ENVIRONMENTAL LEAD EXPOSURE ASSESSMENT FOR CHILDREN FROM PRE-1970'S HOUSING IN ST. JOHN'S, NL

by

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ABSTRACT

Environmental media samples from residential properties throughout St. John's, NL were collected to examine the amount of lead contamination across housing age categories, as previous studies indicated that environmental lead levels on residential properties are above national guidelines, particularly on older properties. **Methods:** Environmental media samples were collected from properties participating in the study. 194 study households were sampled, representing 249 participants aged 6 months to 6 years. These included soil, indoor dust, indoor tap water, interior and exterior paint chips and garden produce samples. Statistical analyses examined the relationship between housing age category and strong-acid extractable lead levels in media samples, as well as, between media and existing data on children's blood lead levels. A bioaccessibility analysis was also undertaken for soil lead. **Results:** Significantly higher ($p < 0.0001$) near-total lead concentrations were found for pre-1970 homes compared to reference post-1980 homes for dust, soil, water post-stagnation and paint. Overall the lead content of household floor dust was found to be correlated with concentrations in paint chips and soil, and to be weakly correlated to children's blood lead levels ($p < 0.0001$, $r = 0.13$). There was also a weak relationship on correlation analysis for tap water stagnation levels 1,2 and 4 and blood lead levels, as well as for our housing age category of pre-1970. Results of a 1 M HNO3 bioaccessibility method had a strong positively correlation to near-total lead concentrations, and only weakly correlated to other soil properties of CEC content: Mg and K for dripline soil samples. **Conclusion:** There are elevated levels of lead in indoor dust, soils and tap water of residential properties throughout St. John's, which may represent an exposure risk to children's health.

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List of Abbreviations and Symbols

Agency for Toxic Substances and Disease Registry (ATSDR)

Blood Lead Level(s) (BLL)

Calcium (Ca)

Canada Mortgage and Housing Corporation (CMHC)

Canadian Council of Ministers of the Environment (CCME)

Cation Exchange Capacity (CEC)

Cation Exchange Capacity – Calcium (CEC-Ca)

Cation Exchange Capacity – Potassium (CEC-K)

Cation Exchange Capacity – Magnesium (CEC-Mg)

Centers for Disease Control and Prevention (CDC)

Confidence Interval (CI)

Dry weight (dw)

Environmental Protection Agency (EPA)

Geometric Mean (GM)

Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES)

Intelligence Quotient (IQ)

Loss on Ignition (LOI)

Maximum Acceptable Concentration (MAC)

Magnesium (Mg)

Sample number (n)

Not Applicable (NA)

Not Detectable (n.d)

Ontario Ministry of the Environment (OMEE)

ppm (parts per million)

Potassium (K)

Student – Newman – Keuls (SNK)

United States Department of Housing and Urban Development (HUD)

United States Environmental Protection Agency (US EPA)

Thesis Structure

This thesis is formatted using the traditional style of the School of Graduate Studies at Memorial University. Theses prepared in this format present the research as a whole. In this case, the thesis is broken down into chapters. It begins with a review of the literature on environmental lead sources and soil lead bioaccessibility, along with a brief overview of the health impacts of lead, particularly on children. Further chapters will address the methodology and results, with a discussion of those results.

The research for this thesis was gathered as part of a large biomonitoring study during 2010 involving multiple parts and funded partly by Health Canada. I was involved in the full scope of the project from the application for ethics to developing and researching the sampling protocols based on previous research and recommendations from groups throughout Canada and the United States, as well as the data collection in homes throughout St. John's. An in-depth analysis was completed by public health colleagues for the larger bio-monitoring study on the children's blood lead levels. The environmental analysis fell under the scope of this thesis.

Chapter 1: Introduction

Prior Research

St. John's, Newfoundland and Labrador, is one of the oldest English-founded settlements in North America (O'Neill, 2008). This record accompanies a long history of lead usage that has left a legacy of lead in the environment. While St. John's has never been a large industrial city with major point sources of lead contamination, its residential and small scale industry sources have left soil-lead levels in the downtown core of the city at much higher levels than comparable urban population centers in Canada (e.g. Victoria) and more similar to much larger North American cities such as New Orleans, Louisiana (Bell *et al*., 2010). Table 1.1 from a study by Campbell (2008) on the analysis of lead concentrations in St. John's compares measured lead levels in parts per million in some Canadian and American cities.

Table 1.1: Soil lead concentrations from cities in Canada and the United States of America. Adapted from Campbell, 2008.

City	Metric	Lead (ppm)	Sampling Method	Reference
Canada				
St. John's, NL	Median	266	Surface soil collected from open spaces, parks, schools and residences	Bell *et al.*, 2003
	Median	744	Surface soil collected around residences mainly in downtown	
Belledune, NB	Median	43-136	One composite sample per garden of 10 sub-samples collected at depths of 5-20 cm	Government of New Brunswick, 2005
Sydney, NS	Median	340	Top 5 cm of soil taken from middle of yard near Coke Oven site	Lambert and Lane, 2004
Victoria, BC	Median	90	Surface soil sampled from boulevards, parks and school yards	Bowman and Bobrowsky, 2003
Trail, BC	Geometric Mean	756	Top 2-3 cm of residential soil sampled	Hilts, 2003
Port Colborne, ON	Median	167	One composite collected from a minimum of 9 sub-samples taken from top 5-10 cm of residential soil	Ontario Ministry of the Environment, 2002
Ottawa, ON	Geometric Mean	42	One garden soil sample taken within 15 m of house	Rasmussen *et al.*, 2001
Iqaluit, NU	Median	13	Samples collected on commercial and residential properties	Peramaki and Decker, 2000
U.S.A.				
Syracuse	Geometric Mean	80	Top 10 cm sampled from sides of streets, parks and residences. Dripline avoided	Johnson and Bretsch, 2002
Chicago	Median	1773	One composite formed from 3-10 sub-samples collected on residential properties around the foundation and in play areas	Shinn *et al.*, 2000

Washington	Median	54-471	Residential topsoil collected at depths of 15 cm, 1 m from home	Elhelu *et al.*, 1995
New Orleans	Median	212	Top 2.5 cm of inner-city open spaces sampled.	Mielke, 1994
	Median	40	Mid-city open spaces	
	Median	28	Suburban open spaces	

A pilot study to investigate lead levels in St. John's soil was started in 2003 to test the hypothesis that the soils of St. John's 'may have become a large reservoir of potentially toxic metals' (Christopher *et al*., 1993), based on evidence of elevated concentrations of metals measured in urban lake sediments (Bell, 2003). This pilot study collected soil samples from a variety of land areas with potential child exposure, including schools, parks, residential properties and open spaces. The samples were collected from both the older downtown area and more recently developed areas of the city. The results of the pilot survey revealed that lead concentrations[1] in 60% of all the samples and 89% of residential samples exceeded the Canadian Council of Ministers of the Environment (CCME) residential soil lead guideline of 140 ppm (Bell, 2003).

Concerns over the moderate to high soil-lead levels and the potential health risk for children prompted a larger study of environmental lead exposure for children in St. John's (Bell *et al.*, 2010). This more systematic study found that 51% (n=1231) of soil samples exceeded the CCME soil guideline, 26% exceeded the United States Environmental Protection Agency (US EPA) guideline of 400 ppm for children's play

1 Unless otherwise specified, when concentration values for soil, dust, paint and garden produce are mentioned in this thesis, it should be assumed that a strong-acid extraction method has been applied to determine the lead concentration.

areas, and 9% exceeded the US EPA guideline of 1200 ppm for soil outside of play areas (Bell *et al*., 2010). The study also measured lead content in house dust of floors, windows sills and window troughs and found that 12% of house dust samples (n=96) exceeded the US EPA guideline for indoor dust (Bell *et al*., 2010) of 40 μg/ft^2 for floors, 250 μg/ft^2 for windowsills, and 400 μg/ft^2 for window troughs. In particular, the study by Bell *et al*. (2010) indicated that higher soil-lead levels were more prevalent in the downtown core of St. John's, the older housing area, and along the perimeter of these houses (dripline samples).

The results of these studies, in turn, led to a biomonitoring survey for environmental lead exposure to determine whether young children (6 months to 6 years) living in older housing in St. John's have an increased exposure to lead in the residential environment, termed the LeadNL Project (Bell *et al*., 2011). This thesis is a part of the larger biomonitoring survey presenting the environmental lead data. More than 250 households took part in the biomonitoring survey during the summer and fall of 2010, of which a total of 194 satisfied the study criteria outlined later on in this thesis. These participants were recruited through a two-stage cluster sampling strategy, using data gathered from the 2006 Canadian census in order to target specific neighbourhoods for canvassing. The biomonitoring study measured children's blood lead levels (BLLs) and lead in soil, indoor dust, indoor and outdoor paint, tap water and garden produce (where planted) in all households where biomonitoring took place. Recruitment of participants from across a selected series of housing age cohorts enabled comparisons of a target population living in older (pre-1970) housing to a reference population living in younger (post-1980)

housing, as well as more refined comparisons of children's lead exposure. The housing age cohorts were further broken down based on the year of construction (pre-1946, 1946-1960, 1961-1970, 1971-1980, 1981-1990, 1991-2000, 2001-2010) and were structured so that they matched neighbourhood-level census data available through Newfoundland and Labrador Community Accounts. The results of this LeadNL study demonstrated that while no children's BLLs in the study population were higher than the Canadian blood lead intervention level of 10 µg/dL. At the time of the study, the GM BLL for children living in pre-1946 housing was significantly higher than children living in houses built after 2000 and is summarized in the paper by Bell *et al*. (2011).

Objectives

This thesis focuses mainly on the analysis of residential environmental lead levels, but also analyzes the environmental lead levels in association with children's BLLs and between housing age cohorts, with the aim of improving our understanding of the local conditions that contribute to the overall lead burden on residential properties. The primary objective is to examine the pathways and exposure sources of environmental lead in different aged housing stock.

Two questions were addressed:

i) Is there a statistical association between children's blood lead levels and the levels of near-total lead in environmental media

ii) Are there statistically significant relationships between lead concentrations in environmental media and housing age

The sampling strategy included soil and indoor dust, as these media were shown to have elevated lead levels in the downtown portion of the city, but also other environmental media that may elevate children's BLLs, including tap water, paint and garden produce.

A secondary objective is to examine the bioaccessibility of soil lead in relation to housing age category and to two residential property areas; the dripline and children's play areas. Additionally, variations in bioaccessibility with soil properties were examined. The bioaccessibility of soil lead was examined because not all of the lead present in an environment medium is necessarily available for uptake by the body.

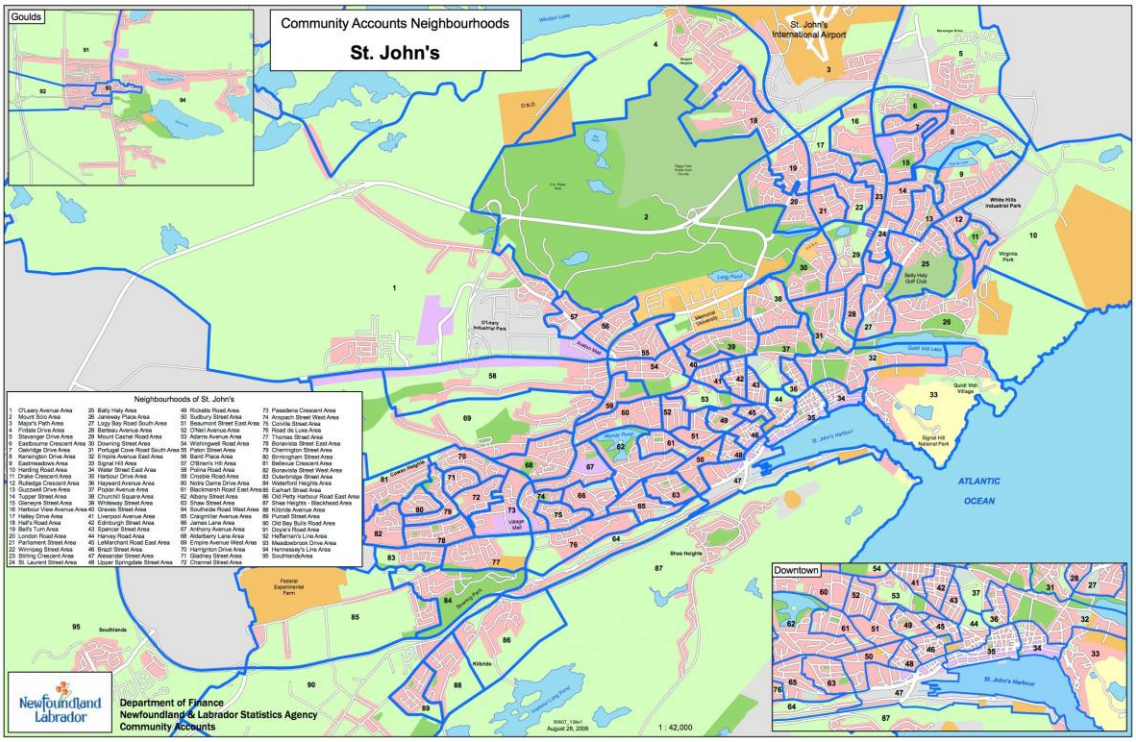

Figure 1.1: Map of St. John's, Newfoundland and Labrador located in Atlantic Canada. Downtown area highlighted in smaller box (communityaccounts.ca – accessed June 1, 2015)

