Journal of Exposure Analysis and Environmental Epidemiology (1999) 2, 106–112

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A field comparison of two methods for sampling lead in household dust

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Comparability of dust lead measurements has been a difficult problem due to different sampling and analysis techniques. This paper compares two dust sampling techniques, the U.S. Department of Housing and Urban Development (HUD) dust wipe method and the Lioy, Wainman, Weisel (LWW) sampler. The HUD method specifies using a moist towelette to pick up as much dust as possible in a specified area and estimates total lead loading. The LWW sampler collects the dust on preweighed wetted filter media, and provides greater standardization of the sampling path and pressure applied. LWW samples were analyzed using inductively coupled plasma mass spectronomy (no samples below minimum detection limit), while HUD samples were analyzed using flame atomic absorption (32% of samples below minimum detection limit). A bootstrapping technique was used in the analysis to contend with those HUD samples below the minimum detection limit. Mixed model equations were generated to predict HUD values from LWW results, and to examine the effects of sampling location, time, and method. The results indicate that the two samplers performed similarly under field conditions, although the LWW sampler produced consistently lower lead loading estimates. LWW values that predicted HUD lead clearance values of 100 $\mu g/ft^2$ for floors and 500 $\mu g/ft^2$ for window sills were 72 $\mu g/ft^2$ and 275 $\mu g/ft^2$, respectively. To examine internal reproducibility, duplicate samples were taken using both the HUD and LWW methods. Correlation results within paired samples indicated a statistically significantly higher ($p < 0.001$) internal reproducibility for lead loading, for the LWW sampler ($r = 0.87$), than for the HUD method ($r = 0.71$). Some of the differences appeared to be related to the analytical methods.

Keywords: *clearance values, dust, HUD dust wipe, lead, LWW sampler, sampling.*

Introduction

Lead dust on household surfaces is known to contribute to elevated childhood blood lead levels. Several dust-collection methods have been designed to effectively measure lead loading and/or lead concentration (Vostal et al., 1974; U.S. HUD, 1990; Lioy et al., 1993), and have been field tested and compared in numerous studies (Sayre and Katzel, 1979; Que Hee et al., 1985; Davies et al., 1987; McArthur, 1992; Farfel et al., 1994a,b; Lanphear et al., 1995; U.S. EPA, 1995; Freeman et al., 1996). Until recently, there has been no standardization of sampling methods. This has led to an inability to compare lead loadings because of differences in sampling efficiency, and/or analytical techniques. The use of premoistened towelettes to wipe up dust, within a defined template, with a fixed number of passes is a method that has been widely used by the U.S. Department of Housing and Urban Development (HUD) (U.S. HUD, 1990). Because of ease of use, and relatively low cost, the HUD method has become the industry standard for the assessment of lead loading. Regulatory standards for lead clearance level testing on hard surfaces, such as floors and window sills, following abatements, are based on this method (U.S. HUD, 1995).

The Lioy, Wainman, Weisel (LWW) sampler (Lioy et al., 1993, U.S. Patent # 5373748), was designed not only for the measurement of lead loading, but also dust loading and lead concentration. Contaminant concentration has been shown to be a useful metric in source ascertainment (Adgate et al., 1995, 1998; Freeman et al., 1997). The LWW sampler was designed to control the pressure applied to the wipe during the collection process in order to obtain a more standardized measurement. Because the applied pressure is limited and the moistened filter paper is held in place by a smooth solid block as it passes over the surface, the LWW sampler is believed to collect less material from the pits and cracks of uneven collection surfaces than the HUD method. A second potential difference which may affect the collection efficiency is, that the detergent in the HUD sampler may solubilize more dirt and dust from surfaces than the water used with the LWW sampler. Adgate et al. (1995) analyzed the quantitative relationships between lead loading, dust loading, and lead concentration, using the LWW sampler in a field trial, but

1. Abbreviations: HEPA, high efficiency particulate air filter; HUD, U.S. Department of Housing and Urban Development sampler; LWW, Lioy, Wainman, Weisel sampler; TLC, Treatment of Lead-Exposed Children Study; TSP, trisodium phosphate.

