Soil Ingestion Rate and Excess Lifetime Cancer Risk in First Nations' People Exposed to Polycyclic Aromatic Hydrocarbons Near In-situ Bitumen Extraction in Cold Lake, Alberta

Graham Irvine

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Abstract

The inadvertent ingestion of contaminated soil is the dominant exposure route of non-volatile and semi-volatile contaminants such as polycyclic aromatic hydrocarbons (PAHs). Quantitative mass balance soil ingestion studies have been used to determine soil ingestion rates for use in human health risk assessments (HHRA) that can be used to predict the likelihood of adverse effects in individuals exposed to hazardous contaminants such as PAHs in contaminated soil. The Cold Lake region of Alberta is one of the three major oil sands regions of Alberta, and PAH concentrations in this oil sand region may be elevated in the atmosphere and the soil, resulting in increased exposures to PAHs. The area is home to Cold Lake First Nation who practice traditional activities and lifestyles that may put them in greater contact with soil than previous soil ingestion studies suggest.

The primary objective of this research was to assess the soil ingestion rate in a group of First Nations subjects inhabiting the Cold lake region, and assess the carcinogenic risk posed by exposures to PAHs in air and soil. The study employed a quantitative mass balance tracer approach to estimate soil ingestion rates, and followed 9 subjects over a 13 day period. Soil and air samples were simultaneously collected to assess PAH contamination.

The mean soil ingestion rate using Al and Si elemental tracers was 52 mg d^{-1}, with a 90th percentile of 220 mg d^{-1}, and a median soil ingestion rate of 37 mg d^{-1}. These values are greater than the soil ingestion rates for HHRA recommended by Health Canada. The mean increase in excess lifetime cancer risk posed by inadvertent ingestion of soil to a First Nations' individuals following traditional activities was 0.02 cases per 100,000 people with a 95% risk level of 0.067 cases per 100,000 people. Exposure to PAHs through inhalation posed a maximum lifetime cancer risk below 0.1 cases per 100,000, people. Thus, this study found no appreciable increase in excess lifetime associated with PAH exposure of First Nations' people in the Cold Lake region.

Résumé

L’ingestion involontaire de terre contaminée est la principale source d’exposition à plusieurs contaminants semi et non volatils, comme les hydrocarbures aromatiques polycycliques (HAPs). Des études précédentes sur l’ingestion de terre utilisant la quantification par bilan de masse ont étés effectuées pour déterminer les taux d’ingestion qui figurent dans les rapports d’évaluations de risques pour la santé humaine (HHRA), de façon à prédire la probabilité d’effets négatifs chez les individus exposés à des contaminants dangereux comme les HAP dans les sols. La région du Cold Lake en Alberta est une des trois principales régions de sables bitumineux de cette province où les HAP sont retrouvés dans l’air ainsi que dans les sols, ce qui résulte en une exposition plus importante aux HAP. Cette région est habitée par les premières nations qui possèdent un style de vie traditionnel et pratiquent des activités qui lui sont relié. Ces activités traditionnelles ont pour conséquence d’augmenter la fréquence du contact des individus avec le sol en comparaison à ce que les études précédentes suggèrent.

L’objectif principal de cette recherche était d’évaluer le taux d’ingestion de terre chez un groupe de sujets de cette communauté, et d’évaluer les risques de carcinogénicité reliés la présence de HAP dans l’air et le sol. Cette étude suit 9 sujets durant 13 jours et applique une approche de quantification par bilan de masse par traceur pour estimer les taux d’ingestion de terre. Les échantillons de sol et d’air ont étés collectés simultanément pour évaluer la contamination par HAP.

La moyenne des taux d’ingestion de terre tracés par Al et Si était de 52 mg d^{-1}, avec un $90^{\text{ième}}$ percentile de 220 mg d^{-1}, et une médiane de 37 mg d^{-1}. Ces valeurs sont plus élevées que celles qui sont recommandées pour HHRA par Santé Canada. L’augmentation de la moyenne du risque de cancer à vie posé par ingestion de terre involontaire aux individus qui pratiquent des activités traditionnelles était de 0.02 cas par 100 000 personnes avec un niveau de risque à 95% de 0.067 cas par 100 000 personnes. L’exposition aux HAP par inhalation posait un risque de cancer maximum à vie en dessous de 0.1 cas par 100 000 personnes. Cette étude n’a pas trouvée d’augmentation significative dans le risque à vie associé avec l’exposition par HAP pour les personnes pratiquant le style de vie des premières nations dans la région de Cold Lake.

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Glossary

ANOVA	Analysis of variance
ASE	Accelerated solvent extraction
AT	Averaging time (days)
BaP	Benzo[a]pyrene, a carcinogenic polycyclic aromatic hydrocarbon, often used as an indicator spices
Bitumen	heavy oil
BTM	Best tracer method
BW	Body weight (kg)
CCME	Canadian council of ministers of the environment
CF	Conversion factor (10^{-6})
Cs	Concentration
CSF	Cancer slope factor
CV	Coefficient of variability
ED	Exposure duration (yr)
EF	Exposure frequency (d yr^{-1})
F/S ratio	Food to soil ratio
GFF	Glass fibre filter
HHRA	Human health risk assessment
In-situ extraction	Bitumen extraction method using of steam for oil sands >100 m below the surface
IR	Intake rate
IR_{air}	Inhalation rate used for risk assessment calculation
IR_{soil}	Inadvertent soil ingestion rate used for risk assessment calculation
Oil sands	Naturally occurring mixture of sand, clay, water and bitumen

PAH	Polycyclic aromatic hydrocarbons
PEF	Potency equivalency factors
PDF	Probability density functions
PUF cartridge	Polyurethane form cartridge
SD	Standard deviation
SE	Standard error

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Chapter 1 - Introduction

1.1 Human health risk assessment of contaminated sites

A site is considered a contaminated site if the concentration of a substance is elevated above background levels to the point that there is risk or the potential of risk of acute or long-term adverse health effects to humans or the environment (Treasury Board of Canada Secretariat 2013). The cause and size of a contaminated site can vary considerably, from abandoned mines contaminated by heavy metals to small oil spills that contaminate soil to large underground oil tanks (Office of the Auditor General of Canada 2012). In Canada there are more than 22,000 federal contaminated sites with many of the larger sites located in the northern territories, Labrador and southwestern British Columbia. The smaller contaminated sites are mostly concentrated along the Canadian-United States border in British Columbia and Saskatchewan, as well as the Atlantic provinces (Office of the Auditor General of Canada 2012). Petroleum-based products are the most common type of contaminant and over 50% of federal contaminated sites have contaminated soils, the remaining sites are classified as having contaminated water, sediment, air or some other form of contamination (Office of the Auditor General of Canada 2012). The large number and variety of contaminated sites in Canada necessitate the need to properly assess the risk of adverse health effects to people on contaminated sites.

Human health risk assessments (HHRA) are used provincially within Canada, and on a national and international level to assess the risk of adverse health effects from exposure to a contaminated medium. A risk assessment consists of four basic steps: hazard identification; dose-response assessment; exposure assessment; and risk characterization (U.S. EPA 2012). Hazard identification is the process of determining if exposure to a contaminant or contaminants could cause an adverse health effect to humans, such as increased incidence of cancer. A dose-response assessment is then conducted through review of available data, to determine at what dose there is a risk of increased adverse health effects from a contaminant. The exposure assessment calculates the dose of a contaminant that a population is exposed to through different receptor pathways and environmental sources. An exposure assessment requires knowledge of the concentration of a contaminant in a medium and the intake rate for the exposure pathway of a contaminant to a population. Soil ingestion is the dominant pathway for non-volatile and semi-volatile contaminants on contaminated sites and as a result soil ingestion rates are essential parts of exposure assessments to be able to calculate the intake rate of a contaminant. Combining the dose-response assessment and the exposure assessment, the risk of excess adverse health effects due to exposure to a contaminant can then be determined.