The Health Effects of Lead

Lead can represent a significant health risk for humans and in particular young children depending on the amount of lead taken into the body. Lead can enter the body through three pathways: inhalation, ingestion or dermal absorption. Dermal absorption of inorganic lead compounds is minimal (OMEE, 1994), while inhalation and deposition of lead in the lungs represents about 25-45% of total intake for children (US EPA, 1998). Of particular concern for children are the smaller particles found in air in the urban environment, as they deposit in the lower respiratory tract where they can easily be absorbed into the bloodstream (US EPA, 1998). Absorption across the intestinal lining from the ingestion of lead represents another important exposure pathway for children, as they have a higher gastrointestinal absorption rate (Ziegler *et al*., 1978). Up to 50% of the lead may be absorbed compared to 10-15% in adults (US EPA, 1998). When lead enters the bloodstream, it is distributed throughout the body with some being excreted while other quantities are stored in the soft tissues such as the kidneys, bone marrow, liver and brain and, more permanently, in bone and teeth (US EPA, 1998). The half-life of lead is about 25 days in blood, 40 days in the soft tissues and 25 years in the bone (US EPA, 1998).

Children are most sensitive to the neurological impacts of lead because their nervous systems are not fully developed and it is easier for lead to cross the blood-brain barrier (Ziegler *et al.*, 1978). Children are also more likely to ingest lead due to their high frequency of hand-to-mouth behavior, which increases the likelihood that contaminated

soil or dust may be consumed (Moya *et al.*, 2004). The health effects experienced by children largely depends on the amount of lead ingested. The current Canadian blood lead intervention level set by Health Canada is 10 µg/dL (Health Canada, 2013). Health Canada also reports that although the intervention level is 10 µg/dL, there is sufficient evidence that blood lead levels below 5 µg/dL are associated with adverse health effects (Health Canada, 2016). In 2012, the Centers for Disease Control and Prevention (CDC) in the United States updated its recommendations for children's blood lead levels to 5 µg/dL being the new reference level (CDC, 2016, Paulson and Brown, 2019). This information was based on their National Health and Nutrition Examination Survey. Some of the general health effects of lead include problems with the developing nervous system, which can lead to mental and behavioural problems. Lead also affects the hematological and cardiovascular systems and the kidneys (ATSDR, 2005). The neurodevelopmental effects of elevated blood lead concentrations include reduced IQ scores (Needleman and Gastonis, 1990, Reyes, 2015, Shah *et al*. 2020). More recently there has been research to indicate that negative health effects may occur at BLLs below 10 µg/dL (Bellinger and Needleman, 2003, Reyes, 2015, Paulson and Brown, 2019, and Reuben *et al.,* 2019) and even at BLLs as low as 1-2 µg/dL (Health Canada, 2013). It is for these reasons that it is important to understand further the environmental levels of lead in St. John's, NL and the bioaccessibility of that lead.

Environmental Lead Sources

There are many potential sources of lead in the environment. Historically, most exposure to Canadians came from inhaled emissions from leaded gasoline combustion and

industrial activities (Health Canada, 2013), but exposure from deteriorated leaded paint has also been a concern. St. John's has a 500-year history of being used as a fishing port; however, permanent urban development only began about 250 years ago (Poole, 1994). St. John's has never been a highly industrialized city, so many of the environmental lead sources in St. John's have come from anthropogenic sources that include coal combustion, and the use of leaded gasoline and leaded paint. It has been hypothesized that the combustion of coal and leaded gasoline, in particular, have created a persistent legacy of lead in soils that may provide a source of lead intake for the general population through the ingestion of soil and dust (Christopher, 1999). For these reasons, it is important to understand the sources and pathways of lead exposure in St. John's, NL.

Indoor Dust

Lead from indoor dust can be from two main sources: tracking in of lead contaminated soil or the breakdown of lead contaminated paint (Tong and Lam, 2000). A study in Sydney, Nova Scotia found that dust lead loadings were higher in entryways than further inside the home leading the researchers to believe that most of the lead sources in the dust were exterior (Lambert and Lane, 2004). von Lindern *et al.* (2003a) and Layton and Beamer (2009) also noted that house dust lead levels were correlated with soil lead. Further research on leaded-house dust indicates that it may be a large contributor to children's blood lead levels (von Lindern *et al.,* 2003b; Lanphear *et al.,* 1998a and Yiin *et al.,* 2000, Gulson and Taylor, 2017, Safruk *et al.* 2017). Lanphear *et al.* (1998a) pooled 12 epidemiological studies to estimate the contributions of indoor dust and soil to children's blood lead levels and found that indoor dust was the strongest predictor. Dixon *et al.* (2009) found, for example, that floor dust samples were significant predictors for

children's blood lead levels, with 4.6% of children having a blood lead level over 10 μg/dL in homes constructed before 1978 where floors had a lead level greater than 12 μg/ft^2. Research from Flin Flon, MB and Creighton, SK by Safruk *et al.* (2017) found GM BLL's were associated with dust from kitchen floors. A systemic review by Frank *et al.* (2019) analyzed results for lead in dust from the United States over the period of 1996-2016 and found: for floors (n =535) it had a mean level of 13 μg/ft^2, and for windowsills (n=380) mean of 214 μg/ft^2, however, unfortunately there was insufficient data for a meta-analysis on window troughs.

The Canadian House Dust Study by Rasmussen *et al.* (2013) found that lead concentrations increase with housing age. The indoor dust guidelines at the time of analysis for this thesis (2012) from the US EPA were 40 μg/ft^2 for floors, 250 μg/ft^2 for windowsills, and 400 μg/ft^2 for window troughs (EPA, 2001). As of June 2019, the guidelines were changed to 10 μg/ft^2 for floors and 100 μg/ft^2 for windowsills (EPA, 2020). There are currently no guidelines in Canada for indoor dust. A recent study by Braun *et al.* (2021), with analysis of residential dust lead levels and childhood blood lead concentrations, found that an increase in floor dust lead from 10 to 40 μg/ft^2 was associated with 26% higher blood lead concentrations that were over 5 μg/dL.

Soil

In St. John's, the natural soil lead levels range from 15 to 139 ppm (Geological Survey of Newfoundland and Labrador, 2007). Anthropogenic sources of lead can raise background lead levels. Potential sources include combustion materials from leaded-gasoline and

point source industrial emitters, as well as the weathering of leaded paint. Once deposited in the soil, if undisturbed, lead can remain there in a relatively immobile form for decades, depending upon soil properties such as the pH and cation exchange capacity (Department for Environment, Food and Rural Affairs and the Environment Agency, 2002). Children playing in lead contaminated soils, particularly those above 1000 ppm, may have a 2 to 7 μg/dL increase in blood lead concentration (Lanphear *et al.,* 2000) as a result of breathing in small soil particles or from inadvertently ingesting soil on their hands (Taylor *et al.*, 2013, Zahran *et al.*, 2010). Lewin *et al.* (1999) and Safruk *et al.* (2017) found a similar relationship between soil and blood lead levels, with increased soil lead levels being associated with elevated blood lead levels in children. Mielke *et al.* (1997) predicted that reducing the soil lead levels in populated areas would reduce blood lead levels. The CCME soil-lead guideline for residential areas in Canada has been set at 140 ppm (CCME, 1999), there is currently no Canadian guidelines for play areas. The US EPA has soil-lead levels set at 400 ppm for children's play areas and 1200 ppm for soil outside of play areas (US EPA, 2011b).

Tap Water

Drinking water is another possible route through which children may be exposed to lead. In Canada at the time of data collection the Maximum Acceptable Concentration (MAC) for lead in drinking water was 0.010 mg/L (Health Canada, 2013). In March 2019 Health Canada decreased the MAC to 0.005 mg/L (Health Canada, 2019). This MAC is based on the level of lead measured in water leaving a drinking water treatment plant. However, as water moves through the distribution line its lead content can increase as a result of contact with lead pipes, connectors and lead solder in the system and in the household

(Health Canada, 2013). Older homes, specifically those built before 1950, represent a greater risk for lead contamination as lead service connectors were commonly used at that time (Health Canada, 2013), unless, however, they have undergone renovations to remove them. Many communities throughout Canada have older water distribution systems that still have lead soldering or pipes (Payne, 2008), as it was not until 1990 that the National Plumbing Code of Canada ruled that there could not be any lead solder in new plumbing or repairs (Health Canada, 2013; CMHC, 2004). The amount of lead that leaches into the water increases as the period of stagnation time (the time the water is standing still in the pipes) increases and, with very soft or acidic (low pH) water (Health Canada, 2013; CMHC, 2004).

Consumption of water must be considered as a possible exposure route, as Lanphear *et al.* (1998b) noted that blood lead levels correlated with higher lead in water and, Miranda *et al.* (2007) noted that changes in water treatment have been linked with broad increases in children's blood lead levels. Water treatment changes and the impact on children's blood lead levels was highlighted by Hanna-Attisha *et al.* (2016) in their paper looking at the Flint drinking water crisis, when a more corrosive water source was introduced. Brown *et al.* (2011) found that lead service lines in the distribution system were a risk factor for elevated blood lead levels even when the lead in water guideline was met. Ngueta *et al.* (2014) noted a change in water lead levels between summer and winter in Montreal which impacted children's blood lead levels, due to higher use of water during the summer months. It is recommended that tap water be run for 30 seconds or more to flush the distribution system, especially if the water has not been used for a couple of hours (US EPA, 2011a, Ngueta *et al.*, 2014).

Paint

Lead was added as a pigment to paints up until the 1960s, mainly for whites and pastel shades, with some containing as much as 50% lead by weight (CMHC, 2004). Leaded paint can represent a serious health hazard particularly if it is chipping and peeling or in reach of a child that can ingest it, and especially in homes built before the 1950s, as this was when paint contained very high levels of lead (Health Canada, 2013). In 1976, a federal law was passed that limited the amount of lead that could be added to interior paints, but it wasn't until 2005 that a similar law was passed for exterior paints. In 2010, the amount of lead that could be added to any consumer paints was limited to 90 mg/kg (Health Canada, 2013). The US EPA (1998) reported that paint is considered one of the most significant high dose sources of lead to a child, and Lanphear and Roghmann (1997) indicated that leaded paint could add large quantities of lead to soil and dust as well as contribute to the lead burden of a child. For these reasons, the US EPA announced that as of April 22, 2010, it is a federal law that any home undergoing renovations built before 1978, where 6 square feet of paint or more will be disturbed, must follow practices that will prevent lead contamination (US EPA, 2011a). Globally many nations still use lead in paint and the World Health Organization has established an alliance to eliminate lead in paint by 2020 due to the toxic effects it can have on children (O'Connor, D *et al.,* 2018). More recently a study from Flin Flon, MB and Creighton, SK found that BLLs in children had a significant positive association with household leaded paint (Safruk *et al.* 2017).

Garden Produce

Plants can take up lead from the soil, so a potential exposure route of concern for this project was produce grown in residential gardens for human consumption. In addition, air-borne lead particulate matter deposited on to the plants can be ingested (Dalenberg and Van Driel, 1990). The uptake of lead by plants is impacted by a number of factors including the type of plant, the condition of the soil such as pH and organic matter content, the concentration of lead in the soil, and lead speciation (Peryea, 2001; Samsoe-Petersen *et al.,* 2002). Amending the soil used to grow produce may help to reduce the uptake of lead by plants.

