

**Final Report:
Characterization of Hexavalent Chromium Concentrations
in Household Dust in Background Areas**

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A. INTRODUCTION

The current study was originally designed as an adjunct to the HCC study (Hudson County/Jersey City Chromium project, NJDEP study SR-06-027) in Jersey City, Hudson County, New Jersey to characterize hexavalent chromium [Cr(VI)] concentrations in house dust. That study was implemented to address the potential impact of Cr(VI) from chromate production residue (COPR) from capped and other chromium waste sites on current and future residential exposure. It was originally assumed that with the remediation of nearly all COPR sites in Jersey City and, given data from studies in the 1990's showing that following remediation of these sites, total Cr levels in house dust returned to background levels, that little or no Cr(VI) would be found in the house dust. However, Cr(VI) was, in fact, found nearly ubiquitously among the homes samples throughout Jersey City, albeit at low levels. This raised a question of the source of the Cr(VI) that was found in Jersey City homes. While the nature of the distribution of Cr(VI) with respect to the presence and absence of known historical COPR sites as well as the distribution of Cr(VI) within each individual house monitored in the HCC study suggested that the COPR was not the source of the Cr(VI) observed, this could not be definitely determined. This is because prior to this study, there were no known data on the occurrence of Cr(VI) in household dust in locations without specific known sources of Cr(VI) contamination. An alternative hypothesis was that Cr(VI) is ubiquitous in urban household dust, or perhaps in household dust in general. This could be the case if Cr(VI) originated from diffuse regional or long-range sources, and/or if Cr(VI) was present in household items and consumer products.

Therefore, this study was undertaken to address several questions:

- To what extent does Cr(VI) occur in household dust in locations with no known sources of Cr(VI) contamination (i.e., background locations)?
- How do levels of Cr(VI) in house dust in background locations compare to those previously found in Jersey City?
- Can the differences in the occurrence of Cr(VI) on different surfaces and different locations in background houses provide information on the extent to which Cr(VI) in house dust arises from within houses as opposed to outside ambient sources?

B. STUDY DESIGN

B.1. Site selection and subject recruitment

The study location of New Brunswick and surrounding areas were chosen to represent the background sites for Cr(VI) levels. Based on the chromium inventory data collected by NJDEP, there are no known nearby (< 1000 m) chromium waste sites or significant industrial chromium releases in New Brunswick areas. Study information was distributed to residents in New Brunswick and adjacent areas by word-of-mouth, local churches and schools. Residents who were interested in participating were contacted by telephone, and informed consent was obtained prior to sample collection. The consent form and a short questionnaire on the building characteristics of their home (age, building materials, etc.) developed for the Jersey City study were modified for this Cr(VI) study. A total of twenty subject houses as specified in the original proposal were recruited and house dust samples were collected. Among the 20 houses, 8 were recruited from New Brunswick, 8 from Highland Park, 2 from Somerset and 2 from North Brunswick. None of these municipalities has known chromium waste sites or nearby chromium

emission sources. As in the Jersey City project, different house types, were selected for sampling in the study (Table 1). However, homes recently constructed or repaired or remodeled (within one year) were excluded from the study due to the known presence of Cr(VI) is. In appreciation for their participation in the study a \$20 gift card were given to the resident upon completion of sampling at each home.

B.2. House dust sample collection

The sampling approach that has been developed in the Jersey City study was used in this background study. Two methods for sample collection were used throughout the study. The preferred method was the LWW sampler using pre-weighed polyester filters to wipe the surface with a template (150 cm²). The second collection method was a free hand wipe method. If LWW sampler could not be used (e.g., the template did not fit on the surface area or the rough texture of the surface may tear the filters), the dust on the surface was wiped by hand. The detailed method of sample collection can be found in the Jersey City final report (Lioy et al., 2008)

A minimum of three surface samples was collected in triplicate from each home (nine samples total). The surfaces sampled include a window well, an appropriate surface in a living area (e.g., living room, bedroom, etc.) and a surface in the basement (if available). A total of 185 dust samples were collected for this project, including three surface samples collected in triplicate from each home and 6 field blanks.

B.3. Sample analysis

Hexavalent chromium

Samples were analyzed by the identical techniques that were employed in Jersey City study. Dust samples were extracted with 5 mL pH = 4 nitric solution, sonicated at 60°C for 45 minutes, and then analyzed by IC/ICP-MS. A calibration curve was constructed from six levels of Cr(VI) and Cr(III) calibration standards (0.5, 1.0, 2.0, 5.0, 10, 25 ng/mL) for quantification. A mid level standard (5 ng/mL) was used for daily check of the instrument performance. If the variation of the response for the mid level standard (5 ng/mL) was greater than 20%, a new calibration curve was re-constructed before sample analysis. The analytical detection limit (ADL) was calculated as 3 times of the standard deviation of seven replicate injections of the lowest level standard, which is 0.038 ng.