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Received 6 October 1997; accepted 7 August 1998.

could not compare these metrics to results found in other studies which used different methods.

To assist in future comparisons of LWW results with clearance standards developed for the HUD method, a study was conducted in which the two methods were used in side-by-side sampling in homes of lead-exposed children. Using both samplers in a field trial allowed comparison of the internal reproducibility, and the individual effects of sampling time (before and after cleaning), room (kitchen or play area), and sampling location (floor vs. sill). The relationship between the HUD and LWW samplers was quantified so that results from studies using the LWW sampler could be directly compared to those using the HUD dust wipe.

Methods

Sampling Overview

All samplings with the LWW and HUD protocols were done at the New Jersey site of the Treatment of Lead-Exposed Children Study (TLC), an ongoing national, multicenter trial of chelation therapy. Children being studied in the TLC study have blood lead levels between 20 and 44 μg/dl, have never been chelated prior to entry into the study, and were between the ages of 12 and 33 months at the time of randomization. Children were referred to the program by physicians and health departments in northern New Jersey, and by self-referral following a recruitment letter. Homes which were not currently undergoing abatement and were considered structurally acceptable, were considered cleanable by study staff, and thus were approved for participation in the study. Prior to randomization, each home was thoroughly cleaned, using a three-pass system. This included HEPA vacuuming, followed by a wet mopping with a solution of trisodium phosphate (TSP), using a two-bucket method, followed by another HEPA vacuuming after drying. Samples were taken prior to the cleaning, and within 24 h after completion. A pair of floor dust wipe samples, one HUD and one LWW, was taken side-by-side on hard surfaces (e.g., wood, linoleum, stone, tile). Carpets were not sampled in these comparisons.

HUD Sampling

Little Ones Baby Wipes Litely Scented (K-Mart Corp.) were used for all HUD dust wipe samples. A cleanable, reusable 1-ft^2 (1/8 in. thick) plastic template marked the floor sampling area. The window sill sampling area was individually measured with each sample. One field blank was taken per house, along with two floor samples, and one window sill sample. One floor sample was taken in the kitchen, as close to the wall as possible. The second floor sample was taken in another room where the child played. Both floor samples were taken from perimeter locations directly under a window (if possible).

The window sill sample was then taken above the play area floor sample. The left side of the window sill was used for precleaning samples, and the right side was used for post-cleaning samples. Duplicates were taken side-by-side with the regular sample. The field personnel used the standard HUD dust wipe sampling technique (U.S. HUD, 1990).

The HUD and LWW samples were collected using a side-by-side protocol on the floor and, when possible, on window sills. Some window sill pairs were taken on different sills, but all second sills selected were similar to the first sill, e.g., proximity to the first sill, size, structure, substrate material, and paint condition.

LWW Sampling

The LWW wipe sampler used, shown in Figure 1, was the commercially available model. It is the second generation LWW sampler and provides greater stability of the sampling media, as compared to the original model, by providing a locking mechanism to hold the filter media in place when sampling. The LWW template was placed directly next to the HUD template. One field blank was taken per day. For every tenth sample, a duplicate sample was obtained for the assessment of internal reproducibility. The following protocol was followed for all sample sets.

Using a pair of nonpowdered vinyl gloves, the technician placed a rectangle of Acquell Polyurethane 1/4-in. thick foam (32 × 22 in.) in a rectangular plastic stamp pad of the same size. Deionized water was placed into the stamp pad until no water ran off when turned upside down. To sample, the field personnel used a LWW sampler, with a 150-cm^2 template, and a prepared sample pack, containing three rectangular pieces of Nucleopore 50 mm × 55 mm PE Drain Disc preweighed filter media, wrapped in aluminum foil. The filter was placed under the small block, and then pushed through the top opening of the frame block, and down onto the stamp pad to wet the filter media. The procedure also locked the small block into the frame block (see Figure 1). The block set was then placed into the template, with the filter media touching the sampling surface. Five passes, both forward and backward, were made with the block set touching the end of the template on every pass. The sampler was designed to minimize the pressure applied to the block, and to provide equal pressure distribution across the sampler block. After sampling, the block set was turned over, and the small block slid out of the frame block. The filter media was then placed back onto the aluminum foil, with sampling side facing up. The remaining two filter pads were then used in sequence with the above protocol. After the two final sample media were used, each was placed sampling side down on top of the first filter media in the aluminum