A study by Finster *et al.* (2004) in Chicago examined produce grown in urban soils to see if there was a relationship between the levels of lead in the soil and the levels of lead in the garden produce. The sampling methods used in this study are based on the Finster *et al.* study methods (2004). The Chicago study reported that all the produce sampled accumulated lead to some degree with most of the lead located in the plant roots and lesser amounts in the shoots and fruit (Finster *et al.*, 2004). Root vegetables, herbs and leafy vegetables had the highest lead concentrations of the edible parts of the plant, and the Chicago study noted that surface adhesion of lead particles was a factor in the lead concentrations, as vegetables washed with a mild detergent had less lead than those not washed in detergent (Finster *et al.,* 2004). Rahlenbeck *et al.* (1999) also found that leafy vegetables had the highest lead concentrations compared to other plant types, as did a study in Ontario, Canada (Ontario Ministry of the Environment, 2007) and in Puerto

Rico (Misenheimer, J. *et al.*, 2018). More recently a study by Byers *et al.* (2020) found lead concentrations were highest in tap root vegetables: turnip, beetroot, radish and carrots compared to fruits in the city of Milwaukee. Often lead does not distribute itself evenly throughout the plant, but tends to accumulate in the roots (Adriano, 2001). Hough *et al.* (2004) measured lead in different vegetable species and noted that the uptake of lead by the plants was relatively small compared to the lead levels in soil or dust. There may still be a general relationship between elevated soil lead concentrations and elevated lead concentrations in plant tissues as found in the Ontario study (Ontario Ministry of the Environment, 2007).

The US EPA (2011b) recommends that root crops not be grown in soil containing more than 1000 ppm lead and that no garden produce be grown in soils containing more than 1500 ppm lead. There are currently no such guidelines in Canada. In 2014 the US EPA had a working group come out with best management practices for gardening in lead contaminated areas. Table 1.2 adapted from the working group's publication summarizes their recommendations. The consumption of produce grown in lead-contaminated soil or in an area where there could be leaded-dust in the air, may contribute to a child's blood lead level as a potential exposure route.

Soil-Lead Concentration (ppm)	Category	Recommendation: Gardening Practices	Recommendation: Choosing Plants[a]
<100	Low risk	• No specific remedial action needed. • Wash hands, produce, clothes (good gardening and housekeeping practices).	• No restrictions of crop types.
>100–400[b] -·-·-·-·-·-▶ 400–1200	Potential risk	• Increasing use of good gardening and housekeeping practices as described in Table 3. • Relocate garden to lower risk garden areas. • Increasing use of soil amendments (*e.g.,* compost, clean fill), barriers (*e.g.,* mulch), and other remedial measures (see Table 3) up to and including raised beds and containers. • Ensure gardeners wear gloves and use tools to reduce soil contact and ingestion.	• Decrease planting of root vegetables or relocate root crop planting to lower risk areas. • Increase use of soil amendments and barriers to reduce soil deposition onto leafy vegetables. • Increase planting of fruiting vegetables, vegetables that grow on vines, and fruit trees.
>1200	High risk	• All of the above good gardening and housekeeping practices. • Raised beds, soil containers, soil replacement (*i.e.,* excavate contaminated soil and replace with soil containing low lead concentrations) are strongly recommended.[c] • Consider finding other locations for garden. • Restrict child access to only established safe areas. • Restrict all gardening by or for children in contaminated soils.	• Select plants with shallow roots for raised beds or areas with replacement soil to ensure that roots do not reach contaminated soil that is left in place, if any, otherwise, no restrictions.

Table 1.2: US EPA Technical Review Workgroup recommendations of best management practices regarding gardening and reducing exposure in lead contaminated soils. US EPA 2014.

Bioaccessibility

While lead may be present in our environment and may represent a human health risk, it is important to consider that not all the lead is necessarily available for uptake. Commonly metals require an aggressive (e.g. hot acid) digestion process in order to be completely released from their matrix (Yang *et al.,* 2003); however, the human body's digestive processes are not that aggressive. This is why bioaccessibility is an important aspect to consider when assessing lead exposure. This research project will examine soil lead bioaccessibility specifically.

Note that although the terms bioavailability and bioaccessibility are sometimes used interchangeably, they are defined differently. Bioavailability may be understood in a narrow sense to refer to the amount or proportion of an orally administered dose that is actually absorbed and reaches the circulatory system of a specific laboratory animal or human subject under a given set of exposure conditions (Oomen *et al.,* 2003; Ehlers and Luthy, 2003; Semple *et al.*, 2004). It is typically measured using *in vivo* dosing experiments. Bioaccessibility on the other hand is typically understood in a more general sense to refer to the amount or proportion of contaminant, such as lead, present in an environmental medium that is potentially available for uptake into the body of the exposed organisms (Semple *et al.*, 2004). It is typically measured using chemical leaching procedures. Based on this understanding, in this thesis the focus is on evaluating the bioaccessibility of lead in sampled soils using a chemical leach procedure and not a human GI assay. Commonly, it is the chemical form of the lead that most affects its bioaccessibility with lead oxides, nitrates and acetates being more soluble than other forms (Yang *et al.,* 2003).

There are many factors that can change the bioaccessibility of a metal in soil. Different soil properties including the soil pH, organic matter and clay content, cation exchange capacity and fertilization may all affect bioaccessibility of the lead (Fytianos *et al.,* 2001). Particle size is considered important, as the current bioaccessibility recommendations from the US EPA indicate that particles with diameters of <250μm (e.g., the combined clay, silt and fine sand fraction) that adhere to hands are most available for incidental

ingestion (US EPA, 2007). Ljung *et al.* (2006) described that soil lead concentrations increased with decreasing particle size, although there is considerable variability in lead availability based on particle size in the literature. A higher pH of the soil has been shown to decrease lead mobility and bioaccessibility (Yang *et al.,* 2003; Cao *et al.,* 2008). Organic matter has a high affinity towards binding heavy metals like lead and can form strong complexes which may affect the release of metals that are available for absorption (Chen *et al.,* 2006; Sklodowski *et al.,* 2006; Saminathan *et al.,*2010). Saminathan *et al.* (2010) also indicated that the more negatively charged sites that are available on soil particles (larger CEC) the greater the tendency for lead retention.

Chapter 2: Methodology

Sampling Strategy

Environmental media samples were collected at those residential properties selected to participate in the LeadNL study during the summer and fall of 2010. A total of 75 of the 95 neighbourhoods in the St. John's municipality were chosen as sampling units for recruitment in the LeadNL study. These 75 neighbourhoods were selected based on the abundance of homes within the targeted housing age cohorts (pre-1946, 1946-1960, 1961-1970, 1971-1980, 1981-2000 and Post-2000). In total, 249 participants from 194 households satisfied study criteria (e.g. had children between 6 months and 6 years old) and were recruited to take part in the LeadNL study. A phlebotomist at the blood collection clinic in the Janeway Children's Health and Rehabilitation Centre collected blood by venipuncture (Bell *et al.,* 2011).

In this study, indoor dust, soil, paint, tap water, and garden produce were the exposure sources sampled. During field sampling, described below, every effort was made to avoid contamination in the sampling process and cross-contamination of samples. For example, strict sampling protocols were followed and sampling tools (e.g. Teflon shovel) and containers (e.g. plastic bags) were used that had certified intrinsically low or no lead content. Sample bags were immediately sealed and labeled after sample collection to avoid loss or cross-contamination.

Soil samples were collected from three areas of residential properties using sampling protocols from the US Department of Housing and Urban Development (HUD, 1995). These three areas were the dripline (perimeter of house), play area and vegetable garden, where present. Areas were sampled depending on their use characteristics as identified by homeowners in a questionnaire delivered at the beginning of the sampling appointment. Soil samples for each area were composites of between 5 and 10 sub-samples collected into plastic bags from either the bare surface soil or just under the vegetation mat (0-5cm max), where no bare soil was exposed. A total of 421 soil samples were collected from 190 study households. Each sample was about 200 grams. This total sample number included duplicate samples that were collected roughly every tenth sample to represent about 10% of the overall soil samples collected. Soil samples were collected from the surface because children are most likely to come into contact with surface soil. Surface soils are often more highly contaminated than underlying soils, because there is typically little movement of added anthropogenic lead through the soil column (Turjoman and Fuller, 1986).

Indoor dust lead samples were collected according to HUD (1995) guidelines from three areas of the house: floor, windowsill and window trough in the rooms most commonly used by the children participating in the LeadNL study. The rooms were identified through questionnaire responses. All dust samples were collected by means of two swipe paths using an 'S' shaped motion with EPA approved *Ghost Wipes*. Floor dust samples were collected using a template of 12"x12" for a 1-square foot area and windowsill and trough dust samples were collected from a 0.1-square foot area. Imperial units were used

to conform to HUD protocols. A total of 2172 dust samples were collected, including 209 duplicates, that were collected roughly every tenth sample for a 10% duplicate representation, from 194 study households.

Paint chips were collected from both indoor and outdoor surfaces in areas where the paint was chipping or peeling, following HUD (1995) guidelines. To collect paint chips, a metal scraper was used to peel paint off onto filter paper, with every effort taken to avoid collecting the substrate under the paint around 2cm in size. A total of 304 paint chip samples were collected from indoor and outdoor locations, 35 of which were duplicate samples representing roughly 12% of the overall sample, from 151 study households.

Cold tap water samples were collected from the water source that the study participant (child) most commonly consumed their drinking water from, based on information from the household questionnaire. This was typically the kitchen faucet and sometimes the bathroom. Sampling protocols were based on the 2009 Health Canada study *Impact of Drinking Water Lead Levels on the Exposure of Young Children to Lead*, in which a minimum of 5 one-litre samples were taken (Prévost and Lemieux, 2009, Levallois, P. *et al.,* 2014). The first 1 litre bottle of tap water was collected after a 5-minute flush period that was meant to completely flush the water lines in the home from the water main to the house. During this time both the water temperature and the flow rate were recorded. The next 4 litres were consecutively sampled after a 30-minute period of stagnation. All water samples were stored at 4°C and were acidified until they had reached a pH of 2 or less

through the addition of nitric acid. A total of 1030 tap water samples were collected from 194 study households.

Where present, garden produce samples were collected in households following sampling protocols developed by Finster *et al*. (2004) for a similar study in Chicago. Samples were collected from both above-ground produce types (e.g. lettuce, tomatoes, strawberries, rhubarb and herbs) and below-ground produce types (e.g. potatoes, onions and carrots). A total of 89 garden produce samples were collected, including 34 duplicates representing ~40% of the total sample, from 34 study households. As there were very few homes with produce available, duplicates were taken at all homes with enough produce.

Laboratory Processing

Following collection, environmental samples were processed in the project laboratory at Memorial University before being distributed for analysis. Once air-dried, soil samples were sieved to remove pebbles and rocks larger than 2 mm and samples were split using a riffle splitter for the following analyses: near-total lead concentration, bioaccessible lead concentration, grain-size distribution, pH, organic content and cation exchange capacity. The bioaccessible lead samples were chosen based on computer randomization. Garden produce samples were processed to mimic common vegetable preparation methods prior to cooking and consumption. Above-ground produce samples were split into two for each household. One half was rinsed with tap water and the other half with mild detergent. Below-ground produce for each household was split into thirds and were either rinsed

with tap water, washed with mild detergent or peeled. Paint, water and dust samples were sent along to the selected laboratories for further processing.

Laboratory Analysis

Near-total lead was measured in all environmental media at Maxxam Analytics in Bedford, Nova Scotia, an accredited ISO 17025 certified laboratory. Analytical methods followed US EPA protocols (Table 2.1). Detection limits for each analytical method are also listed in Table 2.1.

Table 2.1: Laboratory protocols and detection limits for near-total lead measurements in environmental media.

Environmental Media	Analysis Protocol[2]	Measurement Type	Detection Limit
Dust	US EPA SW-846 Method IO-3.1	ICP-MS	0.125 µg
Soil	US EPA SW-846 Method 3050B	ICP-MS	0.5 mg/kg
Water	US EPA SW-846 Method 6020A/EPA 200.8	ICP-MS	0.01 µg/L
Paint	US EPA SW-846 Method 3050B	ICP-MS	50 mg/kg
Garden Produce	US EPA SW-846 Method 6020A/EPA 200.3	ICP-AES	0.18 mg/kg (dw)

dw = dry weight

Quality control was regularly performed by Maxxam Analytics with blanks, certified reference material, duplicates, matrix spikes and spiked blanks being used. Table A.1 in

2 US EPA SW-846 Method IO-3.1 uses concentrated HNO_3 and HCl and heat to digest substrate. US EPA Method 3050B is a strong acid digestion that will dissolve all elements that could become 'environmentally available' by using HNO_3 and H_2O_2. US EPA Method 6020A determines analytes from filtered and acid preserved samples through ICP-MS. EPA 200.8 allows the determination of dissolved elements in ground waters, surface waters and drinking waters. EPA 200.3 measures total recoverable metals in biological tissues.

the appendix provides the results in further detail for the quality control. Table 2.2 displays the overall number of environmental samples collected and the number of duplicate samples collected for each medium.