Total chromium

Approximately 17% of the total Cr(VI) samples were measured for total chromium (N=10), including the samples with Cr(VI) concentration above 10 ppm. Total chromium was microwave-digested with 10 mL of 100% HNO₃, diluted up to 50 mL with DI-water, then determined by ICP-MS analysis. In every batch, one solvent blank and a standard reference material (SRM, certified particulate matter of NIST 1648) were concurrently analyzed to determine the recovery of total chromium. A calibration curve with 7 levels (0.0, 0.3, 0.5, 1.0, 3.0, 5.0, and 15.0 ng/mL) was generated for total Cr quantification. A 10 ng/mL standard (NIST AB, Calibrant A and B, 1811-001, 1811-005, High Purity Standards, Charleston, SC) was used for daily check of instrument performance. If the variability in chromium response is greater than 20%, the instrument was tuned and a new calibration curve was generated again before sample analysis.

Quantification

The concentrations of Cr(VI) and total Cr in solution (ng/mL) were first determined based on their response and corresponding calibration curves. The concentrations in dust mass ($\mu\text{g/g}$) were obtained by multiplying the volume of the extract and dividing by the dust mass collected in milligrams. The Cr(VI) loading was obtained through dividing the Cr(VI) mass, in nanograms, by the sample area in m^2 to obtain a final value of ng/m^2 . It is worth noting that the recovery of total Cr of the SRM samples was low, only $24\pm 3\%$ ($n = 3$). This recovery was similar to that measured in HCC study (Lioy et al., 2008) and previous studies (Kitsa et al., 1992). Since no correction for recovery was made previously, the total Cr concentrations measured in this study were not corrected, either, so the results obtained from this study can be directly compared to previous data.

QA/QC

All the solvents used for sample preparation and sample analysis were checked prior to field sample processing. Six field blanks (3.3% of the total dust samples) were collected throughout the study. An average of 1.23 nanogram of Cr(VI) was obtained from field blank samples. Sixteen (8.9% of the total dust samples) samples, collected side-by-side, were analyzed to report the method precision. The % difference (mean \pm SD) for the side-by-side samples is $36.8\pm 35.4\%$, with a range of 4.8 to 122%. It is worth noting that the distribution of chromium species in house dust samples may not be homogeneous, i.e., the side-by-side collected house dust samples are not equivalent to duplicate samples. The variability measured in the study was primarily contributed by the variability of the dust samples given the good analytical precision of the method ($< 15\%$). Similar sample variability in Cr(VI) concentration in house dust in the HCC study, with an average %difference of $36\pm 33\%$ for 50 paired side-by-side house dust samples (Lioy et al., 2008).

B.4. Data analysis

Statistical analyses, i.e., t-test and ANOVA, were conducted to examine whether there were differences in Cr(VI) concentrations measured in different sampling characteristics such as sampling town, location, and surface matrix as well as housing characteristics such as house age and material covered around the outside of house (e.g., grass, dirt, or mulch). If there were statistical differences ($p < 0.05$) within a group (≥ 3), Duncan's multiple range test was conducted to reveal which one was significantly different from the others. Because of the non-normality of most data (Shapiro-Wilk test; $p < 0.05$); all samples were log-transformed prior to any data analysis. The transformed datasets were approximately normally distributed ($p > 0.05$). Because the distribution of HCC data was not normally distributed, the non-parametric, Wilcoxon rank-sum test (i.e., Mann-Whitney test) was employed to compare the Cr(VI) levels in house dust between this Chromium Urban Background Study (CUBS) and the previous HCC (Hudson County/Jersey City Chromium project) study. For correlation analysis, Spearman correlation was conducted for all cases since the data is not normally distributed.

C. RESULTS

C.1. Cr(VI) concentrations/loadings in dust samples

The concentrations/loadings for all samples analyzed in the study are presented in Table 2 and 3, respectively. Cr(VI) was detected in all of 20 house dust samples collected from the urban background areas, ranging from 0.05 to $56.6\ \mu\text{g/g}$, with a mean (SD) concentration of $4.62(7.79)$

$\mu\text{g/g}$. The loadings were reported from 220 to 169,258 ng/m^2 , with a mean \pm SD of 10,003 \pm 27,886 ng/m^2 . The differences among four towns were not found significant for concentrations ($p=0.3495$) or loadings ($p=0.3441$). Also, the total number of samples in North Brunswick and Somerset were quite small (each $N=6$). Thus, the following analyses were conducted on pooled data, not by stratifying data by each town.