1.2 *Soil ingestion*

The inadvertent ingestion of soil is the dominant exposure pathway for non-volatile and semi-volatile contaminants to humans from contaminated sites (CCME 2006). Soil ingestion rates are consequently an essential part of HHRAs. Soil ingestion referred to herein is the inadvertent ingestion of soil that can occur through soil particles that adhere to hands, food or other objects and are then ingested, but does not include the purposeful ingestion of soil through practices such as soil pica or geophagy. Recommended soil ingestion rates used for HHRA in Canada are 20 mg d^{-1} for adults and 80 mg d^{-1} for children, which differ from the HHRA soil ingestion recommended rates in the United States of 50 mg d^{-1} and 100 mg d^{-1} for adults and children respectively. The recommended soil ingestion rates for use in HHRA are derived from only a handful of mass balance soil ingestion studies that mostly focused on children (Calabrese et al. 1989; 1997; Davis et al. 1990; van Wijnen et al. 1990). A mass balance soil ingestion study employs the use of elemental soil tracers that are relatively high concentrations in soil such as Al, Si, and Ti. These elemental tracers are measured where a subject spends the majority of their time as well as in their food and feces. Using mass balance equations, the soil ingestion rate of a subject can be determined (Calabrese et al. 1989; 1997; Davis et al. 1990**)**. However, soil ingestion has been a relatively small area of research with a focus mostly on children in suburban or urban areas in the United States. There has been little attempt to understand the effects of urbanization, season, lifestyle, or a myriad of other possible contributors to the inadvertent ingestion of soil. Qualitative estimates have suggested that an aboriginal traditional lifestyle may result in higher soil ingestion rates than the current recommended rates used for HHRA (Harper et al. 2007), but the first aboriginal soil ingestion study found soil ingestion rates were lower than the qualitative estimated rates, though higher than recommended rates for HHRA (Doyle et al. 2012).

Traditional activities practiced by aboriginal people in Canada are not only an important part of their cultural identity, but for many are also sources of economic well-being. Many traditional activities such as hunting; gathering foods and medicines; subsistence agriculture; and the drying or smoking of meats put aboriginal people in close contact with soil, and could lead to greater rates of inadvertent soil ingestion. The current Health Canada recommended soil ingestion rates for use in HHRA have not been developed with these traditional activities in mind, which may lead to increased exposure to contaminants as a result of a different soil ingestion rate (Wilson Scientific Consulting Inc. and Meridian Environmental Inc. 2006).

1.3 *Polycyclic aromatic hydrocarbons*

Polycyclic aromatic hydrocarbons (PAHs) are a class of ubiquitous environmental contaminants consisting of two or more fused benzene rings (Boström et al. 2002; Government of Canada et al. 1994). The structure and molecular weight of a PAH affects its chemistry, ranging from semi-volatile to non-volatile, with a large range of water solubilities, and molecular weights (Boström et al. 2002; Lima et al. 2005). An important property of PAHs is their ability to adsorb onto particulate matter, both in water and in air because of their lipophilicity (Government of Canada et al. 1994). PAHs consist of more than 100 identified individual compounds, all with varying properties.

The majority of PAHs that enter the environment are the result of incomplete combustion. In Canada, forest fires are the dominant source of environmental PAHs, but many industrial processes such as aluminum smelting and the upgrading and refining of oil products release significant amounts of combustion derived PAHs (Government of Canada 1994; Lima et al. 2005). PAHs are also present naturally in fossil fuels such as oil. The formation of these petrogenic PAHs through diagenesis favours the formation of alkyl PAHs, differing from pyrogenic PAHs that typically have a much higher percentage of unsubstituted PAHs (Lima et al. 2005). The temperature of combustion is related to the formation of alkylated PAH compounds, with higher temperatures resulting in lesser-alkylated or unsubstituted PAH compounds (CCME 2010). Some plant and microbial processes can also result in the formation of PAHs.

Many PAHs are known carcinogens, mutagens and teratogens. The toxicity of a PAH is closely linked to both the size and structure of a PAH and larger 4-6 ringed PAHs with a bay or fjord region are often more carcinogenic than smaller PAHs. Benzo[a]pyrene (BaP), a 5-ringed PAH is a known carcinogen and the most well-studied PAH (Boström et al. 2002; Choate et al. 2006; Government of Canada et al. 1994). Although there are some acute toxic effects as a result of exposure to PAHs, soil and air quality guidelines have been developed to account for carcinogenic and chronic effects from PAH exposure (CCME 2006). Unfortunately, data is limited for many alkyl-PAH compounds, as until recently, research on carcinogenic activity from PAHs has been focused on the 16 priority PAHs (Bojes and Pope 2007; CCME 2010; Government of Canada et al. 1994).

1.4 *Oil sands and lifetime cancer risk assessment*

The Athabasca region of Alberta is Canada's largest source of heavy oil (bitumen), with the majority of the bitumen extracted in the Athabasca by open pit mining techniques (Burrowes et al. 2011). The Athabasca region is the largest scale oil sands industrial operation in Canada and has open pit mining, *in-situ* bitumen extraction, and bitumen upgrading and refining facilities. There are two methods of bitumen extraction in the oil sands, open pit mining or *in*-situ bitumen extraction. Open pit mining involves the use of large construction vehicles to shovel oil sands into large trucks that deliver it to a crusher to physically break down the size of the oil sands before it is transported to an upgrading facility. Open pit mining is used when the oil sands are less than 70 m below the surface. If the oil sands are deeper than 100 m, *in-situ* extraction is used instead. All *in-situ* extraction methods involve the injection of steam into underground oil reservoirs to heat up the oil sands and reduce bitumen viscosity. The heated bitumen is then pumped out of the reservoir and transported to an upgrader. Oil sands are composed of mostly water, sands and clays and only ~10-12% of an oil sands particle is bitumen. Before bitumen is refined into a petroleum product it is sent to upgraders to remove impurities. The bitumen in the Athabasca region is very close to the surface and in many areas has been exposed due to erosion from the Athabasca River and its tributaries leading to a high natural background concentration of PAHs. Additionally, PAH concentrations have further increased due to the oil sands industry in the region (Akre et al. 2004; Headley et al. 2001, 2002; Kelly et al. 2009). Cold Lake region, the focus of this study, is the largest *in-situ* thermal bitumen extraction operation in the world, but lacks the upgraders, refineries and open pit mining extraction of bitumen (Burrowes et al. 2011). Bitumen extracted in the Cold Lake region is transported to Lloydminsiter, Alberta or Edmonton, Alberta for upgrading and refining.

A human health risk assessment can be used to assess the lifetime cancer risk of a population from exposure to a contaminant. Many PAHs are known carcinogens and the carcinogenicity of a mixture of PAHs is measured using potency equivalency factors (PEF) in relation to BaP. There has been a large amount of research conducted on BaP and the carcinogenicity of BaP has been known as far back as the 1930s. The carcinogenicity of BaP has been well demonstrated through laboratory studies (Boström et al. 2002). The PEF for BaP is set at 1, and the additional 15 priority PAHs are assigned a PEF depending on its carcinogenicity in comparison to BaP. The concentration of each measured PAH is multiplied by its PEF and the final concentration of a mixture is expressed as a PAH concentration in BaP equivalents to determine carcinogenic risk due to exposure to a given contaminated medium. The PEF used for this study were developed by the US EPA and presented in Schneider et al. (2002).

Lifetime cancer risk is assessed by calculating the dose a population receives as a result of exposure to a given contaminant through a particular pathway. In this thesis, the dose of PAH was calculated through both inhalation from atmospheric exposure and soil ingestion. The dose is then multiplied by the inhalation or oral cancer slope factor for BaP depending on the pathway being calculated to determine excess lifetime cancer risk from the exposure to PAHs.

