or to provide weather or physical protection in keeping with the historic nature of the site. Tests were also carried out to determine the presence of lead on the surface of different samples of PVC profiles.

#### 5.1 METHOD DEVELOPMENT

A wipe sampling method was used to measure contamination on exposed hands in the laboratory tests and field study. This allowed an assessment of the average skin surface loading to be made. It should be remembered that all methods of dermal sampling are subject to limitations and the wipe method is no exception (Brouwer *et al.*, 2000). However, this method has previously been validated for lead, with a low background level of lead in the sampling media, good recovery efficiency and an acceptable limit of detection being demonstrated (Hughson, 2005). The suitability of this technique for use for assessing dermal exposure levels to lead and lead compounds has been established (Hughson, 2005).

Three methods of sampling lead from the surface of PVC profiles were considered: wiping, tape striping and micro-vacuuming. The tape striping technique using 2.5 cm Hypafix tape was rejected partly on the basis of poor recovery and partly due to practical difficulties associated with the handling of long strips of tape necessary for sampling entire window frames. Preliminary recovery tests of spiked solutions of lead from PVC profiles for the wiping and micro-vacuuming sampling methods were carried out, with the intention that more comprehensive tests be carried out once the most suitable method for lead recovery from PVC had been identified. The results indicated that despite washing prior to sampling, lead still remained on the surface of the PVC. The recovery tests performed very poorly. However, we are confident that this is a problem with the spiking procedure rather than the sampling and analytical methods. It is likely that a combination of very small amounts being applied together with the use of a hot air drier caused the poor recovery. It is important that if further work is carried out on lead sampling from PVC window frames that more suitable and comprehensive recovery tests are developed.

Micro vacuuming did not prove to be an effective method for removing lead from PVC. Lead was detected on all wipe samples taken from 1990 and 2006 PVC and appears to be a suitable technique for measuring surface lead. However, as indicated above, further work needs to be carried out on sampling efficiency. A significant difference between recovery from the 1990 and 2006 samples was observed with more lead being recovered from 2006 PVC. However, since the amounts were low, it is unlikely that this difference is of any practical significance. One sample of the polyethylene tape covering the 2006 PVC was analysed for lead and found to contain lead at a level comparable with that removed by wiping 2006 PVC. Although it is not possible to draw conclusions from one sample it is likely that lead will be found on the other tape samples.

#### 5.2 TRANSFER OF LEAD TO THE SKIN

Controlled tests were carried out in the laboratory to assess the transfer of lead to the skin by varying the number of skin contacts with lead sheet and ingots. These indicated that, for lead



sheeting pressed on scales and lifting lead ingots, there was a statistically significant increase in the surface loading on the hands as the number of contacts increased. Results from the field survey generally showed a similar pattern with increasing numbers of contacts resulting in increasing surface loading on the hand(s). All surfaces except the gazebo roof were accessible to the public. In general, surface loadings appear higher than for the lab tests. However, it is clear from the results that transfer occurs even with brief or minimal contact with lead.

#### 5.3 SIGNIFICANCE OF THE DERMAL EXPOSURE LEVELS FOUND

There are no published or publicly available reports of consumer exposure to lead in public places and hence it is not possible to make any comparison with our results. Hughson (2005) has reported results for dermal exposure to lead from three different industries (Table 14). It should be noted that gloves were worn or provided and hence results reflect the actual exposure. Hughson reported that lead exposures were low for most routine jobs. These levels are at least one order of magnitude higher than those observed during the laboratory contact tests.

	Dermal lead exposure (μg/cm²)		
	Zinc/lead refinery (n=14)	Lead refinery (n=16)	Lead chemicals (n=14)
Minimum	1.0	1.0	1.4
Maximum	21.3	92.6	160.9
Median	2.7	4.6	16.1
90 <sup>th</sup> percentile	14.2	41.9	88.6

Table 14 Summar	y of dermal lead e	posure by industry	/ (Hughson, 2005)
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Although much lower than those obtained from occupational settings, it is considered that visitors to historic monuments may be exposed to lead through contact with lead on windows, on hand rails or on wooden steps. Visitors will only have occasional, if any, exposure and it is unlikely that such exposure will contribute significantly to their long term blood levels. However, it is recommended that an assessment of the exposure of visitors be made in order to confirm this.

Another group potentially at risk are those carrying out maintenance of existing lead components and installing new ones. Maintenance staff should be advised of the risks and instructed to wear suitable protective clothing and wash thoroughly after contact with lead.

#### 5.4 SIGNIFICANCE OF THE SURFACE CONTAMINATION LEVELS FOUND

In 2001 the Environmental Protection Agency (EPA) in the United States set dust lead hazard levels of 40  $\mu$ g/ft<sup>2</sup>, 250  $\mu$ g/ft<sup>2</sup> and 400  $\mu$ g/ft<sup>2</sup> for floors, window sills and window troughs respectively. These correspond approximately to 0.04  $\mu$ g/cm<sup>2</sup>, 0.27  $\mu$ g/cm<sup>2</sup> and 0.43  $\mu$ g/cm<sup>2</sup>. The highest value from PVC found in the laboratory tests in was 0.45  $\mu$ g/cm<sup>2</sup>, slightly higher than the EPA standard for window troughs. However, it is not known how the levels of lead obtained from wiping PVC would relate to those found from wiping window troughs and no inferences should be drawn about the amount of lead a person living in a house with PVC window frames would be exposed to. Further work would be necessary in order to investigate this.