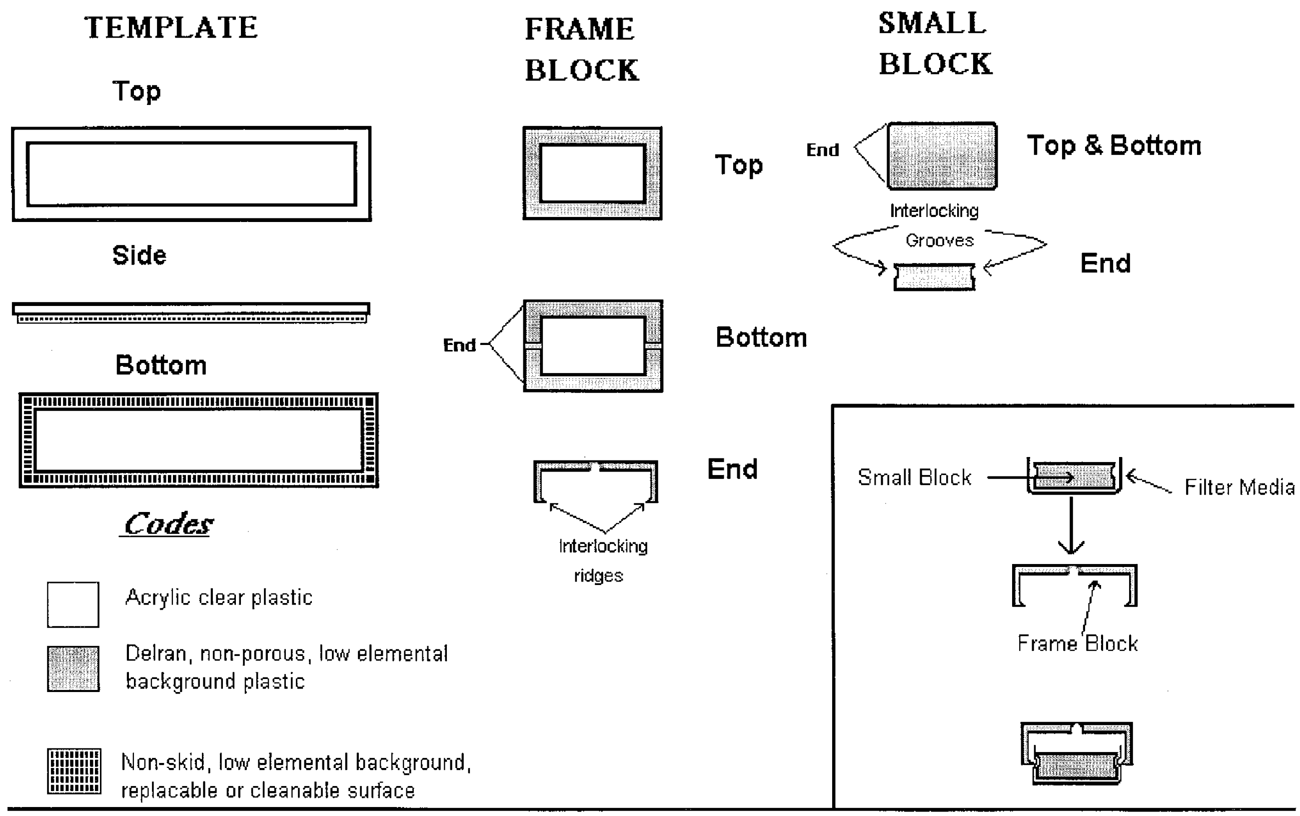

Figure 1. The revised LWW sampler.

foil, to minimize dust loss, and for transportation. For temporary storage, each sample pack was opened and placed in a modified fish tank, covered and sealed to facilitate drying of the filters. After drying, the samples were transported back to the laboratory and then stored in a humidity- and temperature-controlled environment before weighing.