For this thesis, exposure was assessed through the development of a soil ingestion rate and measured PAH concentrations from atmospheric and soil samples collected simultaneously to the to the soil ingestion study. The soil ingestion study informed the soil intake or ingestion rate (IR_{soil}) in combination with recommended values from Health Canada for inhalation rate (IR_{air}). The dose that the population in the Cold Lake region receives can then be used to calculate the lifetime cancer risk to the population. This will be used for risk characterization to communicate the cancer risk to the First Nations' people following traditional lifestyles.

1.5 *Research objectives and hypotheses*

The main purpose of this research is to determine if a First Nations' population inhabiting an oil sands extraction region of Alberta, who engage in traditional activities, is at an increased risk of exposure to PAHs due to elevated soil ingestion and their proximity to oil sands industry, and if this population has an increased lifetime cancer risk as a result of exposure to PAHs. This research consisted of both a soil ingestion study and a concurrent environmental sampling study.

1.5.1 Soil ingestion study

The first objective was to determine the soil ingestion rate for a First Nations' population practicing traditional activities in the Cold Lake region of Alberta, Canada. Based on the previous aboriginal soil ingestion study it was hypothesized that that soil ingestion rates in people that practice traditional activities is greater than the 20 mg d^{-1} rate recommended to be used for HHRA by Health Canada.

1.5.2 Exposure and risk assessment study

The second objective was to determine if there is an increased lifetime cancer risk as a result of exposure to PAHs in the Cold Lake region. It was hypothesized that risk of cancer is greater than background because of the potential exposure to contaminants from oil sands operations.

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Chapter 2 - Soil ingestion to First Nations' people practicing traditional activities in Cold Lake, Alberta

2.1 Abstract

The inadvertent ingestion of contaminated soil is the dominant exposure route on contaminated sites of many non-volatile and semi-volatile contaminants to humans. Few soil ingestion studies have been conducted to date to develop recommended soil ingestion values for human health risk assessments (HHRA). Almost all of the soil ingestion studies used for HHRA have been done on children in suburban/urban environments. There has been only a single soil ingestion study done on aboriginal people practicing traditional activities. This study employed a quantitative mass balance tracer approach on aboriginal people practicing traditional activities to estimate soil ingestion in people in a wilderness area. This study followed 9 subjects over a 13 day period in Cold Lake, Alberta the largest *in situ* thermal heavy oil (bitumen) extraction operation in the world. The mean soil ingestion rate in this study using Al and Si tracers was 52 mg d^{-1}, with a 90th percentile of 220 mg d^{-1} and median soil ingestion rate of 37 mg d^{-1}. These soil ingestion values are greater than the recommended soil ingestion rates for HHRA from Health Canada, and are similar to soil ingestion estimates found in the only other study on aboriginal people.

2.2 Introduction

In North America there are over 60,000 contaminated sites, and Canada alone has more than 22,000 sites that have been identified in the Federal Contaminated Sites Inventory (White and Claxton 2004). Of these 22,000 sites in Canada, 51.9% contain contaminated soils, by far the most impacted medium (Office of the Auditor General of Canada 2012). Human Health Risk Assessments (HHRA) are used to evaluate the potential hazards of soil at contaminated sites, and subsequently, to prioritize sites for remediation and rehabilitation. Assessment of contaminant exposure through inadvertent soil ingestion is a key part of contaminated site HHRA, since ingestion is generally regarded as the dominant pathway on contaminated sites for exposure to non-volatile and semi-volatile contaminants (CCME 1996).

Soil ingestion studies are designed to estimate the amount of soil a population inadvertently ingests, and the values generated are employed in HHRA. Early soil ingestion studies were qualitative or semi-quantitative, and used observation and semi-quantitative analyses to estimate soil ingestion rates in children based on the frequency of hand-to-mouth contact and the amount of soil on a subject's hands (Day et al. 1975; Duggan and Williams 1977; Hawley 1985; Kimbrough et al. 1984; Lepow et al. 1974). Improved quantitative techniques that were subsequently developed employed elemental tracers such as Al, Si, and Ti, and mass balance analyses, to determine ingestion rate. However, the concentrations of these tracers in a subject's feces were assumed to be determined by ingestion of soil particles, and elemental uptake via food and other sources was not considered (Binder et al. 1986; Clausing et al. 1987; Doyle et al. 2010). The resulting soil ingestion estimates were highly variable and ranged from a few mg d^{-1} to upwards of multiple g d^{-1}. Later studies employed improved techniques that monitored a suite of elemental tracers measured in both the soil near the participant's dwelling or school, as well as in food. The most common soil ingestion study are mass balance soil ingestion studies that employ elemental tracers to determine soil ingestion by measuring the concentration of the tracer in soil, and subsequently in the food and feces of a series of subjects followed for a set period of time (Calabrese et al. 1989, 1997; Davis et al. 1990).

There have been only a small amount of these mass balance studies conducted to date, and most focused on children (Calabrese et al. 1989, 1997; Davis et al. 1990; Stanek and Calabrese 1995; van Wijnen et al. 1990). A few studies investigated adults (Calabrese et al. 1990; Davis and Mirick 2006; Stanek et al. 1997). These studies focused primarily on individuals inhabiting suburban and urban areas; consequently, the soil ingestion rate values obtained may not be representative of people practicing a rural or wilderness lifestyle (Harper et al. 2007). These studies are summarized in EPA (2011) and Doyle (2010). The soil ingestion rates employed by Health Canada for use in HHRA are 80 mg d^{-1} and 20 mg d^{-1} for toddlers and adults, respectively, while the EPA uses 100 mg d^{-1} for toddlers and 50 mg d^{-1} for adults (CCME 1996; Health Canada 2012; EPA 2011).

A recent study by Doyle et al. (2012) was the first soil ingestion study that investigated inhabitants of a wilderness areas, and the first to quantify soil ingestion rate in First Nations people practicing a traditional lifestyle. Doyle et al. (2012) recently reported a mean soil ingestion rate of 75 mg d^{-1} (90th percentile of 193 mg d^{-1}) for First Nations' people in central British Columbia who practice a traditional wilderness lifestyle. This value is far less than the Harper et al. (2007) qualitative estimate of 400 mg d^{-1} that was recommended previously, but greater than rates currently recommended by Health Canada and the US EPA.

Currently, there is a lot of speculation in the general media and public sentiment about the contamination to the environment from the Alberta oil sands, but research continues to send mixed messages about the severity of contamination and whether metals and polycyclic aromatic hydrocarbons (PAH) are rising as a result of extraction and upgrading activities (Hall et al. 2012; Kelly et al. 2009, 2010; Kurek et al. 2013; Wiklund et al. 2012). Many aboriginal people that follow traditional lifestyles live or are active in close proximity to oil sands operations, metal refining and processing, precious gem extraction and processing, and other potentially contaminated sites. The current Health Canada and US EPA soil ingestion rates used in HHRA estimates may not adequately protect aboriginal people living in these regions from elevated exposure.

This study employed a quantitative mass balance approach, based on selected elemental tracers, to assess the soil ingestion rate in a First Nations' group practicing a wilderness lifestyle previously used by Doyle et al. (2012). More specifically, the study followed 9 subjects practicing traditional activities in Cold Lake, Alberta, Canada over a 13 day study period. The Cold Lake region is home to the largest *in situ* thermal heavy oil (bitumen) extraction operation in the world. We hypothesize that soil ingestion rates in people that practice traditional lifestyles is greater than current soil ingestion rates used by several regulatory agencies such as Health Canada and the US EPA.