Table 2.2: Environmental media samples and media duplicate sample numbers collected from study homes throughout St. John's, NL.

	Dust	Soil	Paint	Water	Garden Produce
# Samples	2172	354	304	1030	89
# Duplicates	209	67	35	NA	34

*Note: the NA indicates that no duplicates were collected for water samples as replicate analysis was built into the water sampling protocols.

The field duplicates collected were analyzed to determine how representative our sample results were. Overall, all the samples except for garden produce had strong correlations between the duplicate and sample values (Table 3.2). The low correlation for garden produce is likely due to the fact that the concentrations of lead encountered were often close to or below the method detection limit. There were a very small number of produce samples (n=10) with detectable levels of lead.

Table 2.3: Pearson correlation coefficients (r) for duplicate environmental samples. Bolded values indicate a statistical significance ($\alpha = 0.05$).

Environmental Media	r	p-value
Dust	**0.78**	< 0.0001
Soil	**0.87**	< 0.0001
Paint	**0.99**	< 0.0001
Garden Produce	0.13	0.4462

Potentially bioaccessible lead was measured at Howard Mielke's laboratory at Xavier University in New Orleans, using the nitric acid method. This is similar to the method of Minca (2012), which uses the following extraction method: acid pH (1 M HNO_3) shaker time for 2 hours and room temperature (~22 °C). ICP-AES techniques were used to measure lead (Mielke *et al.,* 2005). A second technique used a water-leach method (Garrett *et al.,* 2009). Samples were randomly selected for location and housing age group.

Grain size was measured by fractionation into sand, silt, clay and particles less then 250 μm by dry sieve. A LA-950 Laser Scattering Particle Size Distribution Analyzer was used to measure size distributions.

Cation Exchange Capacity, organic content and pH were measured at the Newfoundland and Labrador Provincial Soil Lab based on their procedural methods. Cation exchange capacity was determined using extraction with an ammonium acetate solution (US EPA, 1986). It was calculated according to this equation: Ca/200 + Mg/120 + K/390 + 8 (8-Buffer pH). The organic content was determined using Loss on Ignition (LOI) that was heated at 430°C for 6 hours (Misra, 2011). The pH of the soil was measured on a 1:1 ratio for mineral soils to buffer solution with buffer pH done using Adams-Evans method (Misra, 2011).

Data Analysis

All results were entered into a database using *Microsoft Access 2003*. Data quality checks were conducted using SAS version 9.2, as were all statistical analyses. Statistical

significance was assessed at the 95% confidence level ($\alpha = 0.05$). Data was not normal so it was log-transformed for analysis and Shapiro-Wilk test done to confirm criteria for parametric statistical analysis. Due to a large number of values having non- detectable near-total lead concentrations for certain media, half the lead detection limit value for each environmental media was substituted for all results including the non- detect results in order to facilitate statistical analysis based on discussion with project statistician (Sears, W., personal communication, 2010). The soil and water results had no non-detectable values. For the dust results, there was a 16% non-detectable rate (with floor having 10%, sill having 37% and trough having 5%), paint had 39% non-detectable rate (with 28% for outdoor samples and 49% for indoor samples), and the garden produce samples had an 89% non-detectable rate overall (with 94% for above-ground produce and 76% for below-ground produce types). Descriptive statistics (geometric mean (GM), median, 95^{th} percentile and range) were calculated by both the full housing age categories (pre-1946, 1946-1960, 1961-1970, 1971-1980, 1981-2000 and post-2000) and by pre-1970 and our reference cohort, post-1980. Statistical tests (t-tests, simple and multiple linear regressions) were carried out to determine whether housing age was a significant predictor of environmental lead levels and whether there were any significant correlations between the environmental media and BLL's. The BLL analyses were done by the larger biomonitoring group and GM values provided to run statistical analyses with the environmental lead levels. Quality control measures from Maxxam Analytics laboratories reports were assessed and summarized (see Appendix for summary Table A.1). Descriptive statistics were calculated by housing age categories (pre-1970 and post-1980) and by soil location (dripline or play area) for the bioaccessibility analysis.

Chapter 3: Results

Environmental media samples were collected from 194 study households and represented 249 participants in total.

Dust Results

The summary statistics for the dust values and by sample location are shown in Table 3.1. The floor dust samples had the lowest GM lead loadings (0.61 μg/ft^2) while the window trough samples had the highest GM dust lead loadings (17.8 μg/ft^2), over 29 times higher than the floor GM. The window troughs had the widest range of loadings (n.d. to 6,101 μg/ft^2).

Table 3.1: Sample size (n), geometric mean (GM), median, 95th percentile and 95% percentile confidence intervals (CI) and overall range for dust lead loadings (μg/ft^2) by sample location on household properties.

Sample Type	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Floor	194	0.61 (0.53, 0.71)	0.60	4 (2.6, 4.04)	n.d. – 407
Sill	184	1.1 (0.81, 1.42)	1.35	18.5 (17.2, 40.7)	n.d. – 1,236
Trough	180	17.8 (13.5, 23.5)	17.8	582 (273, 646)	n.d. – 6,101

Note: n.d. = non-detectable
Summary statistics are based on household-averaged dust lead loadings for each area within the household
Method detection limit 0.125 μg

Statistical analysis using an independent two tailed t- test of housing age categories revealed that there was a significant difference ($p<0.0001$) between dust-lead loadings of pre-1970 homes and our reference cohort, the post-1980 homes for floors ($t = -10.07$), windowsills ($t = -7.46$) and window troughs ($t = -7.27$). The summary statistics per housing age category and dust location are shown in Tables 3.2 to 3.4.

Table 3.2: Descriptive statistics for household-averaged floor dust lead levels ($\mu g/ft^2$) by target (pre 1970) and reference (post 1980) housing cohorts and individual housing cohorts.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	0.98 (0.82, 1.16)	0.85	5.53 (3.52, 5.96)	0.14 - 407.27
Post-1980	58	0.28 (0.24, 0.34)	0.24	0.89 (0.67, 1.16)	n.d. - 200.50
Pre-1946	57	1.33 (1.03, 1.72)	1.13	8.04 (4.68, 10.41)	0.22 - 69.26
1946-1960	26	0.82 (0.59, 1.14)	0.75	3.69 (2.08, 5.80)	0.14 - 24.58
1961-1970	31	0.64 (0.49, 0.83)	0.74	1.70 (1.48, 3.35)	0.16 - 407.27
1971-1980	22	0.42 (0.31, 0.57)	0.38	1.37 (0.92, 2.47)	0.10 - 3.86
1981-2000	33	0.37 (0.29, 0.48)	0.38	1.36 (0.85, 1.82)	n.d. - 200.50
Post-2000	25	0.20 (0.16, 0.23)	0.21	0.36 (0.32, 0.56)	0.08 - 0.61

Table 3.3: Descriptive statistics for household-averaged sill dust lead levels (μg/ft^2) by target (pre 1970) and reference (post 1980) housing cohorts and individual housing cohorts.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	105	2.42 (1.74, 3.36)	3.70	27.82 (25.13, 69.23)	n.d. - 1236.1
Post-1980	58	0.32 (0.21, 0.49)	0.20	7.57 (2.67, 10.14)	n.d. - 68.06
Pre-1946	54	3.54 (2.35, 5.34)	4.90	36.96 (24.62, 88.18)	n.d. - 177.05
1946-1960	22	1.30 (0.57, 2.93)	1.21	16.39 (10.13, 133.00)	n.d. - 1236.1
1961-1970	29	1.91 (0.96, 3.78)	2.67	25.26 (15.72, 133.16)	n.d. - 31.21
1971-1980	21	0.51 (0.23, 1.14)	0.30	9.93 (3.60, 45.28)	n.d. - 20.26
1981-2000	33	0.70 (0.39, 1.27)	0.61	10.22 (5.22, 33.37)	n.d. - 68.06
Post-2000	25	0.11 (0.08, 0.16)	0.06	0.64 (0.29, 0.82)	n.d. - 1.9

Table 3.4: Descriptive statistics for household-averaged trough dust lead levels (μg/ft^2) by target (pre 1970) and reference (post 1980) housing cohorts and individual housing cohorts.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	105	39.54 (28.31, 55.23)	32.99	860.22 (427.75, 1199.21)	n.d. - 6101.09
Post-1980	54	5.40 (3.50, 8.32)	5.40	39.27 (41.48, 158.67)	n.d. - 4154.15
Pre-1946	53	60.28 (38.64, 94.05)	57.43	1171.05 (478.31, 1900.61)	2.7 - 5387.72
1946-1960	22	21.55 (8.80, 52.75)	19.64	860.22 (205.56, 3460.86)	n.d. - 6101.09
1961-1970	30	29.29 (16.47, 52.07)	24.50	521.42 (181.07, 1096.52)	2.80 - 1533.47
1971-1980	21	7.06 (3.66, 13.62)	12.26	35.93 (34.90, 278.44)	1.28 - 70.67
1981-2000	32	8.21 (4.79, 14.07)	5.85	39.27 (48.95, 263.81)	1.30 - 4154.15
Post-2000	22	2.93 (1.48, 5.82)	4.38	14.05 (16.47, 142.80)	n.d. - 55.89

Figure 3.1 graphically displays the differences between the six overall project housing age categories for GM dust-lead loadings for floor, windowsill and trough samples by category. All three locations have a significant difference between GM dust-lead loadings from the oldest housing category (pre-1946) to the younger housing categories of 1981-2000 and post-2000.

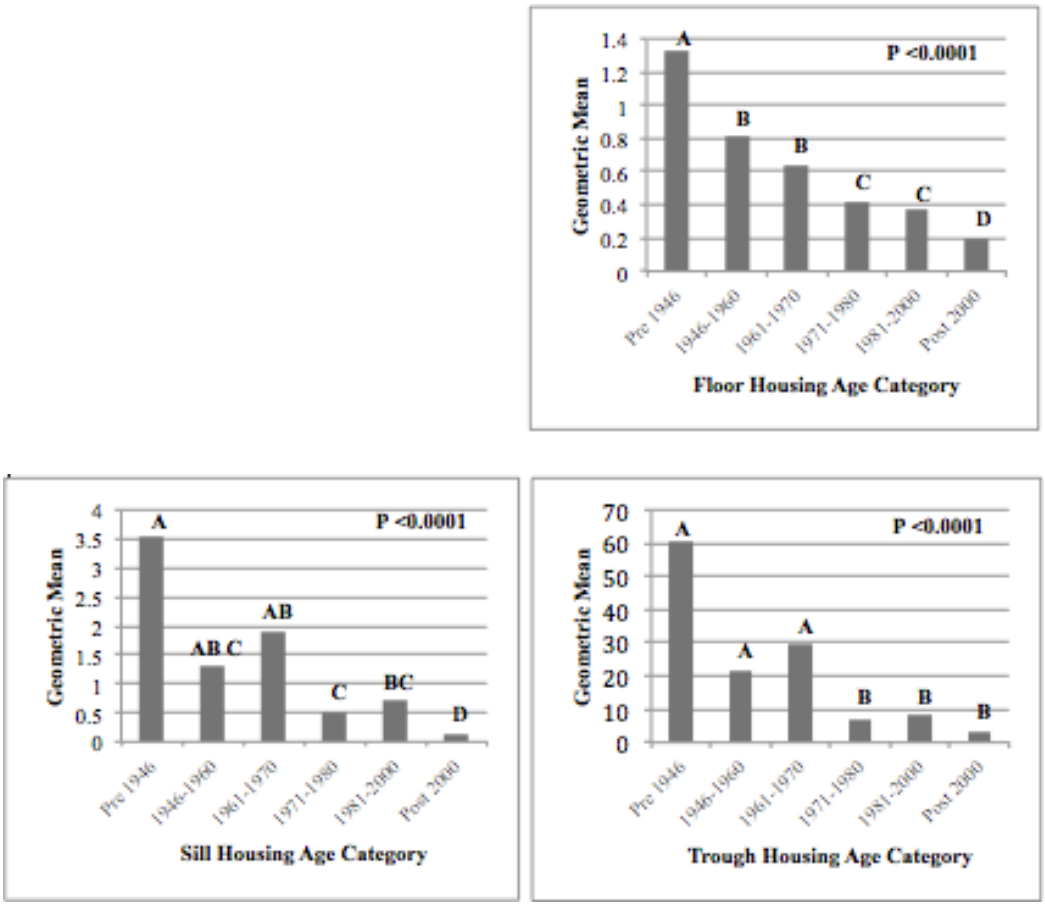

Figure 3.1: Geometric means (µg/ft^2) for dust lead loadings for floor, windowsill and window trough by housing age category. Means associated with the same letter are not significantly different at the 95% confidence level (α=0.05).

