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### PIXE Analysis of Heavy Metals in Soil along the East River

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# PIXE Analysis of Lead Content in Urban Soil



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In partial fulfillment of the requirements for the degree of

*Bachelor of Science in Physics*

2022



# Abstract

We collected samples of soil from along the East River in Queens, New York, near the Hell Gate Bridge, on the Astoria Park side of the bridge in 2019 and on the Randall's Island Park side in 2021. We performed proton-induced X-ray emission (PIXE) analysis on the samples and found that soil closer to the Hell Gate Bridge contained higher concentrations of heavy metals, specifically lead and zinc. Many of the soil samples contained lead concentrations greater than the EPA standard of 400 ppm. We also performed PIXE analysis on a sample of the paint used on the bridge and found similar enhancements of lead and zinc. Our results indicate that the paint used on the Hell Gate Bridge is the cause of enhancements of heavy metals in the surrounding soil.

Keywords: PIXE; urban soil; heavy metals; soil contaminants

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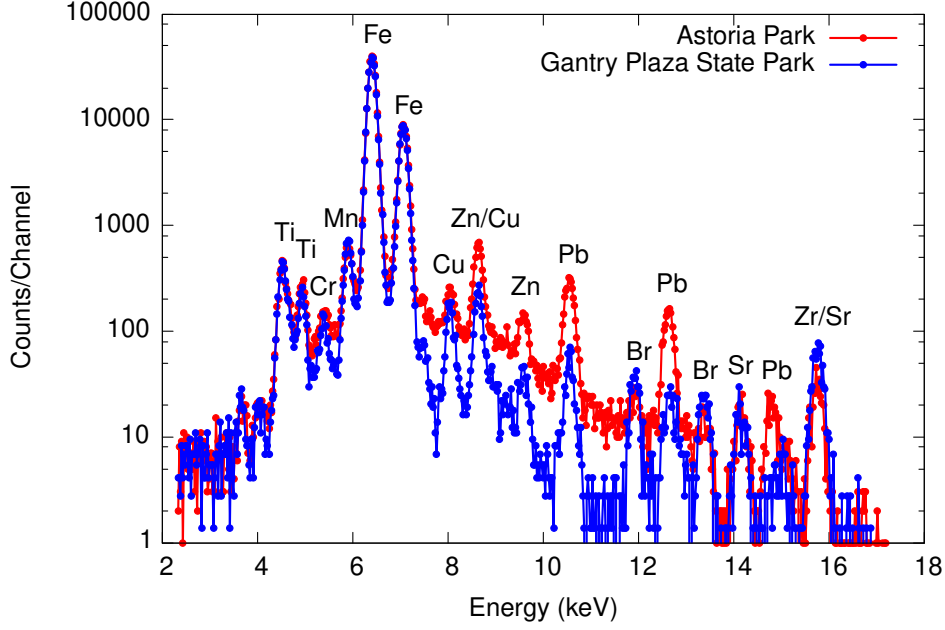
# Chapter 1

## Introduction

My research was inspired by the analysis done by Sajju Chalise in 2017 [1], in which the soil collected from directly under the Hell Gate Bridge in Astoria Park was found to contain an order-of-magnitude higher level of lead than the soil collected from about 5080 m away in Gantry State Park, as shown in Figure 1.1. This high lead content may be due to the fact that lead-based paint was used on Hell Gate Bridge when it was constructed in 1916, and this paint has now contaminated the soil due to paint chips falling off the bridge and mixing with the soil.

Many have thought that leaded gasoline could be pointed to as a cause of these high lead levels, but previous research done on sediment from the core of Central Park Lake points to solid waste incineration as another major cause of urban lead pollution. In 1999, Chillrud et al. collected sediment cores and found that higher concentrations of excess Pb-210 and Cs-137 were present in cores deposited in the 1930s through the 1960s, when solid waste incineration was prominent. Studying the concentrations of vanadium, an element used often in Venezuelan fuel, also uncovered that the decrease in leaded gasoline in the 1970's and 1980's was not the cause of lower lead levels, as it was actually tied to the decrease in Venezuelan fuel imports in 1966 [2]. In this paper, I hope to show that although lead levels due to solid waste incineration have dropped, lead paint is still a viable cause of high lead levels and should be looked at with more concern.

Although 2011 research suggested that bioavailability of lead and other heavy metals is generally low [3], 2012 and 2013 research by Brokbartold et al. showed that  $Pb_3O_4$ ,



**Figure 1.1** PIXE spectra of soil collected from Astoria Park compared to soil from Gantry Plaza State Park [1].

also known as red lead, a compound found specifically in lead anti-corrosion paint such as those used on bridges, was an exception and that its bioavailability and mobility in soil had been greatly underestimated [4, 5]. It has been determined previously that dust and chips from lead paint can easily contaminate adjacent bare soil [6], and that lead and zinc have been found to contaminate water sources due to their use in paint used on a bridge [7]. We believe that the paint used on the Hell Gate Bridge has contaminated nearby soil with lead and zinc in the same way.

In 1989, Madhavan et al. recommended that lead concentration in soil should be no higher than 200 ppm in areas frequented by children 5 years and under and no higher than 600 ppm in areas frequented by children 12 years and under [8]. In December 2000, the USEPA issued a federal guideline that the level of lead in soil should be no higher than 400 ppm [9]. This experiment suggests that much of the soil surrounding the Hell Gate Bridge is much higher than this level, which could lead to many adverse health effects, especially as young children are expected to frequent Astoria Park and Randall's Island. Lead levels in soil have been shown to correlate with levels of lead in children's blood, due to the ability of soil particles to be inhaled as dust or ingested via hand-to-mouth pathways. In New Orleans, it was found that Pb dust in topsoil resulting from

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smelting, industrial discharges, leaded gasoline emissions, leaded paint, and incineration were linked to multiple health adversities [10]. In adults, 20–70 percent of ingested lead and nearly all inhaled lead enters the blood, but children aged 9 months to 3 years are more vulnerable because they absorb lead at higher rates [11].