2.3 Methods

2.3.1 Study area

The study was conducted near Cold Lake, Alberta, Canada, at a site approximately 300 km northeast of Edmonton, Alberta near the Alberta-Saskatchewan border. The Cold Lake region is one of the three major oil sands regions of Alberta along with the Athabasca and Peace River deposit. The region also contains the City of Cold Lake, with a population of approximately 14,000, the Canadian Forces Base Cold Lake, and four native reserves of the Cold Lake First Nations. The aboriginal people of the Cold Lake First Nations belong to the Dene Suline tribe, whose traditional lands ranged from south of Bonnyville to the northernmost point at Peter Pond Lake, Saskatchewan. The area has a humid continental climate, with a lower than average rainfall compared to other Canadian cities, and contains many unpaved roads that can contribute to airborne dust particles. Local bedrock geology is predominantly marine shale of the Lea Park Formation from the Upper Cretaceous (Prior et al. 2013) overlain by various fluvial sediments and glacio-fluvial tills (Fenton and Andriashek 1983).

2.3.2 Soil ingestion study design

The handling and analysis of fecal samples followed the methods previously employed by Doyle et al. (2012) for a soil ingestion study of the Xeni Gwet'in First Nation in the Nemiah Valley of British Columbia. A total of 10 adult volunteers were followed over a 13 day period from August 17 to August 30, 2012. Seven of the study participants were members of Alberta aboriginal communities represented by the same tribal council as Cold Lake First Nations, Tribal Chiefs Ventures Incorporated. The other three participants were field technicians from the University of Ottawa. One participant voluntarily withdrew before the study was completed, resulting in 9 subjects. Only one field technician was present for the entire duration of the study. The study was conducted in August 2011 since the dry conditions common in the late summer likely contribute to enhanced soil ingestion.

Prior to subject recruitment and study initiation, the study was reviewed and approved by the research ethics boards of the University of Ottawa and Health Canada. All participants were interviewed before the commencement of the study and informed about the study objectives, the study location, the nature of their participation, and the handling of their personal information. Each participant signed consent forms, which were pre-approved by both research ethics review boards.

This study used mass balance soil ingestion methods developed and employed by Doyle et al. (2012), which are based on methods established by Calabrese et al. (1989, 1997). Soil ingestion mass balance methods use elemental tracers that are relatively inert (i.e., little or no gastro-intestinal tract absorption), ubiquitous in soils, and have a low food/soil ratios (i.e., low concentrations in food relative to soil). Tracers meeting these requirements include Al, Ba, Ce, La, Mn, Si, Th, Ti, U, V, Y, Zr.

2.3.3 Daily activities

An outdoor base camp was established at English Bay (Figure 2.1). All participants remained at the base camp for the duration of the study, and engaged in a variety of outdoor traditional activities during the day (e.g., fishing, hunting, food gathering). Each participant slept in their own tent and there was a separate tent and trailer setup for food storage and preparation. All food was provided and prepared for study participants, and the exact amount of food consumed by each participant was pre-weighed and carefully recorded. The activities varied from day to day and all activities took place on the Cold Lake First Nations reserves and surrounding region. Activities included hunting and setting traps and snares on the reserve, fishing and setting fishing nets in English Bay (Figure 2.1), collection of medicinal plants on the reserve and surrounding traditional lands, and collection of foods and spices such as blueberries, bear berries, and mint. In accordance with the law, hunting was carried out on the Cold Lake First Nations reserve by participants possessing Indian status cards. Two individuals regularly set traps and snares for rabbits, which were skinned and prepared for meals. There was no attempt to influence the subjects' activities and some of the elders involved in the study chose to remain at the camp for some days during the study. The duration and type of each activity was recorded. Other than being present at camp for breakfast and dinner, the participants were free to do as they pleased, with the exception that they were prohibited from consuming foods not provided as part of the study.

2.3.4 Fecal collection

Fecal samples were collected from each participant from Day 0 to Day 13. To make fecal sample collection as simple as possible, portable commodes were setup at two enclosed locations at the base camp. Each participant was given pre-weighed and labeled sample bags (Fisher Scientific autoclavable polypropylene biohazard sample bags, catalog number 01-826-5). Inside each commode enclosure were plastic zip ties and an ice packed cooler. After fecal collection, the participant sealed their sample bag and placed it inside the cooler. Each evening, samples were transferred to an off-site -20°C freezer. For shipment, fecal samples were stored in ice packed coolers and shipped via air transit. Samples were kept in a dedicated sample freezer prior to processing and analysis.

Fecal samples were processed inside a fume hood except when being transferred from the freezer or to the muffle furnace. Samples were weighed, then dried at 90°C for 72 hours and reweighed to obtain dry weights. Samples were then transferred to crucibles and ashed in a muffle furnace at

500°C for 6 hours and weighed to determine ashed weight. Samples from the same individual for each day were combined and placed into glass scintillation vials in preparation for analysis.

Ashed samples were analyzed by a commercial lab accredited by the Canadian Association for Laboratory Accreditation to ISO/IEC 17025:2005. Samples were analyzed for elemental tracers Al, Ba, Ce, La, Mn, Si, Th, Ti, U, V, Y, and Zr using EPA method 3052, which involves microwave-assisted nitric acid and hydrofluoric acid digestion. Concentrations of elemental tracers with the exception of Si were analyzed using inductively coupled plasma mass spectrometry. Si (%) was measured by sodium peroxide fusion and inductively coupled plasma optical emission spectrometry.

2.3.5 Food collection

All food was provided from lunch on Day 0 to dinner on Day 13, which included breakfast, lunch, dinner and snacks. Average weights of food portions (e.g. meat slices, servings of vegetables) were determined before each meal and after each meal, and each participant was debriefed to determine how many servings of each food item they had consumed. Before the study began on Day 0, each participant was given an initial briefing and asked to report on their activities and food consumption on the days leading up to the start of the study.

Methods for the analyses of food samples were similar to those described for fecal samples, except food samples were not dried prior to being ashed.

All participants ate breakfasts and dinners together each day. Lunches were prepared after breakfast and packed for each participant before each participant departed for the day. Snacks were also provided. Each participant was de-briefed a minimum of twice each day to record how many servings of each food item they had consumed at each meal from Day 0 to Day 13. Day 13 recordings were not used in soil ingestion calculations since fecal samples were not collected past Day 12. Water was purchased in the City of Cold Lake.

2.3.6 Soil collection

Soils were collected at 13 locations (Figure 2.1) by removing vegetation and collecting only the top superficial soil layer into labeled WhirlPak™ bags, which were stored in a freezer at -20°C until shipping to Ottawa in an ice packed cooler. The collected soils correspond to areas where participants of the study spent a significant time during the day, and from the base camp where participants slept and ate. Soils were air dried for 1 week, broken up lightly with a mortar and pestle, and sieved into multiple particle size fractions. Soil fractions < 63 µm were stored in glass scintillation vials until analysis.

2.3.7 Soil ingestion calculation

Using Eq. 1, daily soil ingestion rate values were calculated for each tracer for each individual. Daily soil ingestion estimates were calculated using the following equation (Eq. 1)

$$Sa = \frac{Fc \times Fa}{Sc} - \frac{Ic \times Ia}{Sc}$$

S_a = soil ingested (g)

F_c = concentration of tracer element in feces (µg/g)

F_a = mass of feces (g)

I_c = food concentration for tracer element (µg/g)

I_a = mass of food ingested (g)

S_c = concentration of tracer in soil (µg/g)

The daily soil ingestion rate was calculated for each participant using the food intake on Day 0 to Day 13 with the fecal output (F_c * F_a) offset by one day (e.g., food consumed on day 3 and fecal output on day 4). Mass of food ingested (I_a) was determined by using the food logs and the pre-weighed portion sizes, and I_c was derived from three sources: analysis of the actual food items; tracer concentration values from the study by Doyle et al. (2012); and tracer concentration values of Al, Ba, Mn, Th, and U from the Health Canada Total Diet Study (Health Canada, 2011). Soil concentration (S_c) was the mean tracer level measured in the soils collected during the study. Transit time for element tracers was assumed to be 24 hours to be consistent with previous studies.