C.2. Determinants of Cr(VI) concentrations/loadings

House characteristics and sampling conditions, i.e. sampling surface material, sampling area within the home, age of house, and material around the outside of home (e.g., grass, dirt, and/or much), were analyzed to determine their influences on the measured chromium levels. These characteristics were selected for analysis because they had some reasonable likelihood of being related to Cr(VI) dust concentration.

For surface type, most were wood (43%), vinyl (22%), and laminate (13%), and the remaining surfaces (22%) included a variety of materials, i.e. concrete, cement, and aluminum. No significant differences in either Cr(VI) concentrations (ANOVA test; $p=0.1040$) or Cr(VI) loadings ($p=0.1288$) were found between surface material (Figure 1).

Within each home, we defined 3 different areas where the samples were collected: Living Areas (LA), Basements (BA), and Window Wells (WW). Living Area (LA) includes all locations in the house that are not assigned to basement (BA) or window wells (WW), for example, living room, bedroom, dining room, family room. Not all areas were sampled in each home (some homes did not have basements and window wells were sometimes inaccessible). Significant differences were found in Cr(VI) concentrations for the three different sampling areas within a home (ANOVA test; $p=0.0008$); however, the difference was not significant for Cr(VI) loadings ($p=0.2431$) (Figure 2). The multiple comparison test showed that the Cr(VI) concentrations in living areas (median of 3.67 $\mu\text{g/g}$; $N=25$) were significantly higher than the other two areas such as basement (median of 2.80 $\mu\text{g/g}$; $N=16$) and window wells (median of 1.48 $\mu\text{g/g}$; $N=19$) in the house.

Since significant differences were observed among the three sampling areas within the house, the potential effect of house age and surface material surrounding the house was examined on the Cr-VI levels stratified by sampling area within the home.

Correlation analysis was conducted to examine whether the house age was correlated with Cr(VI) levels measured in each sampling area within the home. House age was not significantly correlated with Cr(VI) concentrations/loadings measured in either sampling areas inside the house, except Cr(VI) loadings in living areas (Spearman correlation; $r=0.75758$; $p=0.0027$). To further pursue the influence of house age, we examined the correlation of house age and Cr(VI) in the living area stratified by surface type (e.g., wood, plastic, stone, and metal) using Spearman correlations. A significant correlation between Cr(VI) concentration and house age was found for wood surfaces in the living area. ($r=0.74545$; $p=0.0133$), which suggests that the Cr(VI) in house dust may partially come from wooden furniture and building materials that were treated with wood stains or preservatives containing chromium. Chromium was reported to be commonly used in wood stains between 1910 and 1970 (Ruetze et al., 1994). A preservative of CCA

(chromated copper arsenate) has been widely used for timber treatment since 1930's (Hingston et al., 2001).

For the material type covering the surface areas surrounding the house (e.g., grass, dirt, and/or mulch, etc.), ANOVA tests were conducted to examine whether the outside material type may affect the Cr(VI) levels measured in each sampling area within the home. There were no significant differences in Cr(VI) concentrations/loadings between different material type outside of the house ($p > 0.05$).

C.3. Comparison of Cr(VI) concentrations/loadings with the HCC study data

Cr(VI) concentrations/loadings obtained in this study (N=60) were compared to those obtained in the previous study in Jersey City (N=292) (Figure 3). A Wilcoxon rank-sum test showed the Cr(VI) concentrations in CUBS (median of 2.47 $\mu\text{g/g}$) was not significantly different ($p = 0.1084$) from the HCC concentrations (median of 2.065 $\mu\text{g/g}$). However, a significant difference was found for Cr(VI) loadings ($p = 0.0373$), with the CUBS loadings (median of 2,912 ng/m^2) were significantly higher than the HCC loadings (median of 1,982 ng/m^2). The location for sampling areas such as living area, basement, or window wells may make further differences in the Cr(VI) concentrations/loadings. Thus, the dataset was stratified into three categories for sampling areas within the home (i.e., basement, window wells, and living area). For the comparison of the Cr(VI) levels measured in basement (N=16 for CUBS and 36 for HCC), the differences were not significant for either concentrations or loadings ($p < 0.05$) between these two studies. For living areas, the Cr(VI) loadings were significantly higher ($p = 0.0229$) in CUBS (median of 3,460 ng/m^2 ; N=25) than in HCC (median of 1,982 ng/m^2 ; N=166). However, a significant difference was not observed in Cr(VI) concentrations ($p = 0.3569$). For window wells, the Cr(VI) concentrations in CUBS (median of 1.48 $\mu\text{g/g}$; N=19) were significantly higher ($p < .0001$) than in HCC (0.23 $\mu\text{g/g}$; N=90). The difference was not significant for Cr(VI) loadings ($p = 0.3542$).