Lead entering the bloodstream can cause severe and irreversible effects in the development of children, specifically the brain. Its neurotoxic effects are well known and studied, specifically the impairment of the prefrontal cerebral cortex, hippocampus and cerebellum, which can cause problems such as attention deficits and memory impairment [11]. The negative correlation between environmental lead levels and intellectual performance is seen, for example, in New Orleans, where children living in communities with higher levels of lead in soil have the lowest school performance scores and, vice versa, children living in communities with lowest lead levels in soil have the highest school performance scores [10]. We believe that the levels of lead found in the soil of Astoria Park and Randall’s Island are cause for concern due to the park being frequented by children.



# Chapter 2

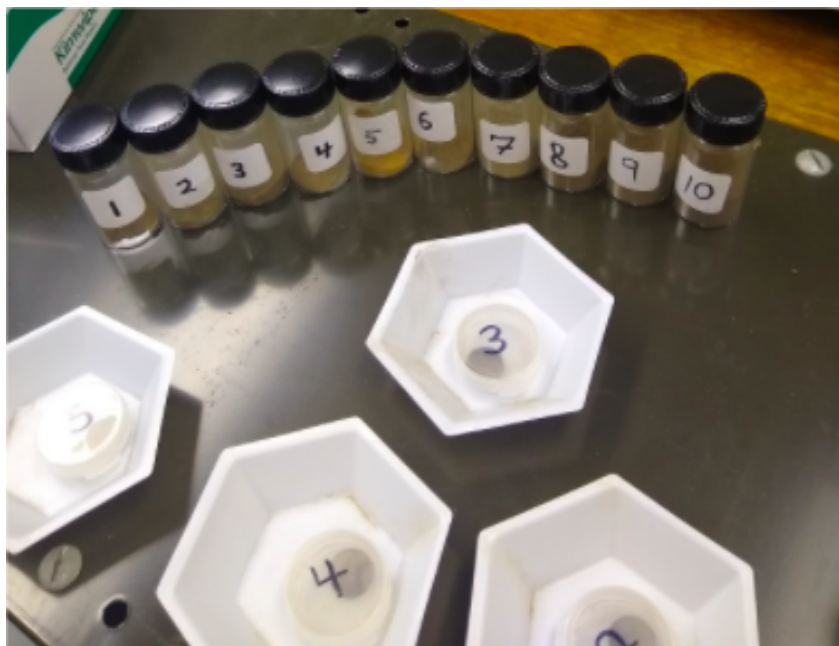
## Experimental Methods

### 2.1 Sample Collection and Preparation

On July 12, 2019, we collected our soil samples from along the East River and around Astoria State Park. In October 2021, we collected samples from Randall's Island, on the west side of the East River. Several paint chips were also collected and tested for enhancements of lead and zinc. In order to determine with greater precision and accuracy the distances between each collection site and the Hell Gate Bridge, we used GPS coordinates from Google Maps, which gave a much more precise location and allowed us to plot the lead and zinc concentrations as a function of distance from the bridge with greater accuracy.



**Figure 2.1** A photograph of the Hell Gate Bridge.

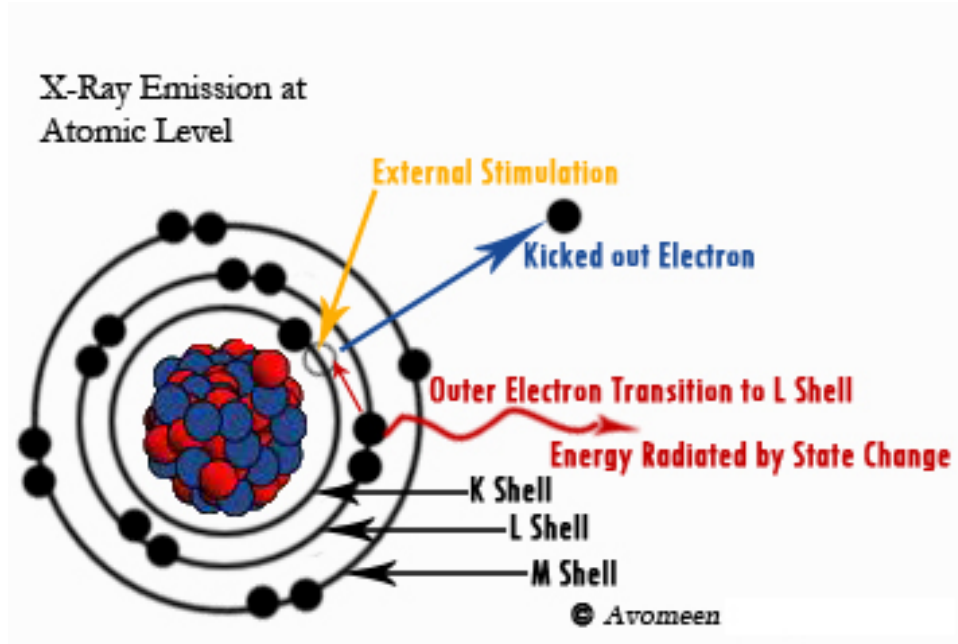


**Figure 2.2** Soil samples in jars and pellets prepared from these samples.

After collection, the soil samples were dried, then sifted and mechanically shaken for 24 hours to produce a fine powder. Pellets were created by hydraulically pressing 0.5 grams of soil with a few drops of polyvinyl alcohol. The pellets were then coated with a thin layer of Al in a vacuum evaporator. Figure 2.1 shows an image of the sifted soil samples in jars, as well as some of the pellets that were made from these samples.

## 2.2 PIXE Analysis

We used the Union College 1.1-MV Pelletron Accelerator to perform PIXE analysis on the collected samples. As shown in Figure 2.2, PIXE analysis works by bombarding a sample with a beam of protons, which interact with the sample by causing inner shell ionization, where the inner shells of atoms lose an electron. When this happens, an outer shell electron falls into the inner shell to take its place, releasing an X-ray with a certain amount of energy characteristic of the element. The amount of X-rays of a certain energy can then be used to determine the concentration of that element [12].