Laboratory Analysis
HUD digestion and analysis was conducted by Azimuth, Inc. contracted with Housing Environmental Services (AIHA and ELLAP Laboratory ID # 9044) using flame atomic absorption spectrometry (Perkin-Elmer FAA), according to NIOSH 7082 analytical method (NIOSH, 1994). The minimum detection level was 25 μg/sample.

LWW digestion and analysis was conducted as follows. Filters were weighed, both pre- and post-sampling, in the humidity and temperature controlled facility. After weighing, the samples were digested in 19% spectrograde nitric acid in a laboratory microwave system (CEM MDS 200, Matthews, NC) using an EPA Soil Sample Protocol (U.S. EPA, 1995). Analysis was done using a flame atomic absorption spectrophotometer (FAA, Model # 3100), or inductively coupled plasma-mass spectroscopy (ICP-MS, Fisons Plasma Quad PQS). Calibration standards were traceable to the National Institute of Standards and Technology (NIST). Quality assurance checks were done with NIST reference materials numbers 981, 2710, and 2711. All analytical runs included both reagent and digestion blanks, as well as lead solution spikes. Ten percent of the ICP-MS samples, and 10% of the FAA samples were evaluated for system spike recovery. The ICP-MS detection limit was 0.2 μg/sample (1 ppb), and the FAA detection limit was approximately 10 μg/sample (0.5 ppm). If the sample's lead level (μg/sample) from the ICP-MS was near the top of the calibration curve, it was run again on FAA. The FAA value was reported. This ensured that all FAA samples were well above the minimum detection level. Acceptable instrument error was within ±20%, although most QC analyses were within ±10%.

Data Analysis
Overall, there were 706 HUD samples and 1268 LWW samples taken in the TLC trial. However, in order to compare the two methods, only those matched sample pairs ($n = 412$) containing both a HUD and a LWW were used in this analysis. None of the LWW reported values was below the ICP-MS minimum detection limits (0.2 μg/sample). By contrast, 32% of the 706 HUD samples

were below the FAA minimum detection limit (25 μg/sample). In the matched sample data set, 26% ($n = 107$ of 412) of the HUD samples were below the detection limit. Originally, the common approach of assigning one-half the minimum detection level (12.5 $\mu g/ft^2$) to each nondetectable HUD value (NCLSH, 1997) was used to compute a Pearson correlation coefficient for matched LWW–HUD pairs. This, however, appeared inappropriate for our analyses. It concentrates all of the nondetectable values at a single point, and violates the basic assumptions underlying the mixed model that we employed (Schlucter and Elashoff).

Because $\log_{10}$ of the lead loading had an approximate Gaussian distribution, it seemed more appropriate to apply a bootstrap technique to assign values to samples below the minimum detection level. Using the HUD data with the nondetectable values removed, the mean and the variance were estimated from the resulting truncated Gaussian distribution. A random number generator (SAS, 1996, RANNOR function) was then used to simulate the nondetectable values as if they had fallen in the lower tail of a Gaussian distribution with the specified mean and variance. The same planned analysis was conducted for each simulated data set, and the mean value of the parameter or test statistic was chosen with the corresponding p-value as the bootstrap estimate. Ten thousand bootstrap replications were used, so that the half-width of the bootstrap confidence interval for each p-value was at most 0.0098.

In order to compare the internal reliability of the two methods, a Fisher's z transformation (Rosner, 1986) was used to compare the correlation coefficients of replicate samples for HUD and LWW. The geometric mean lead loadings, as well as $\log_{10}$ mean lead loadings and standard errors were calculated. Paired t-tests were used to test for significant differences between pre- and post-cleaning LWW dust loadings and lead concentration levels.

For floors, three major predictors were of concern: room (kitchen or playroom), sampling method (HUD or LWW), and sampling time (pre- or post-cleaning). The two- and three-way interactions were also of interest. Sills were only measured in the playroom, so only time and method, and their two-way interaction were relevant. A mixed model (Schluchter and Elashoff, 1990) was then fit with the $\log_{10}$ of the lead loading as the outcome, and indicator variables of room, method, location, and time as the predictors (Analysis # 1).