2.3.8 Statistical analysis

All statistical analyses were conducted using Microsoft Excel™ or IBM SPSS Statistics (IBM Software, Armonk, New York). In cases where distributions were normal and variances across treatment groups equal, an ordinary ANOVA with the Dunnett T3 post-hoc test was employed. A Levene's test was used to test for unequal variance, and the Shapiro-Wilk test used to test for normality. When only two means were compared, the student t-test was used instead of ANOVA. Non-normal distributions were compared using the Kruskall-Wallis test or a Mann-Whitney U test. An alpha level of 0.05 was chosen to determine significance.

2.4 Results

2.4.1 Soil

Twelve elemental tracers were analyzed in samples collected from thirteen sampling sites. The variation in elemental soil tracers used to calculate soil ingestion rate was not large, with CV values ranging from 7% to 44% (Table 2.1). Al had the smallest variation with a CV of 7%, and Si had the second smallest variation with a CV of 9%. The largest soil CV value of 44% was obtained for Th.

2.4.2 Fecal samples

Fecal sample collection was intermittent throughout the study. Participants were encouraged to provide samples each day, but some participants did not provide a daily sample (Table 2.2). The intra-subject variability in fecal dry weight was low for all participants, with the exception of I and J (Table 2.3). Subjects I and J were field technicians that did not participate for the entire 13-day period of the study. Subject I gave only 2 daily samples, had the smallest CV of 20% and subject J has a much larger CV of 88%. After removing subject C from the analysis, the average daily fecal dry weight and the daily fecal weight standard deviation of participants trended downwards over the study duration. Although regression analysis shows this trend was not statistically significant ($F_{(1, 11)} = 2.07$, $p = 0.18$), a larger study may or may not detect a significant difference with higher statistical power.

2.4.3 Food samples

The suitability of each tracer for ingestion rate determination was evaluated for each of the elements examined. Those elements that showed a low food to soil (F/S) ratio and low CV (%) in food were regarded as reliable for ingestion rate determination.

The daily tracer intake through food (Table 2.4) varied depending on the elemental tracer, but the CVs were reasonably consistent ranging from 42% to 64%. Mn is an exception, with an 86% CV daily tracer intake; however, Mn is generally regarded as an unreliable elemental soil tracer because of its F/S ratio of 8.43 (Table 2.4). Ba also had a relatively high CV. The small F/S ratio for Ba makes it seem like a suitable tracer, however, the high CV of 140%, combined with a very high SD for the Ba-determined soil ingestion rate (Table 2.5) suggest that it is not a reliable tracer.

The F/S ratios ranged from 0.02 for Ba to 8.43 for Mn. A small F/S ratio is generally regarded as an indication that the tracer is more reliable for soil ingestion determination due to its near absence in food (Calabrese and Stanek 1993; Stanek and Calabrese 1991).

The aforementioned tracers with the lowest F/S ratio (i.e., Al, Ce, La, and Si), with the exception of Ba, were considered reliable tracers for this study, and used for soil ingestion rate calculations. The F/S ratios for these elements were 0.08, 0.07, 0.07, and 0.10, respectively. Although Ce and La have lower F/S ratios, Al and Si were selected as the most reliable tracers for this study because of their much higher soil concentration and this is consistent with previous works (Calabrese et al. 1989, 1997; Davis et al. 1990; Doyle et al. 2012; Stanek and Calabrese 1991; Stanek et al. 2001). Furthermore, a recent meta-analysis by Stanek et al. (2012) of published soil ingestion studies noted that Al and Si are the most consistently reliable tracers and recommended their use in following soil ingestion studies.

2.4.4 Soil ingestion estimates

The calculated soil ingestion rates range (Table 2.5) from a mean of -378 mg d^{-1} for Th, to 3215 mg d^{-1} for Ti. Similarly, the medians range from a low of -390 mg d^{-1} for Th to a high of 1034 mg d^{-1} for Mn. For reasons discussed in a later section, the majority of these values are likely not indicative of actual soil ingestion rates for the study participants. In comparison, the calculated ingestion rates for the four tracers considered most reliable (i.e., Al, Ce, La, and Si) revealed mean soil ingestion rates over the study duration of 33 mg d^{-1}, 9 mg d^{-1}, 10 mg d^{-1}, and 65 mg d^{-1}, respectively, and median rates of 7 mg d^{-1}, -4 mg d^{-1}, -2 mg d^{-1}, and 37 mg d^{-1}, respectively. As noted, these tracers are generally regarded as the most reliable because of their low F/S ratio; moreover, previous studies have generally regarded Al and Si as tracers that provide the most consistent results. Soil concentrations of Al and Si in this study showed low CV (Table 2.1). In addition, the soil concentrations of Al and Si have a lower CV (%) than other tracers such as Ce and La.

The mean soil ingestion rates show substantial inter-subject variability (Table 2.6), however, there were no significant differences between the mean soil ingestion rates for Al (Welch ANOVA p = 0.63) or Si (Welch ANOVA p = 0.29). Subject B consistently yielded a negative mean soil ingestion rate for all 4 of the reliable tracers, and almost all tracers examined. All other subjects with the exception of subject I, who participated for only 3 days and provided only 2 fecal samples, had positive soil ingestion rates for the 4 most reliable tracers. Soil ingestion rates were not lognormal, so bootstrapping statistics based on 5000 replicates were used to determine upper and lower limit 95% confidence intervals. The bootstrapped, bias-corrected and accelerated (BCa) upper and lower 95% confidence intervals (Table 2.5) are similar to untransformed upper and lower 95% confidence intervals (not shown), varying by approximately 1 to 5 mg d^{-1} for Al, Ce, La, and Si.

Data were not normally distributed, and the differences between sample variances were large. A Levene's homogeneity of variance test found variances to be significantly different between the four tracers Al, Ce, La, Si (Levene, $F_{(3,348)} = 4.748$ $p = 0.003$). The soil ingestion rate sample means were different between Ce and Si (Welch ANOVA, Dunnett T3, p = 0.018), and La and Si (Welch ANOVA, Dunnett T3, p = 0.021), the sample means between Al and Si were not significantly different (Welch ANOVA, Dunnett T3, p = 0.67) (Figure 2.2). Additionally, there were no differences between sample means of Al and Ce (Welch ANOVA, Dunnett T3, p = 0.36), and Al and La (Welch ANOVA, Dunnett T3, p = 0.40). The mean soil ingestion rate for Si was significantly higher than Ce, and significantly higher than La, but there were no significant differences between the other three tracers. There was no significant difference between Al, Ce, La, and Si (Kruskall-Wallis p = 0.074). The average ingestion rate for the four most reliable tracers, Al, Ce, La, Si, were normally distributed (Shapiro-Wilk W = 0.99, p = 0.65) following a natural log transformation. Additionally, Si was also found to be normally distributed (Shapiro-Wilk W = 0.99, p = 0.71) after the same transformation.

The soil ingestion study was arbitrarily separated into two weeks to determine if soil ingestion rate was affected by the participant's familiarity with the study's requirements. The week 1 mean was 36 ± 85 mg d^{-1} for the 4 most reliable tracers and the week 2 mean was 29 ± 92 mg d^{-1}. The data was not normally distributed, nevertheless the analyses conducted failed to reveal any significant differences between the first and second half of the study (Mann-Whitney U = 845, p = 0.44).