**Figure 2.3** A diagram showing how PIXE analysis is used to create X-ray emission from atoms.

## 2.3 PIXE Measurements

We fired beams of 2.2 MeV protons with currents of 5-15 nA, and diameters of 1-2 mm at the samples for 15-20 minutes each in 5 minute intervals. We then measured the emitted X-rays at an angle of  $135^\circ$  relative to the beam using an Amptek XR-100SDD silicon drift detector (SDD). We used a  $56\mu\text{m}$  thick Al filter in front of the detector for the Astoria Park samples, and a  $5\mu\text{m}$  thick Al filter for the Randall's Island samples. We estimated the accumulated charge by removing the sample from the beam and integrating the beam current in a Faraday cup for 5 minute intervals. The total charge incident on the samples ranged from 7 to  $12\mu\text{C}$  with an uncertainty of less than 10%. Also, a charge of  $1\mu\text{C}$  was collected on Ti, Fe, Cu, Ge, Au, and Pb micromatter standards (See section 3.2).

# Chapter 3

## Analysis

### 3.1 GUPIX

GUPIX for Windows, or GUPIXWIN, is a program used to analyze spectra from the PIXE data and calculate the concentrations of elements within the samples [13]. To calculate the concentration of a particular element, GUPIX uses the formula

$$C_Z = \frac{Y_Z}{Y_t H Q \epsilon T} \quad (3.1)$$

where  $Y_Z$  is the intensity of the principle X-ray line for element  $Z$  as determined from the fit to the spectrum,  $Y_t$  is the theoretical intensity determined in GUPIX,  $H$  is an experimental parameter determined by analyzing data from a set of standards,  $\epsilon$  is the detector efficiency,  $T$  is the coefficient for transmission through a filter or absorber between the detector and the target, and  $Q$  is the beam charge incident on the sample.

### 3.2 Calibrations

In order to determine the parameters used to obtain concentrations from GUPIX, we used thin standard targets to calibrate the detector system. These had known concentrations of titanium, germanium, gold, iron, copper, and lead.

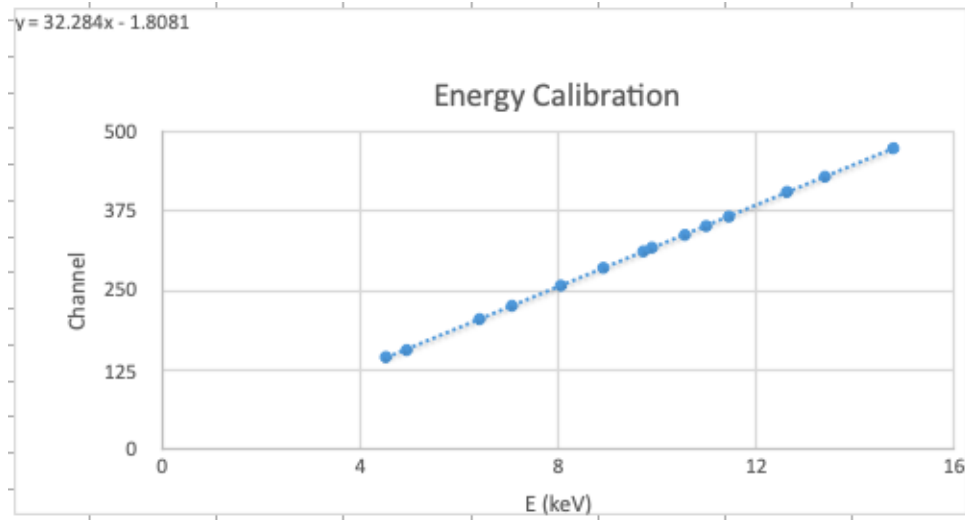
Using GUPIX, we picked out the centroids of the peaks, and matched these with the known energy levels of the transitions of each element, using a table that shows these

energies. We then plotted these and fit them with a line as shown in Figure 3.1 to determine constants A1 through A3 used in Gupix to calibrate the spectra. A5 is determined by

$$A5 = (4.6 \times 10^{-4} A2^2)$$

A4 is determined by using the FWHM of the Fe  $K_{\alpha}$  peak:

$$A4 = \left( \frac{FWHM}{2.354} \right)^2 - 6.404(A5)$$

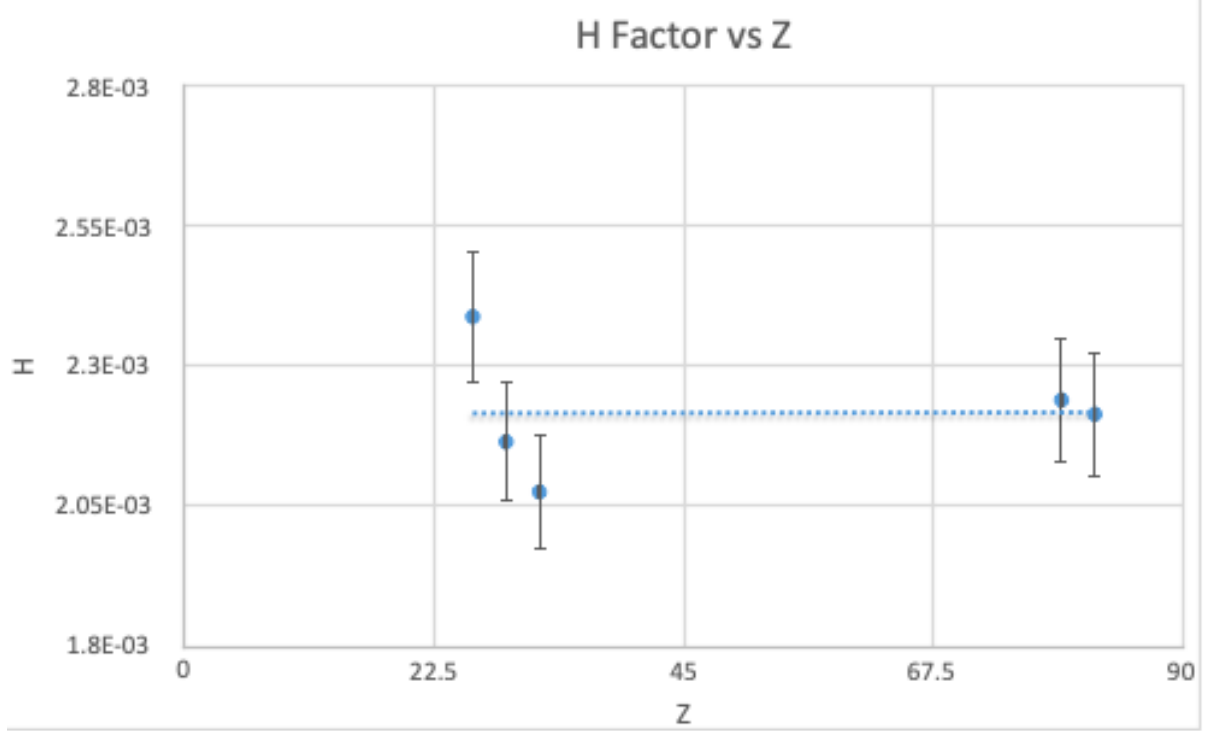


**Figure 3.1** A plot of the channel numbers determined by GUPIX versus the known energies of the peaks. The line is a fit to the data whose slope and y-intercept determine the energy calibration constants. In GUPIX these are A1, A2 and A3.

We then used GUPIX to measure the concentrations of each element in the standards. Using a spreadsheet, we recorded the actual concentration of the standards and the Gupix-measured values on the same table (Table 3.1). We divided the measured concentrations ( $C_m$ ) by the actual concentrations ( $C_s$ ), which gave us an H-value for every sample, each with a different Z and plotted these H-values versus Z. We adjusted the thickness value of our aluminum filter until the points on the plot fell into as horizontal of a line as possible (Figure 3.2). We then used the average H-value as our H-factor in Equation 3.1, and the thickness of the aluminum filter was entered in GUPIX at the value which gave us our H-factor when analyzing the soil. Our final H value was 0.002216 for the Astoria Park samples and 0.002371 for the Randall's Island samples.

Element	$Cs(ng/cm^2)$	$Cm(ng/cm^2)$	Z	H
Fe	51600	123.2	26	0.0023875
Cu	57100	123.6	29	0.0021646
Ge	45200	93.8	32	0.0020752
Au	45600	102.1	79	0.002239
Pb	53080	117.5	82	0.0022136

Table 3.1: A table of the known concentration values (Cs), GUPIX measured concentration values (Cm), H factor and Z for each standard. The average H value was 0.002216.



**Figure 3.2** A plot of the H-factor versus Z which is simply the measured concentration found with H set to 1, divided by the known concentration. The horizontal slope ensures that the H-factor is not dependent on Z.

# Chapter 4

## Results

Table 4.1 shows in detail the concentrations of heavy elements in each sample collected on the Astoria Park side of the bridge, and Table 4.2 shows these results for the Randall's Island Park side. Concentrations are reported only for elements that are present in each sample at a 95% confidence level. The quoted uncertainties were determined by adding in quadrature the estimated uncertainty in charge integration and the fit error reported by GUPIX.

All of the soil samples contained measurable concentrations of the metals Ti, Cr, Mn, Fe, Cu, and Zn, and all but one contained a measurable concentration of Pb. The ranges of the measured concentrations are:  $1378 \pm 30$  to  $9700 \pm 1100$  ppm for Ti;  $69.87 \pm 19$  to  $990 \pm 110$  ppm for Cr;  $194.7 \pm 8$  to  $1210 \pm 130$  ppm for Mn;  $9706 \pm 87$  to  $68709 \pm 137$  ppm for Fe;  $71 \pm 11$  to  $4952 \pm 117$  ppm for Cu;  $105 \pm 14$  to  $2819 \pm 64$  ppm for Zn; 0 to  $190 \pm 30$  ppm for Sr; 0 to  $1000 \pm 100$  ppm for Zr; and 0 to  $20483 \pm 295$  ppm for Pb.

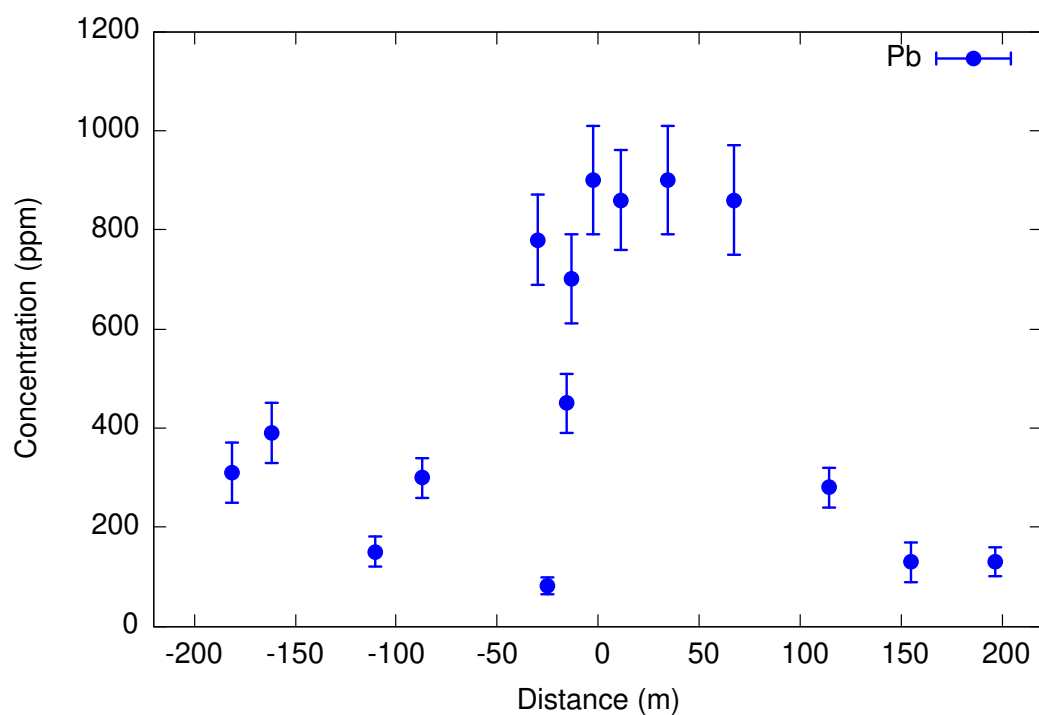
The results shown in Figures 4.1 and 4.2 show the lead and zinc concentrations in the Astoria Park soil plotted as a function of the distance from the bridge, with positive distances being north of the bridge and negative distances being south. Figures 4.3, 4.4 and 4.5 show the lead and zinc concentrations of the Randall's Island Park soil, plotted as a function of distance from the bridge. In this case, we were only able to access the south side of the bridge. The sample taken from directly under the bridge had a Pb concentration more than ten times any of the others, so a second plot was made to show the other data points more clearly. The data indicates higher concentrations of lead and