C.4. Total Cr concentrations and % of Cr(VI) in total Cr

Total chromium concentration was measured in 17% (10 samples) out of total 60 samples, and the summary statistics are presented in Table 4. The average concentration of total Cr was 237 ± 131 $\mu\text{g/g}$, with a median value of 200 $\mu\text{g/g}$. The percentage of Cr(VI) in total Cr was estimated as the ratio of the Cr(VI) concentration at a sampling area within a house (e.g., living space) to the total Cr concentration at the same sampling area. However, since Cr(VI) and total Cr data used to generate the ratio in each case do not come from the same sample, there is an inherent uncertainty in the estimate of the overall ratio due to spatial variation in the ratio within a given location in a house. There is no reason to assume, however, that this uncertainty is biased. The mean ratio of Cr(VI) to total chromium was 8% with a range of 1 to 20%. A Wilcoxon rank-sum test was conducted for the ratios of Cr(VI)/total Cr between CUBS and HCC datasets ($p = 0.6895$), the result supported the assumption that the two ratios were not significantly different from each other.

C.5. Association of total Cr and hexavalent Cr in HCC & CUBS

The associations of the total Cr data and Cr(VI) in HCC and CUBS datasets were examined to explore the potential Cr(VI) sources in different locations. Total Cr consists of Cr(III) and Cr(VI). If the associations of total Cr and Cr(VI) were examined, there would be an auto-correlation due to the Cr(VI) present in both the total Cr sample and the corresponding Cr(VI) sample. Therefore,

the Cr(III) concentration was estimated from the total Cr by subtracting the corresponding Cr(VI) value. As discussed above, the total Cr value and the corresponding Cr(VI) value did not arise from the same sample, but from samples in the same area of the house. This introduces some uncertainty to this examination of association between Cr(III) and Cr(VI). However, as with the estimation of the ratio of Cr(VI) to total Cr, we have no reason to suspect that there is an inherent bias in this uncertainty. We assumed that if all of the Cr(III) and the Cr(VI) arose from the same source, they would be perfectly correlated. If a fraction of the Cr(III) and Cr(VI) arose from the same source and the remainder arose from different sources, the correlation would be smaller and the magnitude of the correlation (i.e., the r value) would reflect the extent to which they arose from a common source. If, however, the Cr(III) and Cr(VI) arose from completely unrelated sources, the r value would be small and the correlation would not be statistically significant (i.e., not significantly different from zero). In Jersey City, Cr in house dust that arose from COPR would contain both Cr(III) and Cr(VI). Therefore, if COPR contributed a significant amount of the Cr(VI) to the total Cr, we would hypothesize a significant correlation between Cr(VI and total Cr. On the other hand, we are not aware of any sources of total Cr other than COPR that would contribute both Cr(III) and Cr(VI) to house dust, therefore, in the background locations, we would hypothesize a non-significant correlation between Cr(VI) and total Cr. The scatter plots between trivalent and Cr(VI) measured in HCC and in CUBS are provided in Figure 4. Spearman correlations were provided in Table 5 for HCC and CUBS, as well as each location within the study. The visual inspection of scattered plots and statistical calculation of associations shows that trivalent Cr and, subsequently, total Cr concentrations were not closely related to hexavalent Cr concentrations for either Jersey City locations or background sites in NJ.

C.6. Summary and Recommendations

The results show low levels of Cr(VI) throughout the urban background study in New Brunswick and vicinities in NJ. Only one sample (with a concentration of 56.57 $\mu\text{g/g}$) out of the total 60 samples was reported above the NJDEP Cr(VI) soil remediation guideline of 20 ($\mu\text{g/g}$). This sample was collected in a basement in New Brunswick. Within four sampling areas in CUBS, the differences were not significant either in concentrations or loadings ($p < 0.05$). The Cr(VI) concentration/loading ranked from low to high levels as window well samples < basement samples < living area samples, which was also observed in HCC database.

The comparison between CUBS and HCC data shows that the Cr(VI) concentrations were not significantly different ($p < 0.05$), indicating the levels found throughout Jersey City were as low as urban background areas in the other areas of NJ investigated in the CUBS study. However, Cr(VI) loadings were significantly higher in the CUBS areas ($p = 0.0373$) than in Jersey City where there are potential outdoor chromium sources (e.g., COPR sites). The difference was primarily due to higher loadings observed in living spaces in participant's homes ($p = 0.0229$) in CUBS than in HCC. The reasons for the differences are not clear. However, it should be noted that loading depends, in part, on the amount of dust that is present on surfaces and so, Cr(VI) loading can vary independently of the strength of the source of the Cr(VI). Therefore, while loading can be predictive of exposure potential, it does not provide a strong indication of the strength of the source of Cr(VI). The urban background study observed that both Cr(VI) concentrations and loadings for samples collected on wood surfaces were higher than on other surfaces (see Figure 1) although the differences were not statistically significant. Spearman correlations between hexavalent Cr loadings and house ages were found to be significant in