Participants were separated into their daily activities to determine if activity type affected soil ingestion rate. No trends were found between expected higher soil contact activities such as attending a rodeo, gathering, or hunting and remaining at camp for the day.

2.5 Discussion

This study was designed to assess the soil ingestion rate for a group of First Nations people inhabiting a wilderness area and engaging in traditional activities associated with a subsistence lifestyle. In some respects it is similar to the recent study of the Xeni Gwet'in First Nation published by Doyle et al (2012). This work extends and improves on the work conducted by Doyle et al, and some earlier soil ingestion studies, by extending the period of subject observation to 13 uninterrupted days, including a greater number of participants, by specifically controlling food consumption over the duration of the study, and lastly, by including foods specifically chosen for their low content of the elemental tracers of interest, particularly Al and Si.

Although the differences between the average soil ingestion rates from this study (i.e., 36 ± 112 mg d^{-1} for Al and 68 ± 151 mg d^{-1} for Si) and Health Canada's recommended soil ingestion rate values for adults of 20 mg d^{-1} or the analogous US EPA value of 50 mg d^{-1} are much smaller than the predicted 400 mg d^{-1} rate by Harper et al. (2007), the 95th percentile ingestion rate values for Al and Si are 268 and 361 mg d^{-1}, respectively. The 95th percentile value for Si is greater than the 95th percentile value of 330 mg d^{-1} from studies used to inform the Health Canada and US EPA soil ingestion rates (Health Canada 2012; EPA 2011; Wilson Scientific Consulting Inc. and Meridian Environmental Inc. 2006). Thus, the results from this study, as well as the previous work by Doyle et al. (2012), suggest that soil ingestion rates currently used in HHRA may be too low for some populations; moreover, some individuals in a population may not be adequately protected using HHRA guidelines advocated in Canada, the USA, and other developed countries around the world (EPA 2011). Although the 95th percentile soil ingestion rate calculated in the present study is greater than the currently recommended ingestion rate value, it is still below the estimate of Harper et al.

(2007), which suggested that First nations people ingest 400 mg d^{-1}. The soil ingestion rates are similar to the recommended ingestion rate for construction workers (i.e., 330 mg d^{-1}) who are regularly engaged in high soil contact activities (EPA 2011).

Comparisons between soil ingestion studies are shown in Table 2.7. The present study doubled the number of soil ingestion estimates from 43 to 87. The mean ingestion rate for the 4 most reliable tracers obtained in this study (i.e., 32 mg d^{-1}) is significantly lower than the analogous value from Doyle et al. (2012) using an independent sample 2-tailed t-test ($t_{(128, 0.05)} = -2.663$, $p = 0.009$). Both studies appear to have similar daily soil ingestion rates for Al, with mean values of 37 mg d^{-1} for the Doyle et al. (2012) study and 36 mg d^{-1} for the current study; however, non-parametric statistical tests still found significant differences between these values, likely attributable to the higher soil ingestion rate variation in this current study (Mann-Whitney U = 1471, p = 0.048). The differences between the Al soil ingestion rates are more apparent with the median values. The Al soil ingestion rate median was 7 mg d^{-1} in the present study, and Doyle et al. (2012) reported 31 mg d^{-1}, similar to the mean Al value for that study. The present study found a 68 mg d^{-1} mean Si soil ingestion rate, whereas Doyle et al. (2012) reported a mean of 49 mg d^{-1} for Si. The difference between the Si soil ingestion rates was not significant ($t_{(94, 0.05)} = -0.71$, $p = 0.94$). The median ingestion rate values for Si in both studies were similar; 40 and 37 mg d^{-1} for the Doyle et al. (2012) study and the current study, respectively. Furthermore, the 90th percentile soil ingestion rates were similar for the two studies, with means of 152, 161, and 236 mg d^{-1} in this study for the four most reliable tracers (i.e., Al, Ce, La, Si), Al, and Si, respectively, compared with 193, 110, and 145 mg d^{-1} for Doyle et al. (2012). Although the 90th percentile ingestion rates are similar for the two studies, both studies showed that average values are quite different than the 90th percentile, which are between 100 mg d^{-1} to greater than 200 mg d^{-1}.

Previous mass balance soil ingestion studies have reported a wide range of soil ingestion rates. Calabrese et al. (1989) measured daily soil ingestion rates in toddlers of 1 to 4 years old, reporting mean values of 153 ± 852 mg d^{-1} and 154 ± 693 mg d^{-1} for Al and Si, respectively. Davis et al. (1990) conducted a study of children aged 2 to 7 years old, and reported soil ingestion rates 39 ± 145 mg d^{-1} and 82 ± 122 mg d^{-1}, for Al and Si, respectively. The best tracer methodology (BTM) was first advocated by Calabrese et al. (1997), where BTM is the median of the four tracers with the lowest F/S ratio (Stanek and Calabrese 1995). This study that first employed the BTM followed 64 toddler participants aged 1 to 4 years old and reported a soil ingestion rate of 7 ± 76 mg d^{-1} for the BTM, 3 ±

96 mg d^{-1} for Al, and -16.5 ± 57 mg d^{-1} for Si (Calabrese et al. 1997). These studies of children, as well as the adult studies discussed below, were all conducted on suburban or urban populations.

Published adult soil ingestion studies report mean daily soil ingestion rates in the same range as the studies of children. The first mass balance adult study by Calabrese et al. (1990), a pilot study to validate the methods employed the children study by Calabrese et al. (1989), followed six adult participants and reported mean soil ingestion rates of 77 ± 65 and 5 ± 55 mg d^{-1} for Al and Si, respectively. Similar to the previous study, the soil ingestion study by Stanek et al. (1997) was also a validation study that assessed soil ingestion in 10 adults to evaluate methods for the soil ingestion study of children by Calabrese et al. (1997). Adults were given capsules of known amounts of soils that were subtracted to determine soil ingestion rates. The reported values, 6 ± 165 for BTM, 12 ± 31 for Al, and -20 ± 37 mg d^{-1} for Si, are generally lower than values published earlier by this group. The study by Davis and Mirick (2006) was the first study that looked at soil ingestion in both the parents and children of the same family simultaneously. They followed 19 families that included a mother, a father, and one child for 11 days, and the means for the children were 37 ± 35 mg d^{-1} for Al and 38 ± 31 mg d^{-1} for Si. The average ingestion rates for the fathers were 68 ± 130 for Al and 26 ± 49 mg d^{-1} for Si. The average ingestion rates for the mothers were 92 ± 218 mg d^{-1} for Al and 23 ± 37 mg d^{-1} for Si.

The aforementioned studies have collectively contributed to a continual improvement in the methods employed to assess soil ingestion rate, and the more recent studies show reduced variability in soil ingestion estimates as indicated by a reduction in standard deviations (SD) in comparison with earlier studies. For example, the SD for the Al ingestion rate was 852 in Calabrese et al. (1989), compared with 96 in Calabrese et al. (1997). Apart from lower SD, the more recent studies also have lower mean soil ingestion rates than the early mass balance studies.

There were 12 different elemental tracers measured in this study to calculated soil ingestion rates. The extreme values of soil ingestion rates calculated using tracers such as Ti with a mean of 3215 mg d^{-1} or Th with a mean of -378 mg d^{-1} are unlikely to be accurate soil ingestion experienced by participants in the study. Th has a much higher F/S ratio of 0.53 and very low soil concentrations. This indicates that most of the Th ingested is from tracers in food and not soil. Transit time error is likely to explain the very low soil ingestion rate calculated, transit time error will increase with higher F/S ratios (Doyle et al. 2010). The very high soil ingestion rate for Ti is likely the result of other sources of Ti besides food or soil, evidenced by the large SD and SE (Table 2.5). Ti is a common additive to consumer products and it is likely that many participants absorbed or ingested Ti through sunscreen, lotions, foods, and toothpastes (Weir et al. 2012). Other tracers with lower F/S ratios such as Y may be useful in soil ingestion studies, but the higher F/S ratio compared to the four chosen tracers (Al, Ce, La, Si) indicate that in this region it would not be as accurate as the four chosen tracers with lower F/S ratios.