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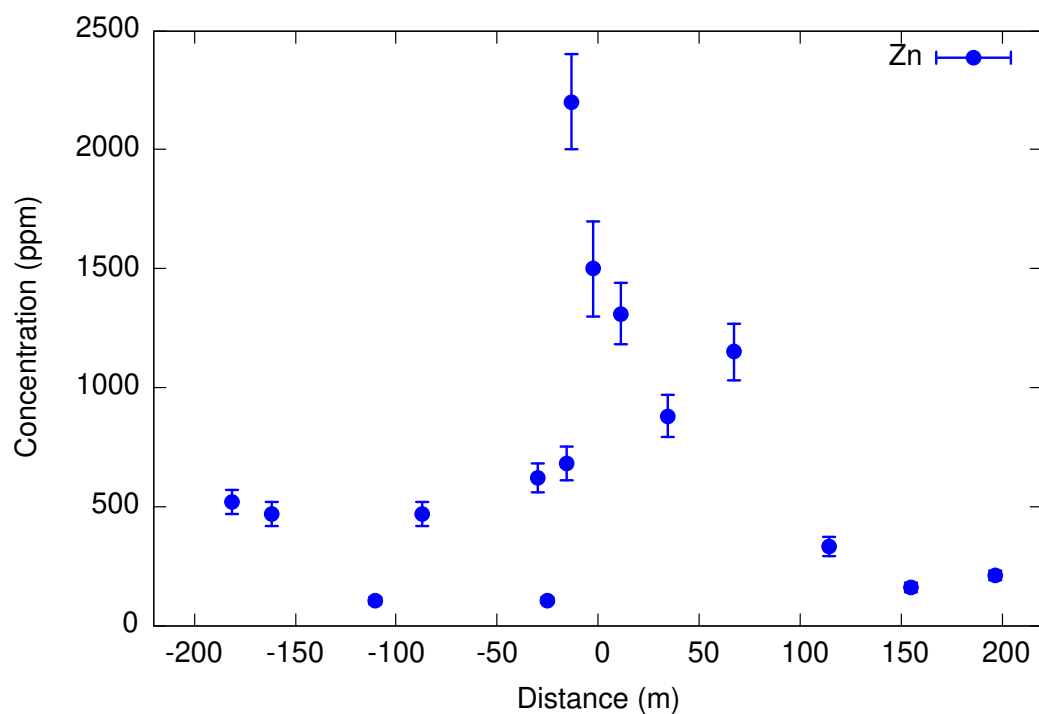
zinc in soil samples taken from distances closer to the Hell Gate Bridge, which supports the hypothesis that the enhancement of lead and zinc found in the soil is due to the paint used on the bridge.

A paint chip taken from the Hell Gate bridge was also analyzed, resulting in the spectrum shown in Figure 4.5. This sample was found to have a high concentration of lead as well as zinc, both metals which have been used in paint pigments and were found in high concentration in the soil.