A planned sequential backwards method was used to find the final model for hypothesis testing. For floors, the hypothesis of no effect of the three-way interaction between room, time and method on the lead loading level was tested. Testing was continued for the two-way interactions and the main effects. At each step in testing, the mixed model was reduced when there was no significant interaction, or main effect. Significant two-way interactions meant that we would examine the main effects within the levels of the other factor. If there were no two-way interactions, the single main effects would be tested. The two-way interaction between time and method was tested for sills. If this was nonsignificant, the main effects of sampling time and sampling method would be examined. The reduced mixed model had $\log_{10}$ of lead loading as the outcome, and sampling location, sampling method, and sampling time as the predictors (Analysis # 2).

To allow conversion of LWW results to HUD lead loading values, another mixed model was fit with $\log_{10}$ HUD as the outcome, and indicator variables of floor or sill, $\log_{10}$ LWW, and their interaction as predictors. Testing was done to determine if the linear relationship between the two methods was the same for floors and sills, by first testing the slopes (Analysis # 3). If the slopes were not significantly different, the model would be reduced, a common slope would be fit, and the intercepts would be compared (Analysis # 4). If the intercepts were different, this model would be used as the final model to describe the relationship and predict clearance values.

Results

The LWW and HUD geometric mean lead loadings are shown in Table 1, by location and time. The LWW sampler collected substantially less lead than the HUD method. Cleaning did not change the lead loading on floors to any large extent, but did make a reduction in window sill lead loading levels. This was true for both sampling methods. The correlation between matched LWW and HUD samples, calculated on the log values, was $r = 0.68$ ($p = 0.0001$, $n = 412$).

The LWW dust loading and lead concentration values which provide additional exposure metrics are shown in Table 2, by location and time. Significant reductions in dust loading were observed on the window sills ($p = 0.014$), which are consistent with the lead loading results. There was no significant change in lead concentration.

The correlations between replicate LWW samples and replicate HUD samples are shown in Table 3. The correlation coefficient was significantly higher ($p < 0.001$) for the LWW replicate samples ($r = 0.87$) than for the HUD replicate samples ($r = 0.71$).

Analysis # 1

For sills, the two-way interaction between time and method was nonsignificant ($F = 1.47$, ndf $= 1$, ddf $= 666$, $p = 0.23$). For floors, the three-way interaction between room, time, and method ($F = 0.075$, ndf $= 1$, ddf $= 666$, $p = 0.81$), the two-way interaction between room and method ($F = 0.61$, ndf $= 1$, ddf $= 666$, $p = 0.29$), the two-way

Table 1. LWW and HUD matched samples by pre- and post-cleaning.

	Floor lead loading ($n = 184$)			Window sill lead loading ($n = 78$)		
	Geometric mean ($\mu g/ft^2$)	Log_{10} mean	Log_{10} standard error	Geometric mean ($\mu g/ft^2$)	Log_{10} mean	Log_{10} standard error
Precleaning						
HUD	72.3	1.86	0.057	605.6	2.78	0.087
LWW	41.7	1.62	0.057	239.9	2.38	0.087
	Floor lead loading ($n = 113$)			Window sill lead loading ($n = 37$)		
	Geometric mean ($\mu g/ft^2$)	Log_{10} mean	Log_{10} standard error	Geometric mean ($\mu g/ft^2$)	Log_{10} mean	Log_{10} standard error
Post-cleaning						
HUD	63.9	1.81	0.072	170.3	2.23	0.127
LWW	34.7	1.54	0.63	123.0	2.09	0.84

Geometric means, $\log_{10}$ means, and standard errors of lead loadings by sample type.

interaction between time and room ($F = 2.33$, ndf = 1, ddf = 666, $p = 0.13$), and the two-way interaction between time and method ($F = 0.06$, ndf = 1, ddf = 666, $p = 0.84$) were all nonsignificant. There was no significant difference between kitchen floor lead loadings and play area floor lead loadings ($F = 1.89$, ndf = 1, ddf = 666, $p = 0.18$).