Soil Results

Overall, dripline samples had the highest GM value at 103 ppm and the largest range of values from 8.5 to 6800 ppm. Play area had the second highest geometric mean at 85.8 ppm followed by the garden samples at 52.8 ppm.

Table 3.5: Sample size (n), geometric mean (GM) with 95% confidence intervals (CI), median, 95th percentile with 95% CI, and the overall range of soil lead values for the different sample locations measured in ppm.

Sample Type	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Dripline	141	103 (81.3, 131)	85	1,601 (781, 1,630)	8.5–6,800
Play Area	182	85.8 (72, 102)	76.8	541 (480, 822)	3.8–1,900
Garden	31	52.8 (34.7, 80.3)	58	481 (206, 764)	2.8–560

Note that the garden soil numbers is fewer then the households where produce was sampled secondarily to soil not being collected at 3 houses.

Statistical analysis using an independent two tailed t-test of housing age categories revealed that there was a significant difference between pre-1970 and post-1980 geometric mean values for dripline, play area and garden soil samples ($t = -14.38$, $p<0.0001$; $t = -14.41$, $p<0.0001$ and $t = -4.17$, $p=0.0003$, respectively). The overall project housing age categories differences are displayed in Figure 3.2 for the dripline, play area and garden samples.

Table 3.6: Descriptive statistics for household-averaged dripline soil lead levels (ppm) by target (pre 1970) and reference (post 1980) housing cohorts and individual housing cohorts.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	72	287.54 (216.13, 382.54)	235.87	1901.35 (1446.15, 3496.75)	8.5-6800
Post-1980	50	28.06 (24.11, 32.65)	26.84	79.35 (55.37, 88.64)	9.4-110
Pre-1946	29	584.78 (377.08, 906.88)	571.35	4401.35 (2268.85, 8966.54)	63-6800
1946-1960	18	307.68 (189.01, 500.86)	256.30	1601.35 (878.58, 4127.30)	31-1600
1961-1970	25	120.20 (84.49, 170.98)	131.35	351.35 (319.42, 966.99)	8.5-540
1971-1980	19	65.31 (51.33, 83.10)	61.35	191.35 (112.25, 240.84)	22-190
1981-2000	27	31.89 (25.78, 39.44)	32.35	83.35 (59.49, 115.86)	12-110
Post-2000	23	24.15 (19.46, 29.97)	23.35	54.35 (42.38, 83.65)	9.4-64

Table 3.7: Descriptive statistics for household-averaged play area soil lead levels (ppm) by target (pre 1970) and reference (post 1980) housing cohorts and individual housing cohorts.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	107	167.48 (137.44, 204.08)	161.35	741.35 (696.03, 1280.60)	3.8-1900
Post-1980	54	27.57 (23.68, 32.09)	24.84	74.35 (56.38, 90.29)	9.5-230
Pre-1946	55	283.06 (220.50, 363.38)	321.35	901.35 (931.69, 2021.22)	3.8-1900
1946-1960	24	134.88 (97.93, 185.79)	146.26	391.35 (319.20, 874.15)	20-400
1961-1970	28	71.92 (52.80, 97.95)	68.70	281.35 (182.43, 480.47)	8.2-530
1971-1980	21	52.65 (39.76, 69.71)	47.35	151.35 (104.18, 252.88)	21-210
1981-2000	30	28.91 (23.04, 36.27)	23.84	98.35 (59.29, 120.62)	13-230
Post-2000	24	25.98 (21.12, 31.96)	26.33	47.35 (45.36, 87.03)	9.5-72

Table 3.8: Descriptive statistics for household-averaged garden soil lead levels (ppm) by target (pre 1970) and reference (post 1980) housing cohorts.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	22	63.98 (39.23, 104.32)	69.45	351.35 (219.29, 1025.23)	2.8-480
Post-1980	7	20.23 (14.18, 28.85)	18.35	40 (27.54, 91.81)	11-39

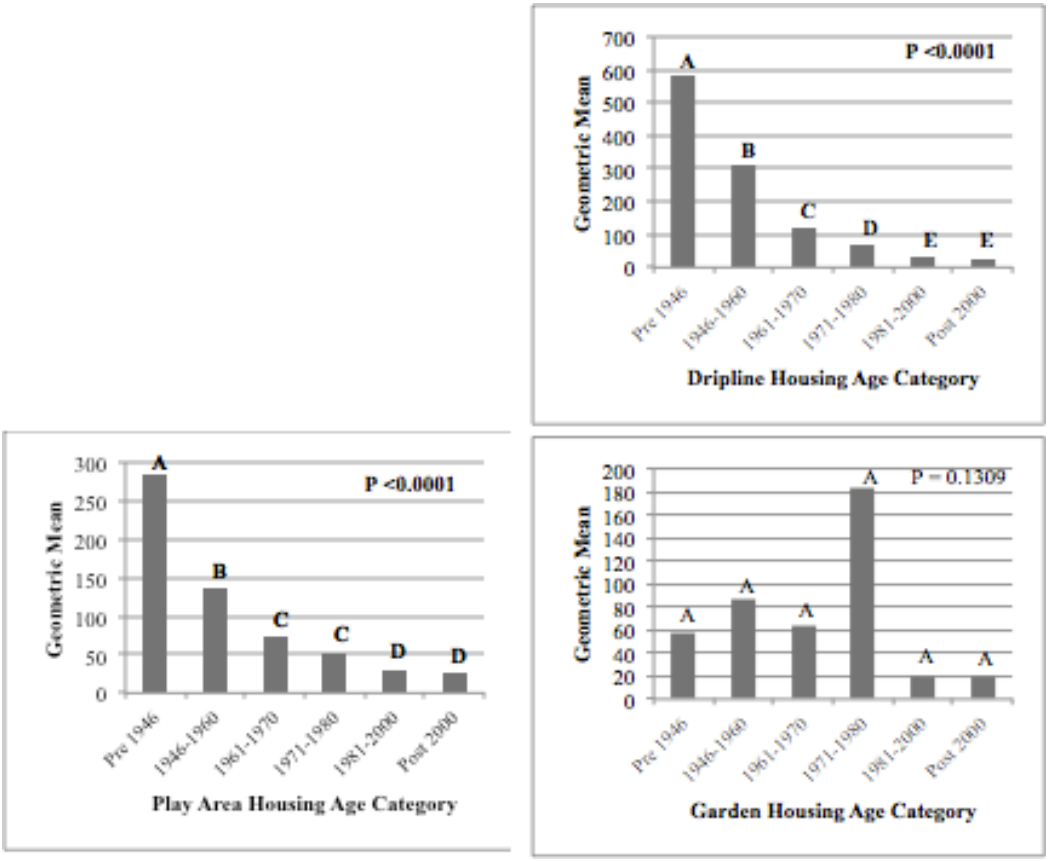

Figure 3.2: Geometric mean (ppm) by housing age category for dripline, play area and garden soil samples from the Student-Newman-Keuls (SNK) test for differences between geometric mean values by housing age cohorts at the 95% confidence level (α=0.05). Means associated with the same letter are not significantly different at the 95% confidence level (α=0.05).

Water Results

Only summary statistics for the bathroom samples are displayed, as very few homes had samples collected in this location, and the majority of study samples were from the kitchen. Table 3.9 displays the geometric means for locations of sampling in each household. The stagnation samples had a slightly higher geometric mean and range as compared to the flush samples. Overall the kitchen samples had a slightly broader range of values than the bathroom samples, but the geometric means for bathroom flush

samples (0.70 µg/L) and stagnation samples (3.08 µg/L) were higher than the kitchen values.

Table 3.9: Sample size (n), geometric mean (GM) with 95% confidence intervals (CI), median, 95th percentile with 95% CI, and the range of tap water lead values (µg/L) for all household samples.

Sample Type	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Kitchen: Flush	194	0.57 (0.52, 0.64)	0.51	2.3 (1.75, 2.45)	0.11–19.1
Kitchen: Average Stagnation	194	0.97 (0.87, 1.08)	0.84	4.31 (2.99, 4.41)	0.15–52.8
Kitchen: Stagnation 1	194	1.46 (1.3, 1.65)	1.28	6.17 (4.93, 7.12)	0.28-52.8
Kitchen: Stagnation 2	194	0.94 (0.84, 1.06)	0.83	4.18 (3.12, 4.48)	0.15-30.3
Kitchen: Stagnation 3	194	0.86 (0.76, 0.98)	0.72	3.76 (2.98, 4.34)	0.16-33.2
Kitchen: Stagnation 4	194	0.74 (0.66, 0.83)	0.65	3.29 (2.35, 3.32)	0.15-31.2
Bathroom: Flush	12	0.70 (0.40, 1.22)	0.56	4.11 (1.63, 9.78)	0.15–4.06
Bathroom: Average Stagnation	12	3.08 (1.36, 6.99)	2.16	44.2 (10.7, 152.5)	0.45-45.02

Results for the different stagnation levels by housing age categories are displayed in Tables 3.10-3.15. Using Satterthwaite's t-test for unequal variances, analysis revealed that there was no significant difference between pre-1970 and post-1980 samples for kitchen flush samples ($t = -1.20$, $p=0.2341$) and for kitchen average stagnation samples ($t = -0.84$, $p=0.4009$). For the different kitchen stagnation levels, stagnation 1 did have a significant difference between pre-1970 and post-1980 samples ($t = -01.99$, $p = 0.0488$) while stagnation 2, 3 and 4 did not have significant differences ($t = -0.93$, $p=0.3521$; $t = -$

0.30, p=0.7618; and t = 0.07, p=0.9436 respectively).

Table 3.10: Descriptive statistics for household-averaged kitchen flush water lead levels (μg/L) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	0.60 (0.51, 0.69)	0.52	3.38 (1.85, 3.96)	0.125–19.1
Post-1980	58	0.52 (0.43, 0.62)	0.44	2.08 (1.28, 2.26)	0.11–3.25
Pre-1946	57	0.73 (0.58, 0.91)	0.65	3.44 (2.18, 4.36)	0.125–19.1
1946-1960	26	0.44 (0.37, 0.54)	0.41	0.92 (0.76, 1.38)	0.16–1.12
1961-1970	31	0.53 (0.38, 0.74)	0.44	4.28 (1.56, 4.45)	0.14–13.1
1971-1980	22	0.63 (0.46, 0.86)	0.55	2.59 (1.38, 3.67)	0.17–3
1981-2000	33	0.54 (0.41, 0.71)	0.38	2.16 (1.36, 3.19)	0.14–3.25
Post-2000	25	0.49 (0.38, 0.63)	0.48	1.77 (0.97, 2.13)	0.11–1.91

Table 3.11: Descriptive statistics for household-averaged kitchen stagnation water lead levels (μg/L) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	0.99 (0.85, 1.14)	0.92	3.64 (2.98, 4.70)	0.23-36.87
Post-1980	58	0.89 (0.73, 1.08)	0.72	4.17 (2.31, 4.21)	0.27-7.35
Pre-1946	57	1.24 (0.99, 1.56)	1.12	5.57 (3.80, 7.73)	0.23-36.87
1946-1960	26	0.79 (0.64, 0.98)	0.75	1.93 (1.46, 2.86)	0.23-2.98
1961-1970	31	0.78 (0.59, 1.03)	0.73	2.23 (1.90, 4.46)	0.23-9.84
1971-1980	22	1.09 (0.75, 1.58)	0.99	4.52 (2.78, 8.99)	0.25-5.96
1981-2000	33	0.99 (0.73, 1.33)	0.69	4.97 (2.70, 6.85)	0.27-7.35
Post-2000	25	0.78 (0.62, 0.98)	0.76	2.10 (1.47, 3.04)	0.31-4.95

Table 3.12: Descriptive statistics for household-averaged kitchen stagnation 1 water lead levels (µg/L) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	1.55 (1.32, 1.81)	1.40	6.14 (4.99, 8.09)	0.3-52.8
Post-1980	58	1.19 (0.97, 1.47)	0.96	6.11 (3.30, 6.26)	0.28-8.86
Pre-1946	57	1.97 (1.56, 2.47)	2.02	7.54 (6.04, 12.32)	0.39-52.8
1946-1960	26	1.20 (0.95, 1.52)	1.22	3.23 (2.34, 4.85)	0.3-3.23
1961-1970	31	1.23 (0.89, 1.70)	1.31	4.13 (3.52, 9.73)	0.3-17.9
1971-1980	22	1.83 (1.21, 2.75)	1.43	11.95 (5.12, 18.60)	0.51-14.3
1981-2000	33	1.50 (0.73, 1.08)	1.15	7.57 (1.54, 2.86)	0.28-8.86
Post-2000	25	0.89	0.85	2.01	0.33-2.89