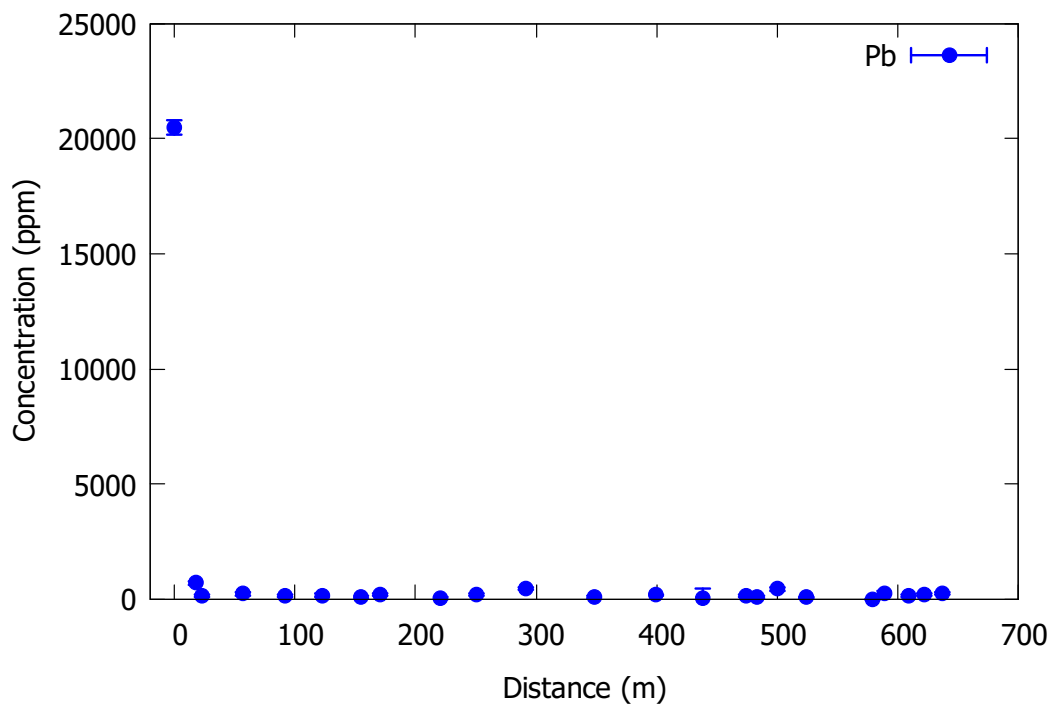




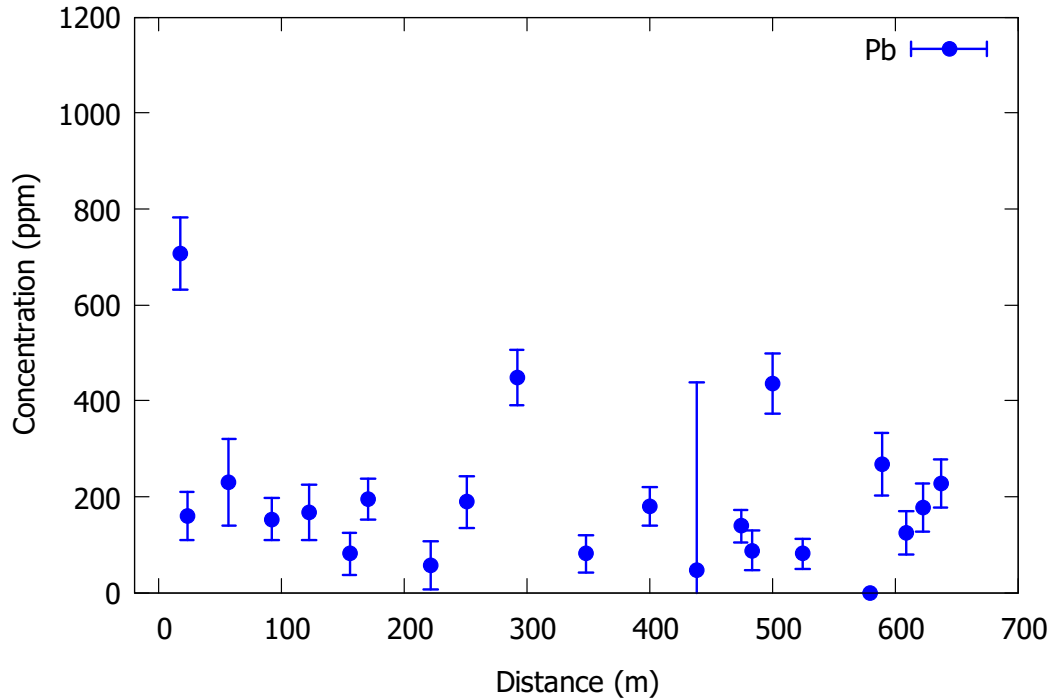
**Figure 4.1** Plot of the lead concentration vs distance from the Hell Gate Bridge on the Astoria Park side. The positive distances are north of the bridge, while the negative distances are south.



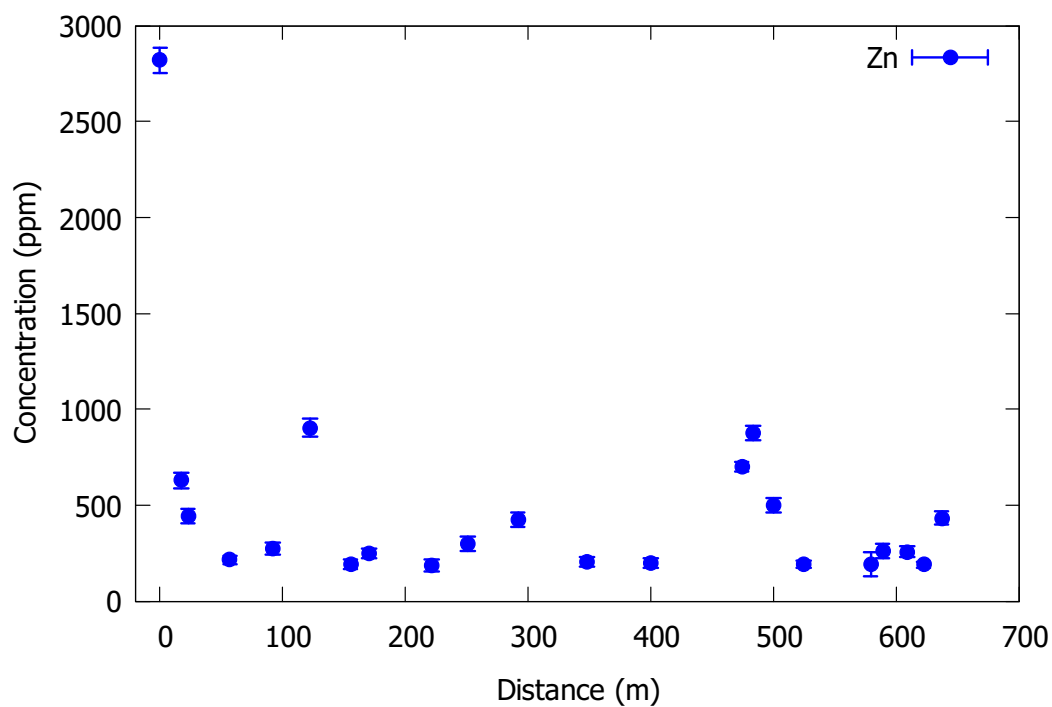
**Figure 4.2** Plot of zinc concentration vs distance from the Hell Gate Bridge on the Astoria Park side. The positive distances are north of the bridge, while the negative distances are south.