Analysis # 2

Because there was no difference in lead loading between kitchen and playroom floors, a model was constructed with $\log_{10}$ of lead loading levels as the outcome, and location (floor or sill), time, and method as predictor variables. For floors, the HUD lead loading values were significantly different than the LWW values ($F = 14.99$, ndf = 1, ddf = 670, $p = 0.001$). We failed to find a significant difference between pre- and post-cleaning lead loading values ($F = 1.20$, ndf = 1, ddf = 670, $p = 0.28$) for HUD or LWW. For window sills, there was a significant difference between the HUD and LWW sampling methods ($F = 6.27$, ndf = 1, ddf = 670, $p = 0.014$). Cleaning was more effective in reducing the lead loading on the window sills, as there was a significant difference in pre- and post-cleaning values ($F = 15.00$, ndf = 1, ddf = 670, $p = 0.0001$).

Analysis # 3

In order to determine if the linear relationship was the same for floors and sills, the slopes were compared. There was no significant difference between the slope of the floor equation and the slope of the sill equation ($F = 0.20$, ndf = 1, ddf = 262, $p = 0.70$). Since the slopes were not significantly different, a mixed model was fit with a common slope and separate intercepts. The difference in intercepts (floor vs. sill) was then tested.

Analysis # 4

Where FLOOR and SILL are the indicator variables for the sampling location, and LWW and HUD are the lead

Table 2. Geometric means, $\log_{10}$ means, and $\log_{10}$ standard deviations of dust loadings and lead concentrations from matched HUD–LWW data set.

	Dust loading (mg/ft^2)			Lead concentration ($\mu g/g$)		
	Geometric mean	Log_{10} mean	Log_{10} S.D.	Geometric mean	Log_{10} mean	Log_{10} S.D.
Floors						
Pre ($n = 184$)	599.2	2.78	0.46	0.076	−1.12	0.61
Post ($n = 113$)	431.0	2.63	0.44	0.086	−1.07	0.53
*Window sills**						
Pre ($n = 78$)	1071.5	3.03	0.45	0.230	−0.64	0.90
Post ($n = 37$)	588.8	2.77	0.42	0.219	−0.66	0.81

* In a paired t-test, only the difference in dust loading on window sills, between pre- and post-cleaning, was significantly different ($t = -2.604$, $n = 32$, $p = 0.0140$). All others were not significant.

Table 3. Reproducibility of LWW duplicates, HUD duplicates, and LWW–HUD matched pairs-correlation coefficients.

Lead loading	Pearson	*n*
HUD duplicates	0.71	127
LWW duplicates	0.87	141
LWW–HUD matched pairs	0.68	412

All coefficients are significant at $p = 0.0001$.

loading levels in $\mu g/ft^2$, the model (in exponentiated form) is:

$$HUD = 10^{[(B_0,F * FLOOR) + (B_0,S * SILL)]} * LWW^{B_1} \quad (1)$$

where $B_0,F = 0.877$ (s.e. = 0.072) is the intercept for floors, $B_0,S = 1.223$ (s.e. = 0.106) is the intercept for sills, and $B_1 = 0.605$ (s.e. = 0.039, $p < 0.01$) is the common slope. There was a significant difference ($F = 22.65$, ndf = 1, ddf = 263, $p < 0.0001$) of 9.2 $\mu g/ft^2$ between the intercepts for the floors and the sills. Using this equation, the LWW lead loadings that predict the HUD clearance values of 100 $\mu g/ft^2$ for floors and 500 $\mu g/ft^2$ for window sills, on these surfaces, were 72 $\mu g/ft^2$, and 275 $\mu g/ft^2$, respectively.

In order to assess the effect of the bootstrapping techniques, and the different detection limits of the FAA and the ICP-MS on the results of the mixed model analysis, a supplementary analysis was done. Those HUD samples from the original HUD data set ($n = 706$) that were below the detection limit (32%) were entered as one half (12.5 fg/ft^2) the minimum detection level (25 fg/ft^2). So as to treat the samplers similarly, we took the bottom 32% of the LWW samples and changed their lead loading value in the same manner, as if they had been reported as below the detection limit. This resulted in changing 32% of the LWW values (below 24.33 fg/ft^2) to 12.17 fg/ft^2. The Pearson correlation coefficients were then recalculated for both the HUD duplicates and the LWW duplicates, producing $r = 0.77$ and $r = 0.85$, respectively. While much closer, the coefficients were still significantly different ($p = 0.05$).