living areas ($p=0.0027$) as well as on wood surfaces within living areas ($p=0.0133$). In HCC data, similar correlations were found between Cr(VI) loadings and house ages in living areas ($r=0.38532$; $p<.0001$) as well as on wood surfaces in living areas ($r=0.38796$; $p=0.0002$). The stronger associations were observed in CUBS ($r>0.75$), which is probably because more older houses were sampled in the CUBS than HCC. This indicates the potential indoor chromium sources may contribute to the increase of hexavalent Cr loadings in homes. In the past, the use of furniture stains or wood preservatives containing Cr(VI) appears to have been common and may be a significant Cr(VI) sources in the house as these materials degrade and mix with house dust over time.

Total Cr concentrations were measured in 10 samples (approximately 17% of total samples) and 17% of Cr(VI) in total Cr were estimated for the side-by-side collected samples. The total Cr levels reported in the study (237 ± 131 $\mu\text{g/g}$) were lower than those in Jersey City (771 ± 753 $\mu\text{g/g}$). The correlations between Cr(III) and Cr(VI) in both CUBS and HCC provided no indication that total Cr (or trivalent Cr) and hexavalent Cr have the same source. They appear to occur independently each other, probably from different sources. This is the opposite of what would be expected if COPR were a significant source of Cr(VI) in Jersey City. However, the analysis was conducted on very limited number of samples ($N=10$ for CUBS and $N=31$ for HCC), therefore, further studies are needed to verify the findings and investigate the potential sources that may contribute to the levels of hexavalent Cr in residential homes.

The study provides the valuable information of hexavalent Cr concentrations/loadings in house dust in urban background areas. The result shows that the hexavalent Cr loadings or possibly concentrations in household dust may be linked with wooden materials. The association of total (or trivalent) Cr and hexavalent Cr will be clearer when more paired total and hexavalent Cr data are available.

D. References

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APPENDIX

Table 1. Housing characteristics

Characteristics	N	Don't know
House type		1
Single family	14	
Town/row house	2	
Multi-unit	3	
House age reported		8
Min	5 years	
Q1	46 years	
Median	60 years	
Q3	90 years	
Max	100 years	
Yard material		
Grass	19	
Dirt	4	
Mulch	7	
Have a basement	17	
Home with inside smoker	2	
Any renovation		
Add a room	0	
Put up/take down wall	1	
Replace window	0	
Refinish floor	0	
Exterior painting	0	
Interior painting	3	
Heating system		
Hot water	13	
Forced air	6	
Electric	1	
Air conditioning		
Central	8	
Window	12	

Table 2. Cr(VI) concentration (ppm or µg/g) by geographical location in the study

Town	N	Mean	SD	CV	Med	P5	P95	Min	Max
Highland Park	24	4.85	4.64	96%	2.89	0.42	15.0	0.16	18.4
New Brunswick	24	4.67	11.3	241%	2.27	0.34	10.0	0.05	56.6
North Brunswick	6	3.86	4.13	107%	2.17	0.76	11.4	0.76	11.4
Somerset	6	4.25	3.81	90%	2.62	1.81	11.8	1.81	11.8
All	60	4.62	7.79	169%	2.47	0.35	13.4	0.05	56.6

Note: No duplicate or blank samples. Same for all tables in below.

Table 3. Cr(VI) loading (ng/m²) by geographical location in the study

Town	N	Mean	SD	CV	Med	P5	P95	Min	Max
Highland Park	24	12,698	34,445	271%	4,138	595	44,083	318	169,258
New Brunswick	24	10,511	27,786	264%	2,681	495	22,143	252	138,115
North Brunswick	6	2,018	1,831	91%	1,255	220	4,450	220	4,450
Somerset	6	5,173	4,787	93%	3,544	1,042	13,334	1,042	13,334
All	60	10,003	27,886	279%	2,912	407	33,113	220	169,258

Table 4. Ratio of Cr(VI) to total Cr (ppm or µg/g)

Analyte	N	Mean	SD	CV	Med	P5	P95	Min	Max
Total Cr ^a	10	237	131	55%	200	89	444	54	515
%Cr(VI) of the total Cr estimated ^b	10	8%	6%	74%	6%	2%	17%	1%	20%
%Cr(VI) of the total Cr by HCC study ^c	31	12%	11%	94%	9%	1%	33%	0.3%	51%

Note: ^aThe recovery determined by SRM is 24±3% (N=3) and no correction for the recovery.

^bEstimates were based on replicate samples on the same sampling location, not from the identical sample.