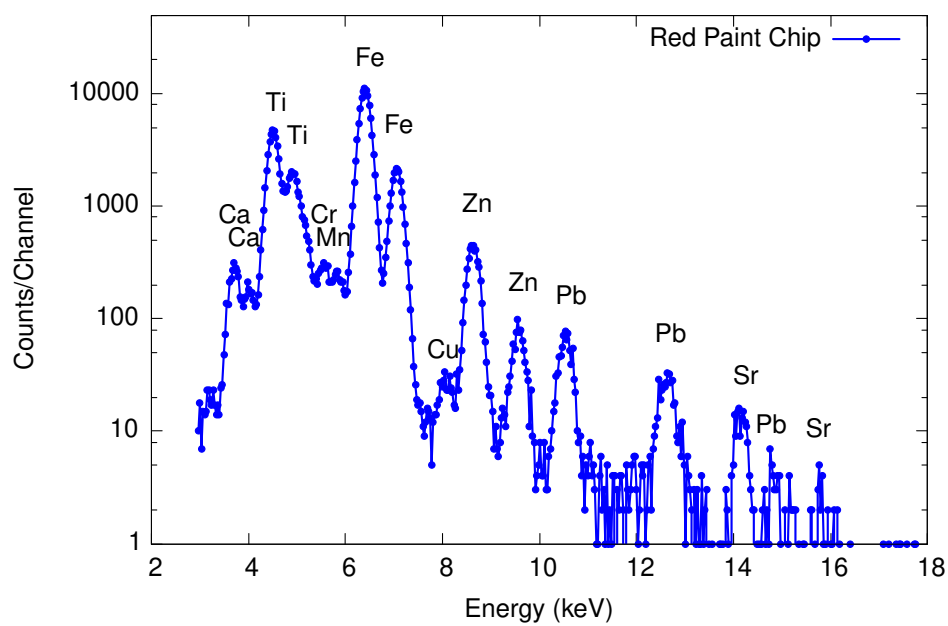
**Figure 4.3** Plot of the lead concentration vs distance from the Hell Gate Bridge on the Randall's Island Park side south of the bridge.



**Figure 4.4** Zoomed in plot of the lead concentration vs distance from the Hell Gate Bridge on the Randall's Island Park side south of the bridge, excluding the data point right under the bridge.



**Figure 4.5** Plot of zinc concentration vs distance from the Hell Gate Bridge on the Randall's Island Park side south of the bridge.



**Figure 4.6** PIXE spectrum taken from the paint chip.

Table 4.1: Concentrations of heavy elements measured in the soil samples collected in Astoria Park.

Sample #	GPS Coords.	Concentration in ppm								
		Ti	Cr	Mn	Fe	Cu	Zn	Sr	Zr	Pb
1	40.7816552 -73.9206039	7400±800	200±30	700±80	37000±4000	280±30	1310±130	190±30	400±60	860±100
2	40.7816219 -73.9207816	8200±900	820±90	800±100	54000±5000	790±80	1500±200	140±20	500±70	900±110
3	40.7816064 -73.9209418	7700±800	500±60	680±80	46000±5000	230±30	2200±200	110±20	830±100	700±90
4	40.7815376 -73.9211427	5800±600	220±30	390±50	24000±2000	114±15	620±60	57±13	370±50	780±90
5	40.7815755 -73.9211112	5700±600	420±50	510±60	24000±2000	82±11	106±13	78±14	510±60	82±17
6	40.7811294 -73.9215644	8000±800	750±80	1210±130	59000±6000	580±60	470±50	110±20	430±60	300±40
7	40.7810093 -73.9217857	5500±600	630±70	200±40	21000±2000	71±11	105±14	87±16	560±70	150±30
8	40.7806333 -73.9221555	6200±700	400±50	420±50	30000±3000	170±20	470±50	66±16	470±60	390±60
9	40.7805310 -73.9223513	5900±600	380±50	490±60	33000±3000	160±20	520±50	100±20	610±80	310±60
10	40.7814879 -73.9208335	7800±800	140±30	760±90	34000±3000	134±18	680±70	130±20	530±70	450±60
11	40.7819629 -73.9206143	6800±700	990±110	610±70	42000±4000	170±20	880±90	54±16	470±60	900±100
12	40.7821284 -73.9202877	9700±1100	530±60	660±80	52000±5000	220±30	1150±120	140±20	580±80	860±110
13	40.7824135 -73.9198736	6300±700	97±19	540±60	31000±3000	110±15	330±40	93±18	550±70	280±40
14	40.7827435 -73.9196349	4600±500	250±30	360±50	25000±3000	86±16	160±20	85±16	360±50	130±40
15	40.7829865 -73.9192608	4500±500	210±30	410±50	23000±2000	85±13	210±20	59±15	370±50	130±30
16	40.7834607 -73.9187387	5800±700	350±50	610±70	31000±3000	107±16	210±30	88±19	670±90	50±30
17	40.7857947 -73.9159492	5200±500	140±20	490±60	29000±3000	180±20	280±30	103±19	370±60	190±40
18	40.7815475 -73.9205348	6200±700	180±30	550±60	26000±3000	72±11	200±20	110±20	680±90	90±30
19	40.7807509 -73.9208801	6900±700	200±30	550±60	34000±3000	106±14	200±20	93±18	740±90	290±50
20	40.7807849 -73.9196668	6100±600	160±20	440±50	27000±3000	82±12	390±40	120±20	480±60	170±40
21	40.7806470 -73.9189674	6300±700	110±20	510±60	30000±3000	109±16	230±30	72±18	1000±100	290±50

Table 4.2: Concentrations of heavy elements measured in the soil samples collected in Randall's Island Park.