Table 3.13: Descriptive statistics for household-averaged kitchen stagnation 2 water lead levels (μg/L) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	0.96 (0.82, 1.12)	0.86	4.12 (3.05, 4.90)	0.19-30.3
Post-1980	58	0.85 (0.97, 1.04)	0.72	3.84 (2.32, 4.35)	0.16-12.4
Pre-1946	57	1.21 (0.95, 1.53)	1.08	5.15 (3.80, 7.87)	0.19-30.3
1946-1960	26	0.78 (0.61, 1.00)	0.84	1.97 (1.58, 3.43)	0.22-3.34
1961-1970	31	0.75 (0.56, 1.00)	0.73	2.54 (1.92, 4.77)	0.22-8.43
1971-1980	22	1.07 (0.69, 1.65)	0.84	4.53 (3.21, 12.70)	0.15-7.59
1981-2000	33	0.92 (0.67, 1.25)	0.62	3.84 (2.61, 6.86)	0.2-12.4
Post-2000	25	0.77 (0.60, 1.00)	0.74	2.95 (1.57, 3.51)	0.16-4.05

Table 3.14: Descriptive statistics for household-averaged kitchen stagnation 3 water lead levels (µg/L) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	0.87 (0.74, 1.02)	0.74	3.76 (2.94, 4.86)	0.16-33.2
Post-1980	58	0.83 (0.67, 1.04)	0.67	5.16 (2.52, 5.25)	0.16-16.5
Pre-1946	57	1.11 (0.86, 1.45)	0.95	7.44 (4.05, 9.19)	0.17-33.2
1946-1960	26	0.67 (0.53, 0.84)	0.67	1.79 (1.28, 2.61)	0.21-1.87
1961-1970	31	0.69 (0.52, 0.90)	0.67	1.77 (1.66, 3.89)	0.16-7.2
1971-1980	22	0.90 (0.63, 1.30)	0.81	3.76 (2.23, 6.93)	0.17-3.85
1981-2000	33	0.91 (0.54, 1.01)	0.81	5.16 (1.76, 4.68)	0.24-16.5
Post-2000	25	0.74	0.64	1.74	0.16-10.6

Table 3.15: Descriptive statistics for household-averaged kitchen stagnation 4 water lead levels (μg/L) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	114	0.73 (0.63, 0.86)	0.65	3.89 (2.31, 3.93)	0.15-31.2
Post-1980	58	0.74 (0.61, 0.90)	0.65	3.48 (1.97, 3.63)	0.15-5.91
Pre-1946	57	0.89 (0.70, 1.14)	0.75	5.31 (2.94, 6.27)	0.18-31.2
1946-1960	26	0.63 (0.49, 0.80)	0.61	1.37 (1.26, 2.73)	0.18-3.84
1961-1970	31	0.59 (0.45, 0.77)	0.59	1.49 (1.38, 3.16)	0.15-5.82
1971-1980	22	0.80 (0.56, 1.14)	0.65	3.48 (1.94, 5.92)	0.17-3.61
1981-2000	33	0.75 (0.56, 1.01)	0.62	4.41 (2.05, 5.15)	0.21-5.91
Post-2000	25	0.72 (0.55, 0.95)	0.69	2.15 (1.52, 3.51)	0.15-3.43

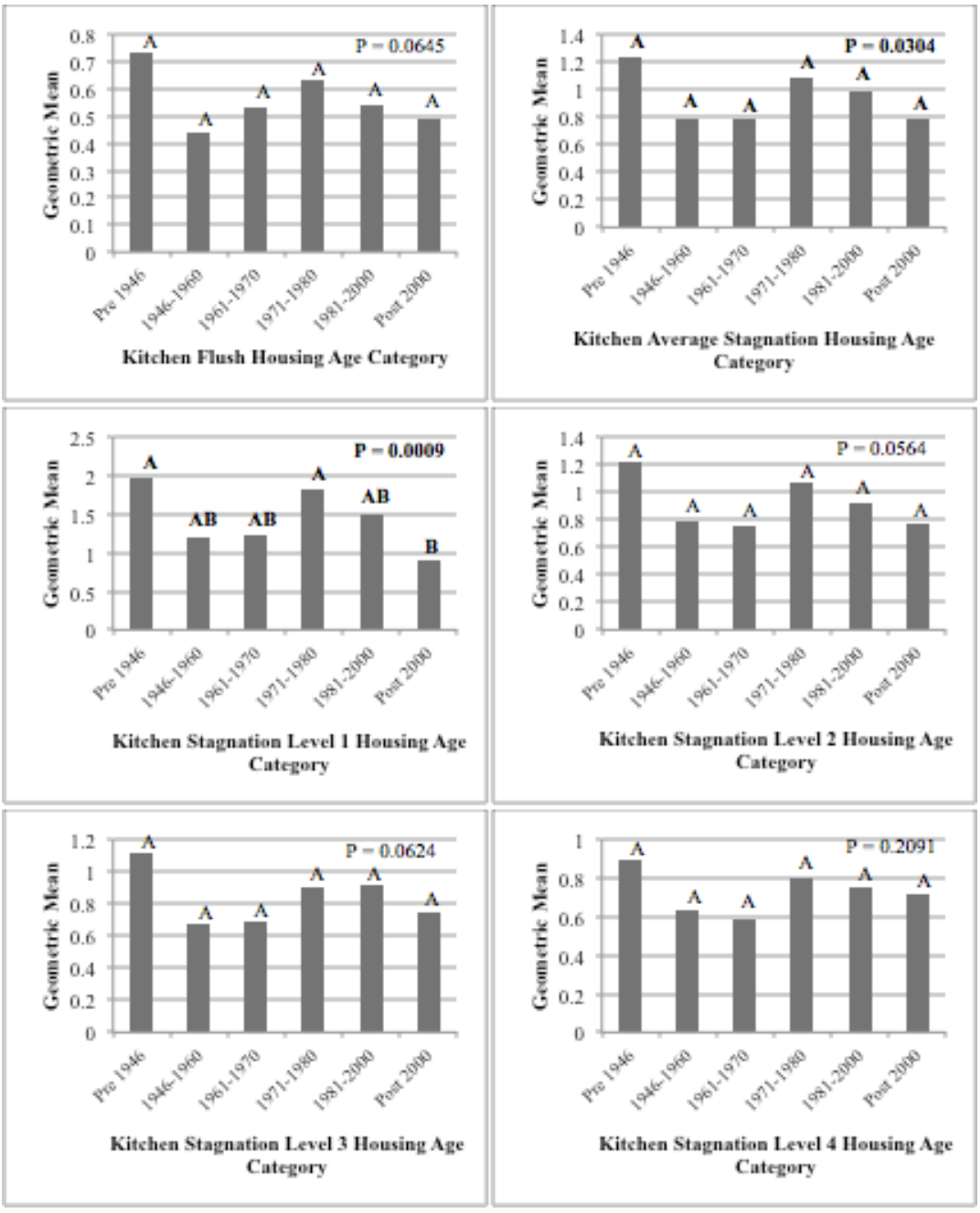

Figure 3.3: Geometric mean (µg/L) graphs by housing age category for kitchen flush, kitchen average stagnation and the 4 stagnation levels from the Student-Newman-Keuls (SNK) test for differences between geometric mean values by housing age cohorts at the 95% confidence level (α=0.05). Means associated with the same letter are not significantly different at the 95% confidence level (α=0.05).

Paint Results

Interior paint samples had a slightly higher GM (399 ppm) and a wider range of total lead values (n.d. - 110,000 ppm) than exterior samples (GM 335 ppm; range n.d. - 52,850 ppm. Table 3.16).

Table 3.16: Sample size (n), geometric mean (GM) with 95% confidence intervals (CI), median, 95th percentile with 95% CI, and the overall range of paint lead values (ppm) by sample location.

Sample Type	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Exterior	117	335 (228, 490)	240	27,028 (9,748, 31,593)	n.d.–52,850
Interior	112	399 (263, 604)	470	31,028 (16,102, 57,894)	n.d.–110,000

Note: n.d. = non-detectable

Analysis of the housing age categories using Satterthwaite's t-test for unequal group variances revealed that there was a significant difference between pre-1970 and post-1980 homes for exterior and interior paint lead values ($t = -8.59$, $p<0.0001$ and $t = -8.58$, $p<0.0001$; Tables 3.17 and 3.18 respectively).

Table 3.17: Descriptive statistics for household-averaged indoor paint lead levels (ppm) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	80	670.44 (374.39, 1200.59)	662.48	34494.90 (34494.90, 136989.74)	n.d.-110000
Post-1980	19	38.58 (27.65, 53.84)	27.50	224.24 (81.61, 234.62)	n.d.-350
Pre-1946	38	2398.01 (1054.46, 5453.46)	4338.71	86027.50 (51500.03, 666355.31)	n.d.-110000
1946-1960	19	428.71 (140.06, 1312.29)	561.50	29027.50 (5302.52, 183696.00)	n.d.-29000
1961-1970	23	118.12 (52.19, 267.34)	27.50	1827.50 (992.43, 13006.64)	n.d.-18966.67
1971-1980	13	49.09 (24.74, 97.41)	27.50	527.50 (150.62, 1370.32)	n.d.-500
1981-2000	16	41.11 (27.73, 60.96)	27.50	224.24 (88.82, 312.01)	n.d.-350
Post-2000	3	n.d. (NA)	n.d.	n.d. (NA)	n.d.

Note: n.d. = non-detectable

Table 3.18: Descriptive statistics for household-averaged outdoor paint lead levels (ppm) by housing age cohorts and pre-1970 versus reference cohort post-1980.

Housing Age	n	GM (95% CI)	Median	95th percentile (95% CI)	Range
Pre-1970	81	534.83 (328.17, 871.63)	452.25	21027.50 (10453.75, 47281.29)	n.d.-52850
Post-1980	21	38.34 (26.12, 56.26)	27.50	267.50 (97.40, 327.06)	n.d.-400
Pre-1946	43	446.51 (230.61, 864.53)	373.84	12868.99 (6520.10, 50903.70)	n.d.-41000
1946-1960	18	479.29 (129.94, 1767.91)	364.70	23935.49 (7965.88, 502341.98)	n.d.-52850
1961-1970	20	870.19 (347.16, 2181.22)	885.82	32338.65 (7477.54, 136837.80)	n.d.-36000
1971-1980	15	198.71 (67.19, 587.67)	217.50	11027.50 (1481.21, 47536.05)	n.d.-11000
1981-2000	18	40.52 (25.88, 63.43)	27.50	421.73 (106.38, 441.49)	n.d.-400
Post-2000	3	n.d. (NA)	n.d.	n.d. (NA)	n.d.

Note: n.d. = non-detectable

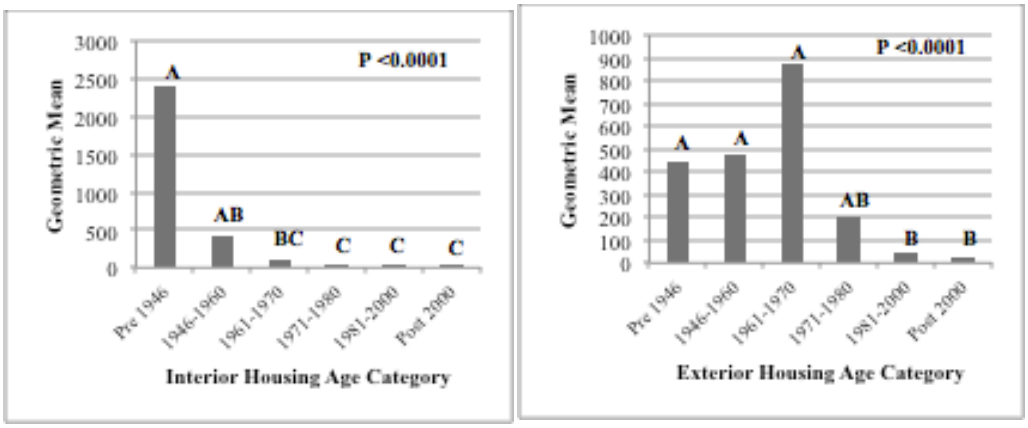

Figure 3.4: Geometric means (ppm) by housing age category for exterior and interior paint samples from the Student-Newman-Keuls (SNK) test for differences between geometric mean values by housing age cohorts at the 95% confidence level (α=0.05). Means associated with the same letter are not significantly different at the 95% confidence level (α=0.05).