^cObtained from the Final Report for Exposure and Health Effects in Hudson County: Phase I

Table 5. Spearman correlations for trivalent and hexavalent chromium in HCC and CUBS

Study	N	Spearman Correlation (p value)	Location	N	Spearman Correlation
HCC	31	-0.25025 (0.1745)	DP	2	NA
			Freedom	8	0.20360 (0.6287)
			Garfield	5	0.10000 (0.8729)
			Lafayette	4	-0.80000 (0.2000)
			Other	8	0.02381 (0.9554)
			SH	4	0.40000 (0.6000)
CUBS	10	0.41818 (0.2291)	Highland park	5	0.60000 (0.2848)
			New Brunswick	3	0.50000 (0.6667)
			North Brunswick	1	NA
			Somerset	1	NA

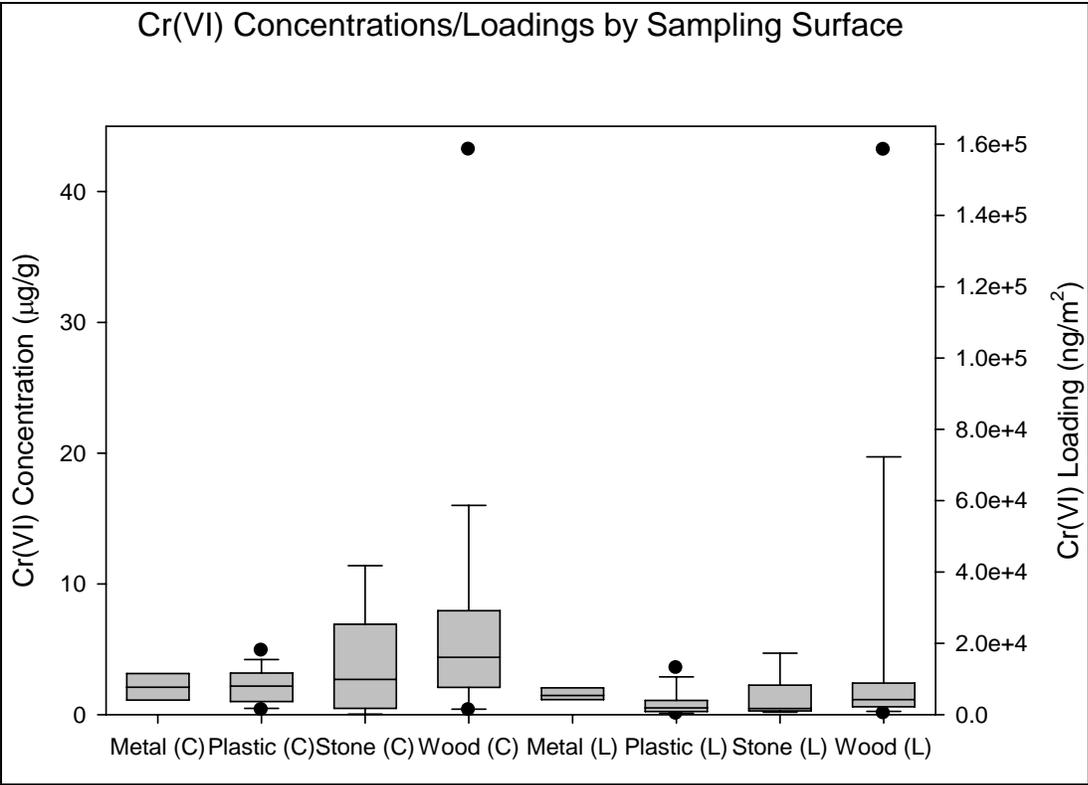


Figure 1. Boxplot of Cr(VI) concentrations/loadings by sampling surface

Note: (C) denotes the hexavalent Cr concentration (µg/g) and (L) denotes the loading (ng/m²)