The soil ingestion rate variance reported in our study is similar to several of the previous soil ingestion studies summarized above, but greater than that noted in Doyle et al. (2012) for both Al (Levene's test F = 7.93, p = 0.006) and Si (Levene's test F = 10.8, p = 0.001). Unlike the Doyle et al. (2012) study, the participants in the current study did not provide a fecal sample for each day of the study (Table 2.2); nevertheless, all participants reported not defecating on those missed days.

Although it is unlikely that the amount of soil contact associated with the activities in the current study was constant across the activities, most of the activities can be considered medium soil contact activities and would be common in a wilderness setting. The type of traditional activity did not appear have an effect on soil ingestion rate determined in this study, as participants who attended rodeos, a high soil contact activity had very similar soil ingestion rates than participants who remained at camp for the day. For example, hunting and gathering fruits and medicines were the two most popular activities in the study (i.e., 8 subjects for 9 days). In contrast, on two of the days, four participants attended a rodeo as observers, an activity that could reasonably be regarded as "high contact" due to the dusty and arid nature of the environment but differences in soil ingestion rates were not observed. In either case, it is reasonable to assert that the participants' activities in the current study differ from activities that are typical of a suburban or urban community. Thus, it is also reasonable to assert that inadvertent soil ingestion rate values from previous urban/suburban studies would not be expected to be representative of soil ingestion in a First Nations community where wilderness, subsistence lifestyle is common (Calabrese et al. 1989, 1990, 1997; Davis and Mirick 2006; Davis et al. 1990; Stanek et al. 1997). Indeed, the soil ingestion mean values determined in this study for aboriginal people leading traditional lifestyles in the Cold Lake region and the earlier study by Doyle et al. (2012) are greater than reported mean values from previous soil ingestion studies used by Wilson Scientific Consulting Inc. and Meridian Environmental Inc. (2006) to recommend Health Canada's risk assessment guidelines. It should be noted that this study did not examine ingestion of soil via traditionally prepared foods. Some foods eaten during the study were caught/harvested and prepared by the study participants, and soil adherence to traditional foods was not measured.

The aforementioned studies by Doyle et al. (2012), Davis and Mirick (2006), and Stanek and Calabrese (1991) summarize the assumptions employed for mass balance tracer determination of soil ingestion rates. It is generally agreed that effective tracers are (1) low in food or water, and (2) high in soil, and (3) not absorbed in the gastrointestinal tract (Stanek and Calabrese 1991). Moreover, accurate determination of soil ingestion rate requires a one to one correspondence between tracer ingestion and fecal output. Unfortunately, inter- and intra-subject variability in the daily ratio between tracer intake and tracer excretion contribute to inter-day variations in calculated ingestion rates. Additionally, all studies assume that the time it takes for a tracer to travel through the body, or tracer transit time, is 24 hours; however, this value also varies across time and subject, and can depend on a number of factors such as age, diet and gender (Doyle et al. 2012; Stanek and Calabrese 1991; Madsen and Jensen 1989). Madsen and Jensen (1989) found a mean transit time of 28.5 hours, which is the summation of the mean transit times for each section of the gastro-intestinal system. A 24 hour transit time allows fecal samples to be collected on a daily basis for determination of daily soil ingestion rates. Previous studies generally collected samples from participants followed for only a few days and errors introduced by what is referred to as “transit time misalignment” can be large. However, as the duration of a study increases, the food tracer intake should balance with the fecal tracer output (Stanek and Calabrese 1991). Thus, the current study benefited from an extended duration as well as dietary control. The meals provided included foods specifically highlighted by Doyle et al. (2012) as low in Al and Si (i.e., fresh meats, fruits, and vegetables). Processed foods were avoided, as recommended by Doyle et al. (2012), since these have been found to have elevated levels of elemental tracers. Although source error (i.e., contributions of tracer from unknown sources) could not be entirely removed from our study, it was minimized by controlling food intake and providing an encampment for the participants. It should be noted that the tracers examined here are known to have low gastrointestinal tract absorption rates (Calabrese et al. 1989, 1997; Doyle et al. 2012; Stanek and Calabrese 1991). Finally, the uniformly high Al and Si concentrations in the collected soils (Table 2.1), with low CVs of 7% and 9%, meet the criteria established by Stanek and Calabrese (1991) for a good quality tracer study.

Further work should focus on determining soil adherence to traditional foods, and the effect on soil ingestion rates in First Nations people (Harper et al. 2007). The accuracy of soil ingestion rate values for First Nations people practicing traditional, subsistence lifestyles could be improved by examining the relationships between ingestion rate and the frequency of specific activities; moreover, the season of the activity. Although the logistics of a study that effectively examines the relationship between ingestion and activity may prove difficult, the study could be highly focused and examine only a small number of subjects over a shorter period of time. Such a detailed examination of ingestion rate for First Nations' people may prove very useful for risk assessments of remote contaminated regions or areas being considered for resource exploitation. One would expect that soil ingestion rates will be lower in winter, but this is still speculative since all soil ingestion studies conducted to date have been done in summer and early fall (Stanek et al. 2011).

2.6 Conclusions

This study is the longest continuous soil ingestion study conducted to date, spanning 13 consecutive days. The mean soil ingestion estimates obtained over the study duration were 33 mg d^{-1}, 65 mg d^{-1}, and 32 mg d^{-1} for Al, Si, and the mean of the most reliable tracers (Al, Ce, La, Si) are comparable to the previous study of a First Nations population by Doyle et al. (2012), of 37 mg d^{-1}, 49 mg d^{-1}, and 74 mg d^{-1} respectively and similar to the adult soil ingestion rate values recommended by Health Canada and the US EPA (i.e., 20 mg d^{-1} and 50 mg d^{-1},respectively). The hypothesis that soil ingestion is greater for First Nations' people practicing a traditional subsistence lifestyle, in comparison to an urban/suburban population is supported in the sense that the ingestion rates determined here, particularly the 95th percentile ingestion rates, which ranged from of 361 mg d^{-1} for Si, were greater than mean and 95th percentile soil ingestion rates determined for urban or suburban adult populations (Wilson Scientific Consulting Inc. and Meridian Environmental Inc. 2006). Nevertheless, as noted earlier, the mean as well as the 95th percentile estimates are lower than Harper et al.'s (2007) prediction of soil ingestion rates for First Nations people (i.e. 400 mg d^{-1})

The results of this study support the continued use of mass balance soil ingestion methods to examine a wilderness or rural population. Like the Doyle et al. (2012) study, Al and Si were found to be reliable soil ingestion tracers, along with Ce and La, which also have low F/S ratios.

The soil ingestion rates presented here only represent rates in First Nations' people involved in moderate soil contact activities over the time period examined, and thus, may not be representative of other regions, other seasons, or other activities. Although this study did not detect a statistically significant effect of activities on soil ingestion rate, follow-up studies that include higher soil contact activities (i.e., rodeo participation) and other seasons may be able to detect relationships between activity/season and ingestion rate, and thereby contribute to an improved understanding of non-dietary, inadvertent soil ingestion in First Nations' people.