Discussion

Wipe samples provide a logical approach for estimating the extent of lead contamination on smooth surfaces. The HUD wipe method has been widely adopted because of its reasonable reproducibility and ease of use. Federal clearance standards are now defined in terms of this method.

The LWW sampler provides more detailed information about the lead exposure of a surface than the HUD method. While the HUD method only yields an estimate of lead per square meter of surface, the LWW sampler also provides an estimate of the amount of dust collected from the surface and the concentration of lead in the dust. The results presented above for the LWW sampler confirm that the reduction in lead loading associated with cleaning is related to the removal of dust with no effect on lead concentration. Thus, it appears that the use of wet mopping with TSP does not selectively remove lead particles. The LWW sampler was designed to improve the standardization of the sampling process by providing partial control of the amount of pressure that the technician can apply to the wiped surface. It is likely that the internal reproducibility achieved for lead loading, obtained from side-by-side samples, reflects this design feature.

The LWW sampler collects less lead from a given surface than does the HUD towelette. Applying manual pressure to the towelette probably leads to more efficient collection from pits and fissures in the surface than is obtained with the LWW sampler in which uniform pressure is applied to the collecting medium through a small block. The significantly higher intercept for the floors as compared to the smoother sills is consistent with this. The difference between the samplers may be greatest on uneven surfaces. The detergent on the towelette used in the HUD method may also contribute to lead and dust recovery. Although there is an overall difference in gross recovery, a relationship exists between the two sampling methods throughout the range of lead loadings represented in this study. Thus, the slope of the mixed model equation was the same for floor and sills (Analysis # 3).

The two samplers were affected similarly by field conditions as reflected by the absence of significant interaction terms with room or with pre- and post-cleaning status. It is not clear whether the extra lead recovered by the HUD method reflects contamination that is likely to contribute to a child's exposure. But in as much as it is recovered with an intentional wipe-up approach (including detergent), it may well collect lead that is not available to most children. Conversely, the LWW sampler may not recover all the lead on a pitted surface that is available to a child. The extra lead collected by the HUD method may be related to the detergent on the towelette, or the result of manual pressure applied on the sampling surface.

Because the LWW lead loading results are consistently lower than the HUD lead loading results, an LWW lead loading cannot be directly compared to published HUD post-abatement clearance standards. The mixed model analyses provide an opportunity to translate LWW lead loading values to HUD lead loading values (Analysis # 4, Eq. (1)), although additional studies are needed to examine this relationship on different types of surfaces. As established by HUD, the present post-abatement clearance standards are 100 and 500 $\mu g/ft^2$ for floors and window sills, respectively. The LWW values that predict these HUD values are 72 μg/ft^2 and 275 μg/ft^2, for floors and window sills, respectively.

Field sampling with the HUD method is usually supported by FAA analysis, resulting in a significant number of samples below the detection limit of the method. It is clear from the side-by-side samples analyzed here that the reproducibility of the LWW method supported by more sensitive laboratory techniques is better than the HUD–FAA combination. The supplementary analysis in which the bottom 32% of specimens done by both methods were assigned a value midway between zero and the 32nd percentile value suggests that some, but not all, of the better reproducibility of the LWW method are accounted for by the more sensitive analytic technique. A different study design would be needed to resolve this issue definitively.

In summary, both the LWW and HUD sampling methods have advantages. The HUD method is quicker to use in the field and does not require preweighed filter media. It is robust for rough and pitted surfaces since it does not matter if the dust wipe rips, provided it is still able to gather the dust from the surface. The LWW method provides information on dust loading and lead concentration in addition to lead loading. These additional data are useful in evaluating the specific effects of cleaning and the scope of improvement through a cleaning approach. The LWW provides lower estimates of lead loading than does the HUD method, which should be taken into account when interpreting clearance values.

Acknowledgments

The authors are grateful to the families who participated in the study and to Richard Madison of the Treatment of Lead Exposed Children Study (TLC), Newark, NJ. This study was partially funded by National Institutes of Health Grant # N01ES3563, and National Institutes of Environmental Health Sciences Research Center in Environmental Health Sciences (P30) Grant # ES05022-10.

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