A large number of studies (including Lanphear et al., 1996, 2005b, Tohn et al. 2000, Jacobs et al., 2002) on exposure to lead in residential houses in the United States have been published



which indicate that lead-based paints and the dust it generates are important sources of lead exposure, particularly for children. Although these were originally reported in  $\mu g/ft^2$ , they have been transformed to  $\mu g/cm^2$  for the purposes of comparison with this study. In a study to investigate the relationship between lead contaminated house dust and blood lead levels in 205 children, Lanphear *et al.* (1996) reported geometric mean lead dust loadings of 0.01, 0.02, 0.18 and 2.97  $\mu g/cm^2$  for uncarpeted floors, carpeted floors, window sills and window troughs respectively. Tohn *et al.* (2000) studied lead dust loadings in 22 homes which were to have lead hazard control work carried out on them. They reported pre-intervention median loadings of 0.02, 0.02, 0.29 and 5.87  $\mu g/cm^2$  for uncarpeted floors, carpeted floors, carpeted floors, window sills and window sills and window troughs respectively. Jacobs *et al.* (2002) carried out a survey of a national representative random sample of 831 housing units, geometric mean dust loadings of 0.001, 0.01 and 0.10  $\mu g/cm^2$  were reported for floors, window sills and window troughs respectively. Further, 90<sup>th</sup> percentiles of 0.01, 0.19 and 3.04  $\mu g/cm^2$  were also reported in this study.

However, it is difficult to make comparisons between the findings described above and the PVC surface lead levels reported here. The source of lead in the aforementioned studies is leaded paint, and presence of lead on various surfaces suggests that lead has been released into the indoor environment. In contrast, the source of the surface lead levels was the PVC itself, and it is unclear whether lead is released to the indoor environment. The results summarised in the above paragraph are provided for the purpose of comparison and no conclusions regarding exposure arising from PVC should be drawn.

#### 5.5 POTENTIAL RISK TO HEALTH

The question of whether lead in PVC constitutes a health hazard has been a long running issue. PVC products in the home which may contain lead include window frames, vinyl window blinds, lead-based paint in older homes and toys. In 1996 the U.S. Consumer Product Safety Commission (CPSC) issued a statement warning of the hazard from imported vinyl miniblinds, recommending that parents of young children remove these from their homes. In 1997 Greenpeace published a report claiming that hazardous levels of lead were to be found in children's PVC toys. However, these findings were subsequently questioned by the CPSC (1997) who conducted a study on a number of the same products. Although some of the products were identified as containing lead, their results indicated that children would not be exposed to hazardous levels of lead during normal use. Both Greenpeace and CPSC used wipes to sample the products, however, Greenpeace weathered the products by exposing them to alternating cycles of heat and light. CPSC felt that such weathering would not occur during normal use.

Many reports linking blood lead levels to children's intelligence have been published (among them Lanphear *et al.*, 1996, Canfield *et al.*, 2003, Chen *et al.*, 2005, Lanphear *et al.*, 2005a, 2005b). Further, since one of the main sources of household lead is lead-based paint (Lanphear and Roghmann, 1997) lead in paint is implicated in influencing blood lead levels and hence intelligence. As far as we are aware, there are no studies linking PVC to blood lead in children. The dose arising from exposure to lead in paint must be very much greater than that, if any, arising from PVC since the main source of exposure for young children is likely to be the ingestion of flakes of paint having a high lead content.

#### 5.6 RECOMMENDATIONS FOR FURTHER RESEARCH

Although levels of lead removed from lead stabilised PVC were low and exposure of residents is likely to be minimal, the proposed sampling of PVC window frames *in situ* should confirm this. The results of this study suggest that there is a small but statistically significant difference



in lead surface levels for new and relatively old PVC frames. Any further *in situ* measurements should include a range of lead stabilised PVC frames. In addition, it is recommended that samples from the surrounding surfaces be collected, for example, window sills and floors. Furthermore, dermal contact studies should be carried out to assess the transfer of lead from the PVC window frames and possible ingestion exposure of lead.

#### 5.7 SUMMARY OF CONCLUSIONS

In summary, laboratory tests on lead sheeting and ingots indicated that generally as the number of contacts with the lead surface increased, the amount transferred to the hands increased. A similar pattern was observed in the field studies carried out at historic buildings. Although visitors to these buildings are likely to have minimal, if any, exposure, it is recommended that an assessment of their exposure and risks be made. Lead was detected on the surfaces of new (2006) and old (1990) PVC window frames. Although unlikely that the low levels of lead found on the surface of the PVC window frames will present a significant health hazard to residents, it is recommended that an assessment of lead on *in situ* PVC frames and on other surfaces and dermal contact studies are carried out in order to confirm this.



## 6 STATEMENT OF QUALITY

IOM recognise and adopt accepted UK guidelines for good survey practice

This project was carried out under the IOM project management system, which includes preparation of a written protocol for the research and periodic auditing of the work by experienced senior scientists not actively involved in the study.

IOM has UKAS accreditation for several measurement techniques. While the laboratory analysis of all samples collected under this study is covered by the UKAS accreditation, the sampling protocol is a non-standard research procedure and cannot easily be accredited. However, the sampling procedures followed the general quality procedures required by the overall quality management system. Sampling and analytical quality assurance included appropriate calibration checks, replicate analyses and blank samples

Data processing and reporting was subject to the internal data processing control procedures. Raw data is stored for five years and can be audited by the sponsor.







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## **APPENDIX 1 – SURFACE AREA OF HANDS**



Subject	Area	ı (cm²)
Subject	Left hand	Right hand
А	150	156
В	102	100
С	137	142
D	127	130
Е	117	125
F	169	177

#### Table A1 Surface area of hands