Sample #	GPS Coords.	Concentration in ppm								
		Ti	Cr	Mn	Fe	Cu	Zn	Sr	Zr	Pb
1	40.7836013 -73.9229964	4076±33	213.7±33	736±19	68709±137	710.3±56	2819±64	0	0	20483±295
2	40.7837567 -73.9229642	3868±74	253.5±12.6	559.8±20	24381±488	551.4±49	629.1±40	0	0	707.4±76
3	40.7835247 -73.9232459	4086±74	294.3±14	630.6±22	19647±391	1027±51	443.7±35	0	0	160.3±50
4	40.7834178 -73.9236254	2875±60	146.1±9	274.5±13	15393±334	230.4±31	215.4±24	0	0	230.5±90
5	40.7831352 -73.9238936	3543±67	206±12	420.8±17	19741±409	490.1±42	274.4±29	0	0	153.2±44
6	40.7827732 -73.9239509	4157±71	176±11	579±19	22848±404	4952±117	904±46	0	0	168±57
7	40.7827324 -73.9244371	2963±41	151.8±10	342.2±15	16795±235	210±34	190±25	0	0	81±43
8	40.7829479 -73.9248257	2799±54	628.3±17	322.6±14	17970±359	172.2±30	249.9±24	0	0	195±43
9	40.7827539 -73.9253688	2983±24	181.9±11	289.8±14	17814±68	189±40	187.5±30	0	0	58.09±50
10	40.7825955 -73.9256605	4616±84	323.1±16	457.2±20	24497±451	258.8±52	301.9±37	10	0	188.9±54
11	40.7822894 -73.9259985	3832±71	167.3±11	416.3±17	24631±475	497.3±46	425.5±35	0	0	448.5±58
12	40.7818133 -73.9263857	3177±23	173.3±9	328.7±13	18664±65	171.4±32	207.6±25	0	0	81.13±39
13	40.7816559 -73.9269869	1378±30	69.87±5	194.7±8	19714±371	242.9±32	199.8±23	116.2±134	0	179.8±41
14	40.7813185 -73.9272330	2898±56	168.2±9	346.2±14	18189±360	354±39	393.5±30	0	0	263.5±46
15	40.7811818 -73.9276260	1978±148	109.7±60	220.1±83	12911±452	160.2±22	700.7±26	0	0	139.7±34
16	40.7811640 -73.9277416	3482±37	237.5±10	403±13	24321±255	308.9±39	874.7±37	0	0	88.16±42
17	40.7812553 -73.9280591	3409±72	273.8±13	342.7±17	19647±438	224.8±40	499.3±38	0	0	436.8±63
18	40.7815861 -73.9286233	1761±21	245.5±10	267.8±12	9706±87	158.8±21	190.1±19	0	0	81.54±32
19	40.7817610 -73.9295490	4015±83	534.4±18	547.5±22	25151±526	270.4±53	263.1±38	0	0	268.2±65
20	40.7819309 -73.9295038	2549±40	601.8±35	281.6±38	16121±103	251.1±69	190.6±63	0	0	0
21	40.7820967 -73.9299443	2978±57	98.62±8	460±15	19571±374	123±32	358±29	0	0	125.1±46
22	40.7823130 -73.9301824	2978±60	291.4±12	444.6±17	21450±431	239.2±42	192.1±15	0	0	178.3±50
23	40.7823599 -73.9303765	4339±52	238.1±13	545.8±18	24603±298	285.8±43	434±35	0	0	226.9±50

# Chapter 5

## Discussion

All but two of the 44 total soil samples contained lead concentrations exceeding the New York State Department of Conservation (NYDEC) unrestricted use soil standard of 63 ppm [14] and 10 of them had levels above the EPA standard in play areas of 400 ppm [9]. The lead levels measured in 31 of the 44 samples fall within the range of 82 to 401 ppm reported for various locations in Astoria Park in a recent study conducted by the New York public radio station WNYC [15]. The concentrations of the other elements measured in this study fall within the ranges expected for soil in this region [16] and do not exhibit clear enhancements around the bridge.

Overall, the results shown in Figures 4.1, 4.3 and 4.4 indicate that much of the soil in both Astoria Park and Randall's Island contains levels of lead higher than the EPA recommended concentration of 400 ppm. Figures 4.2 and 4.5 show that soil near the bridge also contains high enhancements of zinc. This, along with Figure 4.6, which shows high enhancements of lead and zinc in the paint chip, supports our hypothesis that the high levels of heavy metals in the soil are most likely caused by the paint used on the bridge.

Several methods exist for preventing high levels of lead in soil from entering the bloodstream and causing health risks, including implementing physical barriers (i.e. covering contaminated soil with clean soil, mulch and/or gravel) and in situ soil amendments that can be mixed into the soil in order to immobilize the lead compounds and decrease their bioavailability [17]. Research on bioavailability of lead in soil has been done by Cheng et

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al., who found that the mobility of metals in soil is in the order of:  $\text{Zn/Cd} > \text{Pb} > \text{Cr/Ni}$ . Although the mobility levels of these metals were found to be low, maintaining a neutral to basic pH level is still necessary for keeping the levels as such [3]. Further research should be done on the possibilities of decreasing lead bioavailability in New York City.

# Chapter 6

## Conclusion

We collected 44 samples of soil taken from various points along the East River near the Hell Gate Bridge in Queens, New York. All of the soil samples contained measurable concentrations of the metals Ti, Cr, Mn, Fe, Cu, Zn, and all but one contained measurable concentrations of Pb. Ninety-five percent of the samples had Pb levels above the NYDEC unrestricted use soil standard and 22 percent of them exceeded the EPA standard for play areas. This work provides strong evidence, shown by the lead and zinc enhancements directly under the bridge, for the contamination of soil in Astoria Park and Randall's Island by metals, especially lead and zinc, used in the paint on the Hell Gate Bridge. The levels of lead in the soil immediately surrounding the bridge and in the nearby park are a cause for concern due to the area being frequented by children.

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