Garden Produce Results

Above-ground and below-ground produce samples were collected from 34 study households and analyzed for lead content (ppm dw). Table 3.19 displays the summary statistics. Very few of the garden produce samples had detectable lead levels, with 10 samples (representing 11% of all samples) having values that were above the laboratory detection limit of 0.18 mg/kg dw. The below-ground samples had a slightly higher geometric mean (0.15 ppm) and larger range (n.d. - 0.39 ppm) compared to above-ground samples (GM 0.11 ppm; n.d. - 0.19 ppm). Due to the small number of samples with detectable lead concentrations, no further analyses were made to determine garden produce lead differences over time or based upon different preparation methods.

Table 3.19: Sample size (n), geometric mean (GM) with 95% confidence intervals (CI), median, 95th percentile with 95% CI, and the overall range of garden produce lead values by sample location in ppm.

Sample Type	n	GM (95% CI)	95th percentile (95% CI)	Range
Above-Ground	28	0.11 (0.10-0.12)	0.22 (0.17, 0.23)	n.d.–0.38
Below-Ground	6	0.15 (0.11-0.19)	0.34 (0.27, 0.44)	n.d.–0.44

Relationships between Lead Concentrations in Environmental Media

To determine the basic relationships between the measured lead levels in different environmental media sampled in this study, linear regression analyses were performed with selected media. All three dust locations were significantly correlated with interior paint. Floor and trough dust lead levels were significantly correlated with exterior paint. All three dust locations were also significantly correlated to soil samples from the dripline and play area. There were no significant correlations between lead levels in garden soil and lead in household dust. Lead levels from garden soil samples also showed no significant correlations with paint lead levels, while the rest of the soil samples did have a significant correlation with interior and exterior paint, with interior paint having a stronger correlation than exterior paint (Table 3.20).

Table 3.20: Correlation coefficients for geometric mean household lead concentrations in selected environmental media.

	Dust: Floor	Dust: Sill	Dust: Trough	Paint: Interior	Paint: Exterior
Paint: Interior	**0.20**	**0.04**	**0.22**		
Paint: Exterior	**0.08**	-0.01	**0.04**		
Soil: Dripline	**0.35**	**0.22**	**0.27**	**0.32**	**0.19**
Soil: Play Area	**0.30**	**0.21**	**0.24**	**0.34**	**0.10**
Soil: Garden	0.02	0.01	0.05	-0.03	-0.01

Note: Bolded values indicate a significant relationship (α=0.05).
Geometric mean for each house by sample location has been used to determine the correlation coefficients.

Relationships between Lead Concentrations in Environmental Media and Children's Blood

Linear regression analysis was also undertaken to evaluate relationships between blood lead levels and lead levels in the environmental media. Positive and significant relationships were found for all dust locations, play area soil and most of the water categories, although all were weak (Table 3.21).

Table 3.21: Correlation coefficients of blood lead levels (BLL) and environmental media results from study households.

Environmental Media	r
Dust: Floor	**0.13**
Dust: Window Sill	**0.12**
Dust: Window Trough	**0.06**
Paint: Interior	-0.00
Paint: Exterior	-0.01
Soil: Dripline	0.01
Soil: Play Area	**0.06**
Soil: Garden	0.02
Water: Kitchen Flush	**0.02**
Water: Kitchen Average Stagnation	**0.03**
Water: Kitchen Stagnation Level 1	**0.05**
Water: Kitchen Stagnation Level 2	**0.02**
Water: Kitchen Stagnation Level 3	0.01
Water: Kitchen Stagnation Level 4	**0.02**

Note: Bolded values indicate a significant relationship (α=0.05)

To examine the combined effects of environmental media lead, housing age categories and blood lead levels, a multiple linear regression analysis was used. Due to the data set having a number of missing values across households (e.g. no paint at some homes or no garden soil at others), the analysis could only be completed to the level of household averages for overall dust, soil, paint and flush and stagnation water based on discussion with the biomonitoring team. Housing age category was included in the model due to its level of importance in our study design. When using the regression analysis, the best predictive model was the one that included the housing age category for pre-1970 and post-1980, average household dust, average household soil and average household stagnation water data ($n = 168$, $F = 8.98$, $r^2 = 0.18$). A second model that included household averaged paint lead as an additional variable increased the r-squared value to 0.23. To further look into how well these variables were describing the relationship with

children's blood lead levels, a backward selection method was applied to the multiple linear regression. The model that included the household averaged paint lead was first run. Using this, only 128 of the total 194 households were included in the analysis, due to missing variables. Housing age was the first variable to be removed from the model, as it contributed less than 1% to the variance, followed by paint, soil, and water stagnation. Only household average dust was found to be a significant variable in the model at $p<0.0001$ with an r^2 value of 0.23. When the model was run leaving out household average paint, 168 of the total 194 households were included in the analysis. Soil lead was the first variable removed followed by housing age category. Both household averaged dust lead and water stagnation were left in the model with dust being a significant variable at $p<0.0001$ and stagnation water at $p=0.0448$ with an r^2 value of 0.17%.

Soil Bioaccessibility Results

There were 95 different samples collected from participating households of dripline or play area soil. These bioaccessibility samples were split from the overall soil sample collected at the chosen households based on computer randomization for samples in the pre-1970 and post-1980 categories and to have a similar number between the dripline and play area (Table 3.22).

Table 3.22: General numbers of samples collected for the bioaccessibility analysis from study homes throughout St. John's, NL.

	# Samples
Dripline	48 (22 post-1980 age group)
Play area	47 (14 post-1980 age group)
Pre-1970	59
Post-1980	36

Looking at the geometric mean values, it is evident that both lead measures are higher for the dripline versus play area samples and for the pre-1970 versus the post-1980 homes (Table 3.23). Furthermore, the percentage of near-total lead that is potentially bioaccessible is consistently high, averaging about 83%. Lead results from the water leach method could not be included in the analysis as the majority of the samples were undetectable for the methods used (method detection limit 0.9 ppm).

Table 3.23: Geometric mean values for the lead content and soil characteristics from household bioaccessibility analysis with 95% confidence intervals (CI).

Sample Area	CEC (meq/100gm)	pH	Organic Content (%)	Near-total Pb	Bioaccessible Pb		Grain Size <250 (μm)
				EPA 3050B (ppm)	1 M HNO_3 (ppm)	% bioaccessible	
Dripline	17.2 (8.1-45.1)	5.4 (4-6.9)	9.3 (2.1-16.9)	283.9 (13-1900)	223.2 (10-1410)	78.6	75.4 (30.76-99.86)
Play area	19.8 (2.7-34.1)	5.3 (4.1-6.8)	10.6 (1.2-17.4)	216.8 (19-900)	191.9 (14-736)	88.5	74.8 (22.65-98.5)
Pre-1970	19.3 (2.7-45.1)	5.4 (4-6.8)	9.8 (1.2-17.4)	368.8 (21-1900)	304.8 (16-1410)	82.6	73.1 (22.65-99.86)
Post-1980	17.2 (8.7-26.5)	5.3 (4.3-6.9)	10.2 (5.3-17.2)	57.2 (13-630)	48.5 (10-541)	84.8	78.5 (54.56-98.26)

To examine the objective of looking at bioaccessibility according to housing age (pre-1970 and post-1980), linear regression analyses and correlation analyses were undertaken. Tables 3.24 and 3.25 show the correlation coefficients for the EPA near-total method (for comparison) and the nitric acid bioaccessibility method, along with the soil characteristics.

Table 3.24: Correlation coefficients for bioaccessible (1 M HNO_3) and near-total (EPA 3050B) lead analyses and soil characteristics for pre-1970 soil.

	CEC (meq/100gm)	Grain Size <250µm	pH	Organic Content	CEC-Ca (meq/100 gm)	CEC-Mg (meq/100 gm)	CEC-K (meq/100 gm)	Grain Size – Sand (µm)	Grain Size – Silt (µm)	Grain Size – Clay (µm)
1M HNO_3	- 0.07	0.06	-0.01	0.14	0.01	**-0.27**	0.13	-0.13	0.07	**0.44**
EPA 3050B	-0.02	0.08	0.003	0.17	0.04	-0.23	0.15	-0.15	0.09	**0.45**

Note: Bolded values indicate a significant relationship (α=0.05)
N = 59

Table 3.25: Correlation coefficients for bioaccessible (1M HNO_3) and near-total (EPA 3050B) lead analyses and soil characteristics for post-1980 soil.

	CEC (meq/ 100gm)	Grain Size <250 µm	pH	Organic Content	CEC-Ca (meq/ 100gm)	CEC-Mg (meq/ 100gm)	CEC-K (meq/ 100gm)	Grain Size – Sand (µm)	Grain Size – Silt (µm)	Grain Size – Clay (µm)
1 M HNO_3	-0.15	0.05	-0.26	0.17	-0.11	-0.29	0.17	0.07	-0.03	-0.07
EPA 3050B	-0.19	0.07	-0.25	0.08	-0.13	-0.32	0.13	0.05	-0.01	-0.07

N = 36

In the pre-1970 soils, the CEC-Mg has a weak negative correlation and the clay content of grain size is positively correlated for the 1M HNO_3 extraction method (Table 3.24). Results for the pre-1970 soils near-total EPA extraction method are very similar to those for the HNO_3 extraction, with only clay content being positively correlated. When examining the post-1980 soils, all coefficients are low and there are no significant

correlations with either the 1M HNO_3 extraction method or near-total results (Table 3.25).

Table 3.26: Correlation coefficients for bioaccessibile (1M HNO_3) and near-total (EPA 3050B) analyses and soil characteristics for play area soil.

	CEC (meq/ 100gm)	Grain Size <250 µm	pH	Organic Content	CEC-Ca (meq/ 100gm)	CEC-Mg (meq/ 100gm)	CEC-K (meq/ 100gm)	Grain Size – Sand (µm)	Grain Size – Silt (µm)	Grain Size – Clay (µm)
1M HNO_3	0.06	-0.11	-0.12	0.23	0.08	-0.17	0.20	0.13	-0.12	0.28
EPA 3050B	0.10	-0.08	-0.12	0.25	0.11	-0.15	0.23	0.10	-0.10	0.29

N = 47

Table 3.27: Correlation coefficients for bioaccessibile (1M HNO_3) and near-total (EPA 3050B) analyses and soil characteristics for dripline soil.

	CEC (meq/ 100gm)	Grain Size <250 µm	pH	Organic Content	CEC-Ca (meq/ 100gm)	CEC-Mg (meq/ 100gm)	CEC-K (meq/ 100gm)	Grain Size – Sand (µm)	Grain Size – Silt (µm)	Grain Size – Clay (µm)
1M HNO_3	-0.05	-0.06	0.12	-0.09	0.14	**-0.33**	**0.29**	-0.18	0.05	**0.47**
EPA 3050B	-0.05	-0.06	0.12	-0.09	0.13	**-0.34**	0.27	-0.19	0.06	**0.47**

Note: Bolded values indicate a significant relationship (α=0.05)
N = 48

For the play area analyses, all the correlation coefficients are low, and there is insufficient evidence to conclude that there are any significant linear relationships present (Table 3.26). For the dripline correlation analyses, the 1M HNO_3 bioaccessibility method, CEC-

K was weakly positively correlated while grain size-clay was moderately positively correlated, and the CEC-Mg was weakly negatively correlated. Results for the EPA near-total method for dripline soils are very similar to those for the 1M HNO_3 extraction. The CEC-Mg had a weakly negative correlation and grain size-clay was moderately positively correlated (Table 3.27).

In contrast to the above-mentioned relatively weak relationships, Pearson's correlation shows a clear positive relationship between the EPA near-total lead concentrations and the amount that is bioaccessible, for the 1M HNO_3 analysis (Table 3.28).

Table 3.28: Pearson correlation coefficients for bioaccessibility (1 M HNO_3 method) analyses to total (EPA 3050B method) lead concentrations.