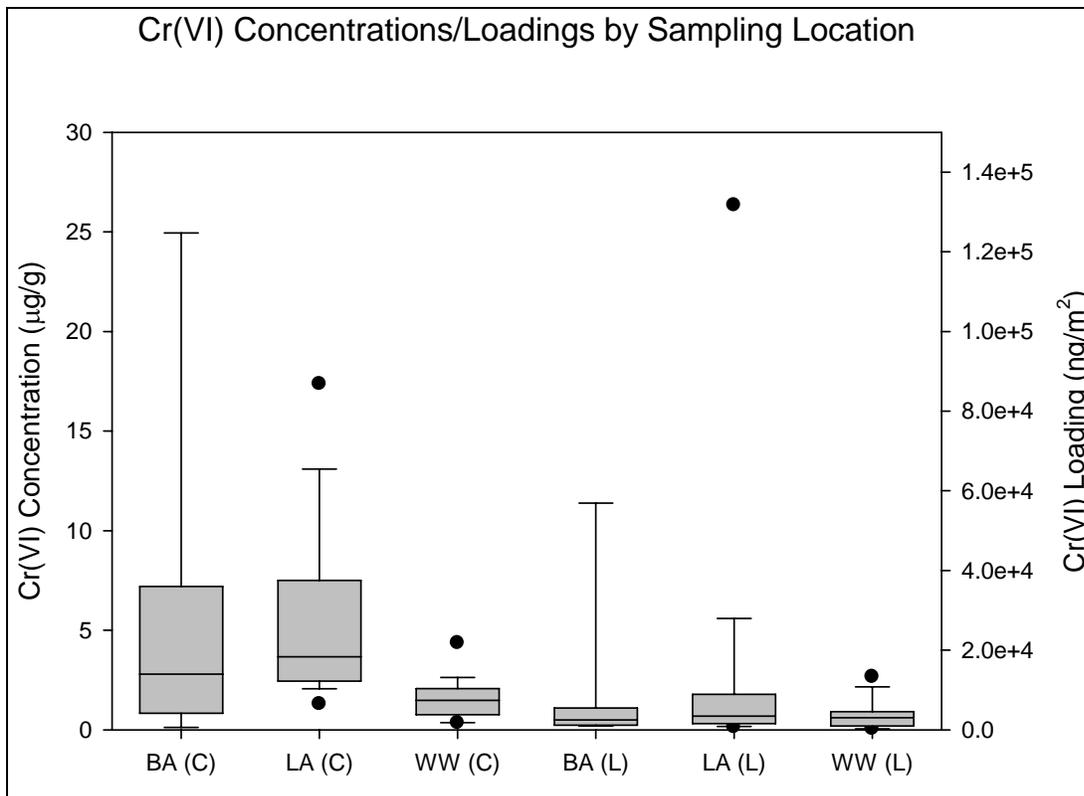


Figure 2. Boxplot of Cr(VI) concentrations/loadings by sampling location

Note: BA means a sampling location of basement, LA and WW mean the locations of living area and window wells, respectively.