	Pearson Correlation	p-value
Pre-1970 HNO_3	0.99	<0.0001
Post-1980 HNO_3	0.98	<0.0001
Dripline HNO_3	0.99	<0.0001
Play area HNO_3	0.99	<0.0001

Chapter 4: Discussion

The findings of this study suggest that lead levels in St. John's, NL are indeed higher in and around pre-1970's housing. A significant difference is detected for dust, soil, water post stagnation, and paint when cohort groups were analyzed for pre-1970 versus post-1980 homes. While none of the GM dust lead loadings for the 3 locations exceeded the previous US EPA guidelines or current US EPA guidelines (10 μg/ft^2 for floors, 100 μg/ft^2 for windowsills and 400 μg/ft^2 for window troughs (EPA, 2020)), there were homes that did exceed these values. The Pre-1970 and particularly pre-1946 housing age cohorts have houses in them that do exceed these guideline values, with the upper ranges being much higher than the US EPA guidelines recommend. The geometric mean values for dust loading for each location are lower than what has been found in a large systematic review done by Frank *et al.* (2019) for lead results in the US from 1996-2016. The older housing age categories having higher values is consistent with The Canadian House Dust Study by Rasmussen *et al.* (2013), which found older housing age categories had higher dust lead concentrations. The findings also suggest that lead contaminated house dust is the major exposure source of lead influencing children's blood lead levels. These findings are in-line with other published studies including a large pooled epidemiological study by Lanphear *et al.* (1998a) which found that dust was the strongest predictor of children's blood lead levels and others which found dust to be a large contributor to children's blood lead levels (von Lindern *et al.,* 2003b and Yiin *et al.,* 2000, Safruk *et al.*, 2017, Braun *et al.* 2021).

The geometric mean soil results in this study do not exceed the current soil-lead guideline for residential areas in Canada set at 140 ppm (CCME, 1999) or the current US EPA level of 400 ppm for children's play areas (US EPA, 2011b). The highest values were seen in the dripline area with a GM of 103 ppm, however sampling did reveal that there are a number of households in the pre-1970s cohort that have elevated soil-lead levels. The highest value was 6800 ppm for the dripline at one household, 1900 ppm for the play area at another household and 480 ppm for the garden area in this pre-1970 cohort. This is important to note for the city of St. John's, as multiple previous studies have found a positive correlation with higher soil lead levels leading to elevated blood lead levels (Lewin *et al.* 1999, Lanphear *et al*. 2000, and Safruk *et al.* 2017).

The water sample results show that the geometric means calculated do not exceed the MAC recommended by Health Canada in 2019 of 0.005mg/L (Health Canada, 2019); however the ranges show that there are some households in the pre-1946 category that do exceed this amount. There was also very little difference between the flush and stagnation categories per housing age group, with the exception of Stagnation level 1 being significantly different between our pre-1970 and post-1980 samples. The household average water stagnation samples taken after a 30-minute period are also suggested to be a significant exposure source, but only when the regression model with the most households was included. Lanphear *et al.,* (1998b) found that blood lead levels correlated with water lead and other researchers have noted that lead service lines are a risk factor for elevated blood lead (Miranda *et al.,* 2007 and Brown *et al.,* 2011).

Correlation analysis however allowed a look into the relationships between children's blood lead levels and environmental media levels, as well as, a closer examination of the relationship between lead levels in different environmental media to see the effect of sample location. Specifically, the correlation analysis between environmental media revealed that lead levels in interior paint had a weak correlation with the lead concentrations in dust at all locations as compared to exterior paint, which would be expected since it was interior dust that was sampled. Previous research has indicated that the lead in household dust may be from lead contaminated paint sources (Tong and Lam, 2000 and Lanphear and Roghmann, 1997). There was also a weak relationship between lead in house dust and soil lead contents, particularly the floor dust and dripline soil lead. This relationship follows on evidence from other studies indicating a strong relationship between interior dust lead and soil lead (Tong and Lam, 2000; Lambert and Lane, 2004; von Lindern *et al.*, 2003a, Safruk *et al., 2017*). There was a weak relationship between interior paint and dripline soil and play area soil, which suggests that perhaps interior paint was used to paint the exterior of homes. There is evidence of a relationship between paint lead and soil lead due to it chipping off into the soil (Lanphear and Roghmann, 1997).

While dust and water stagnation lead levels were shown to have a weak relationship through a multiple regression analysis with children's blood lead levels, correlation analysis allowed a more in-depth evaluation of the relative importance of the locations sampled within study households. Specifically, correlation analysis showed that lead in dust from the floors and the windowsills were weakly correlated relative to that of lead in dust from the window troughs. Moreover, window trough samples had much higher

geometric mean lead levels, 29 times higher than the floors. The results show that lead in dust from the floors has the strongest relationship to blood lead levels, and, are in line with previous results (Dixon *et al.* (2009), Safruk *et al.* (2017), and Braun *et al.* (2021), all who found that floor dust samples best predicted children's blood lead levels. The correlation between the water stagnation levels and children's blood lead levels was significant but weak.

The investigation of the relationship between children's blood lead levels and housing age was one of the main study objectives, as the early pilot studies in St. John's found that older homes had higher lead levels in dust and soil (Bell *et al.*, 2010). Housing age alone was not found to be a significant contributor to children's blood lead levels, but it was found to have a significant relationship with the environmental media samples. There was a significant difference between environmental lead levels in dust, soils and paint for pre-1970 versus post-1980 homes. This was expected due to lead in paint largely being phased out in the 1960's (CMHC, 2004). Leaded gasoline use decreased throughout the 1970's and was prohibited in Canada in 1990 (Health Canada, 2009), all of which would have led to decreasing lead-bearing particulate matter in soils over time.

The housing age category of pre-1970 had a weak relationship with children's blood lead levels. This is consistent with the Canadian House Dust Study by Rasmussen *et al.* (2013) that found higher dust lead loadings in older homes and Safruk *et al.* (2017) that found higher blood lead levels in older age housing categories. The strongest predictors in this study were household dust and water stagnation. This is again consistent with the

Canadian House Dust Study by Rasmussen *et al.* (2013) and Braun *et al.* (2021).

One of the limitations with this study were that sample size was relatively small, and it was very difficult to obtain samples of all environmental media at each household. Due to this, the number of households included in the full multiple regression analysis was small, which may be part of the reason why it is difficult to discern a definitive relationship with housing age categories and blood lead levels.

Bioaccessibility

This study shows that there are elevated levels of lead in residential properties throughout St. John's. These high levels are most likely a result of anthropogenic deposition over the years. The near-total lead concentrations (EPA 3050B) exceeded the CCME guideline of 140 ppm in all housing age categories except for those from the post-1980 housing age category. None of the sample populations exceeded the US EPA guideline for play area samples of 400 ppm. These high levels of total lead may represent an exposure risk for human health, as studies have found links between human health problems and lead in soil (Bierkens *et al.,* 2012; Isaac *et al.,* 2012), particularly because this study shows a high correlation between the amount of lead in the soil and how much is potentially bioaccessible.

Analysis showed that the 1 M HNO_3 bioaccessibility method had a strong positive correlation to near-total lead concentrations (EPA 3050B). This is consistent with what other studies have found (Minca, 2012). Minca et al. (2012) concluded that the 1M HNO_3 bioaccessible analysis could be used to estimate bioaccessible soil lead when compared to a relative bioaccessibility leaching procedure developed by Drexler and Brattin (2007).

This Drexler method is the physiologically based leach procedure of a pH around 1.5 and is the basis of the US EPA 2008's standard operating procedure used in this study. This could account for the finding of similar correlations to total lead concentration.

When examining bioaccessibility by housing age category, the results show that the pre-1970 concentrations using the nitric acid method are higher than the post-1980 results. This was expected as older housing has been exposed to the lead products such as gasoline, paint and coal combustion over the years while the newer homes would not have the extent of this exposure.

When examining the relationship of bioaccessible lead by dripline location and play area location, analysis showed that the dripline concentrations were higher for the nitric acid method as compared to the play area. This too was expected as the dripline represents an area where lead has been deposited over the years from weathering of exterior surfaces coated with leaded paint.

Looking at the soil factors that were significantly correlated, in the dripline analyses, the nitric acid method had a relatively weak negative correlation with the Mg component of the cation exchange capacity, a relatively weak positive correlation with the K component and a stronger positive correlation with the clay portion of the soil. For the pre-1970 housing soil, there was also a somewhat weak negative relationship between results for the HNO_3 method and the Mg component of the cation exchange capacity and a stronger positive correlation with the clay-size fraction. The clay aspect being significantly correlated follows with results from other studies looking at lead behavior

(Elless *et al.,* 2007, Juhasz *et al* 2011, Yan *et al.* 2019) that found that decreasing particle size was correlated with increased lead bioaccessibility.

Looking at the other soil characteristics included in this study, pH was not found to be a significant predictor of bioaccessibility. Miguel *et al* (2012) also found that soil pH did not influence bioaccessibility to any significant extent. This is in contrast to other studies (Yang *et al.,* 2003; Cao *et al.,* 2008 and Saminathan *et al.,* 2010) that found an increasing pH of the soil decreased lead mobility and bioavailability. This study also showed no significant correlation with the organic content of the soil, again in contrast to other studies that have showed soils amended with humus materials have decreased extraction of lead (Yang *et al.,* 2006, Sipos *et al.,* 2005). Cation exchange capacity was shown to have a significant correlation when broken into its components of Mg and K, however coefficients for overall CEC were weak and non-significant. Mg and K components of the CEC both were significantly correlated with the nitric acid bioaccessibility method for the dripline soils, although the magnitude of the coefficients was not large. The lack of a significant relationship to overall CEC contrasts with results of previous studies that have found CEC to be a significant predictor of lead bioaccessibility (e.g., Saminathan *et al.*, 2010 and Yan *et al.* 2019). Saminathan *et al.* (2010) found that the larger the CEC the greater the retention for lead. It is likely that our small sample size has contributed to the discrepancies between our results and other published results, and it is something to possibly analyze further on a larger scale in the future.

Chapter 5: Conclusion

There are elevated levels of lead in residential properties throughout St. John's that may represent an exposure risk to human health, particularly in the older housing age cohort of pre-1970. None of the guidelines for environmental media from Canada or the US EPA were exceeded in this study in the statistical analysis. However, the raw data does show a number of houses in the oldest housing age cohort (pre-1946) have lead levels above the recommended guidelines for dust, soil and tap water.

Examination of BLLs and environmental media showed that floor dust had a weakly positive correlation ($r = 0.13$) to BLLs, and there was a weaker but still significant relationship for our water stagnation levels 1,2 and 4 to BLLs ($r <= 0.05$). The housing age category of pre-1970 also had a weak relationship with children's BLLs.

Bioaccessibility analysis showed a strong positive correlation between the 1 M HNO_3 and near-total lead analysis (EPA 3050B), and the soil factors there were weak correlations for the Mg component of CEC, K and clay size.

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Appendix

Table A.1: Quality Control (QC) of environmental media analysis from Maxxam Analytics laboratories. Range, mean and median values are listed respectively under each media type and heading except for # QC batches and method blank value recovery.

Environmental Media	# QC Batches	QC Recovery (%)[1]	Matrix Spike Recovery (%)[2]	Spiked Blank Recovery (%)	Method Blank Value[3]	Method Blank Value Recovery (µg)
Dust	136	78 – 110 97.7 98	NA	76 – 107 96.6 98	0.204 – 1.34 0.4 0.35	0.125 (all batches)
Soil	41	90 – 111 100.9 101	80 – 105 92.2 92	83 – 111 97.6 98	1	NA
Water	56	NA	92 – 113 100.7 101	93 – 110 101.2 102	0.017 – 0.25 0.097 0.024	0.01 (4 batches) 0.1 (50 batches) 1.0 (2 batches)
Paint	36	94 – 104 100.0 100	85 – 99 91.2 91.5	85 – 99 92.4 92.5	53 - 82	50 (35 batches) 100 (1 batch)
Garden Produce	12	44 – 61 51.7 52	77 – 112 95.8 95	92 – 111 100.3 100.5	ND	NA

Note: [1]Garden produce had 8 batches with a QC standard recovery percentage.
[2] Soil had 4 batches with a matrix spike recovery percentage, paint had 16 batches with a matrix spike recovery percentage, and garden produce had 11 batches with a matrix spike recovery percentage.
[3]Dust had 13 batches with a method blank value, soil had 1 batch with a method blank value, water had 3 batches with a method blank value, and paint had 2 batches with a method blank value
NA – Not Applicable
ND – Not Detectable

Method blank is indicative of potential contamination. These are processed and carried throughout the entire sample preparation and analytical processes.
Matrix spikes and spiked blanks are used to determine precision and bias. They are split samples spiked with identical concentrations of the analyte of interest. The spiking occurs prior to sample preparation and analysis.
For soil, water, paint and garden produce, matrix spikes and method blanks were analyzed at a minimum frequency of 5%.
For dust method blanks and spiked blanks were analyzed at a frequency of 1 for every 20 samples with a minimum of 1 for each digestion batch.