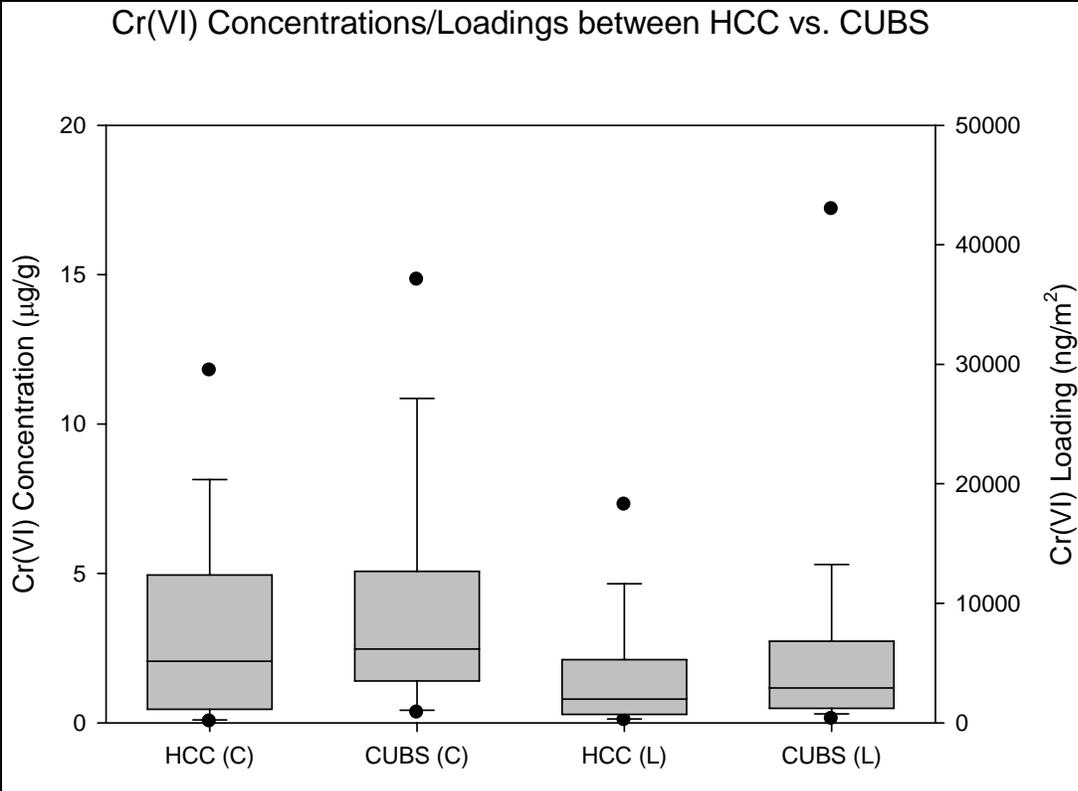
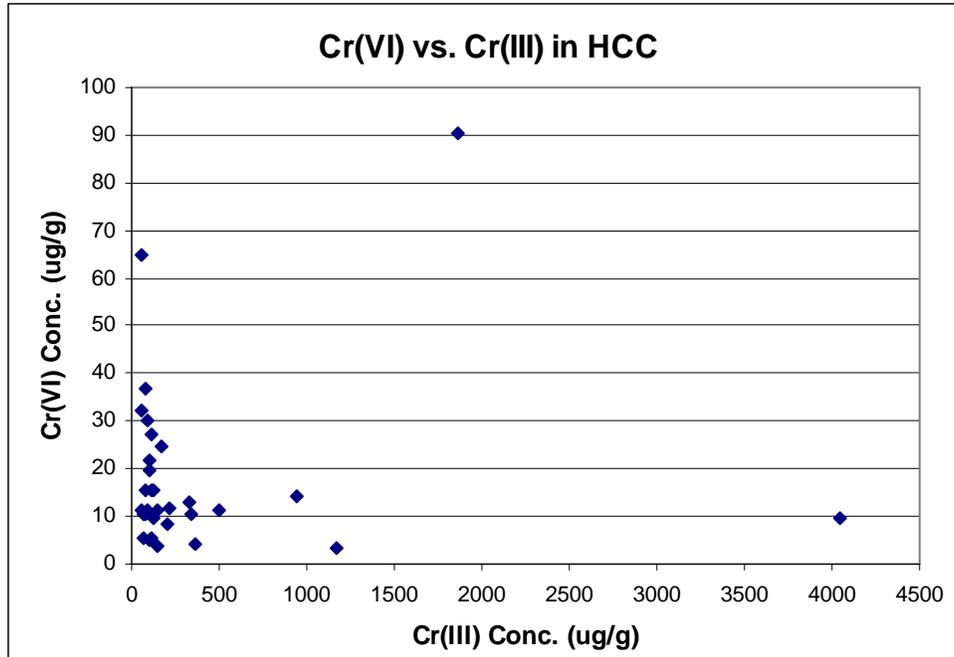
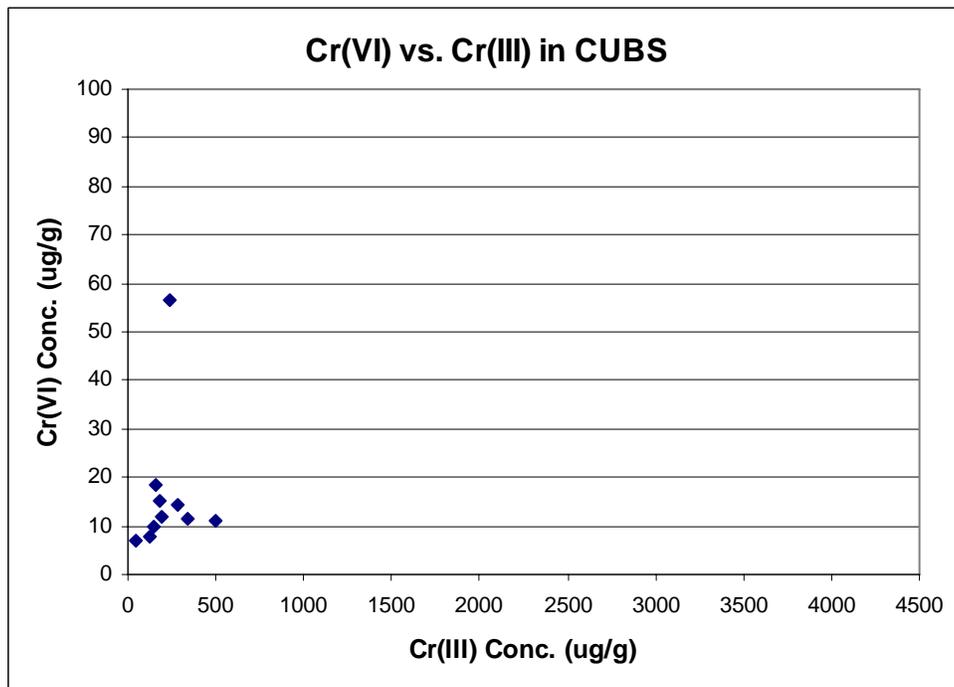


Figure 3. Boxplot of Cr(VI) concentrations/loadings between HCC vs. CUBS



(a)



(b)

Figure 4. Scatter plots for Cr(VI) vs. Cr(III) in HCC (a) and CUBS (b)