2.7 References

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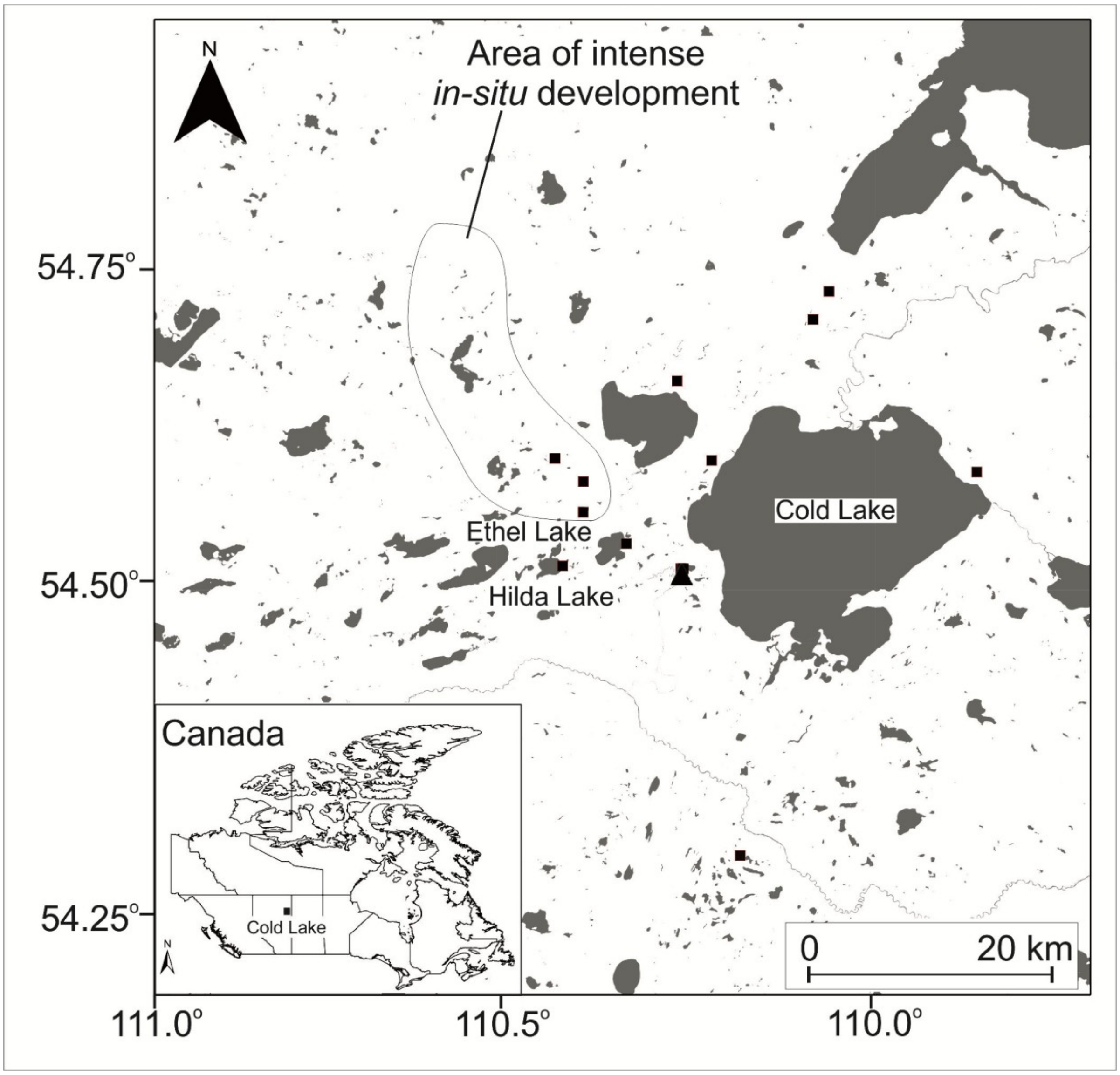

Figure 2.1: Map of the Cold Lake region. Sampled soil sites are marked with a black square. The base camp is marked with a black triangle.

Table 2.1: Soil concentrations of 12 elemental tracers examined at 12 sampling sites. Table shows the mean, standard deviation, coefficient of variation and sample size for each elemental tracer used to estimate soil ingestion rate. Soil concentrations in µg g^{-1}.

Soil	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Y	Zr
Mean	42538	598	55	26	497	329296	9	1879	51	2	13	130
SD	9623	98	16	7	155	28164	4	422	16	1	4	40
CV (%)	7	16	30	28	31	9	44	22	32	27	31	31
n	13	13	13	13	13	9	13	13	13	13	13	13

Table 2.2: Missed fecal sample collection days. Days marked with an X indicate that the subject provided a fecal sample. Note that subject I left the study on Day 3 when subject J arrived. Subject H, I, and J were field technicians.

	Subject								
	A	B	C	D	E	F	H	I	J
Day									
0	X			X	X	X		X	
1	X	X		X	X	X	X	X	
2	X	X	X	X	X	X	X		
3	X	X	X	X	X	X	X		X
4	X			X	X	X	X		X
5	X			X	X	X	X		X
6	X			X	X	X	X		X
7	X	X	X	X	X	X	X		X
8	X	X	X	X	X	X	X		X
9	X	X		X	X	X	X		X
10	X	X	X	X	X	X	X		
11	X	X	X	X	X	X			
12	X	X	X	X	X	X	X		
13	X	X	X	X	X	X	X		

Table 2.3: Fecal dry weight values for each study participant. Table shows the mean, standard deviation, CV (%), median, lower and upper 95% confidence intervals and sample size (n) for each subject over the study duration.

		Dry Weight (g)					
Subject	n	Mean	Standard Deviation	CV (%)	Median	Lower 95%	Upper 95%
A	13	27.3	13.3	49	27.3	19.9	40.4
B	10	32.1	13.9	43	36.5	18.9	44.2
C	5	99.5	60.0	60	78.4	15.4	56.9
D	13	27.2	16.9	62	21.8	19.9	40.4
E	13	50.8	16.0	31	49.3	19.9	40.4
F	13	49.3	15.9	32	48.2	19.9	40.4
H	11	46.4	21.9	47	40.9	19.3	42.7
I	2	27.2	5.5	20	27.2	8.5	82.3
J	7	21.2	18.6	88	11.8	17.2	50.2

Table 2.4: Fecal dry weight values for each study participant. Table shows the mean, standard deviation, CV (%), median, lower and upper 95% confidence intervals and sample size (n) for each subject over the study duration.

	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Y	Zr
Daily tracer intake (food)												
Mean ($\mu g\ d^{-1}$)	3,546	842	3.98	1.92	4187	32898	4.85	272	18.6	1.06	1.82	19.6
SD	1878	750	2.20	0.98	3607	15216	2.84	131	7.91	0.48	1.08	12.6
CV (%)	53	89	55	51	86	46	59	48	42	46	59	64
n	108	108	108	108	108	108	108	108	108	108	108	108
Fecal												
Mean ($\mu g\ g^{-1}$)	1205	277	1.17	0.56	1304	10390	0.44	1292	2.47	0.37	0.43	4.78
SD	749	130	0.55	0.26	572	6525	0.55	1695	1.20	0.15	0.20	4.35
CV (%)	62	47	47	46	44	63	127	131	49	42	48	91
n	87	87	87	87	87	74	87	87	87	87	87	87
Food/soil ratio												
Mean	0.08	0.02	0.07	0.07	8.43	0.10	0.53	0.14	0.37	0.56	0.14	0.15
SD	0.04	0.03	0.04	0.04	7.27	0.05	0.31	0.07	0.16	0.26	0.08	0.10
CV (%)	53	140	55	51	86	46	59	48	42	46	59	64
n	108	108	108	108	108	108	108	108	108	108	108	108

Table 2.5: Summary of soil ingestion rates calculated for each of the 12 elemental tracers examined. Soil ingestion rate expressed as mg d^{-1}.

	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Y	Zr
Mean	36	318	12	12	1998	68	-378	3215	-183	196	-7	19
Standard Deviation	117	1662	72	78	10107	152	461	5622	238	626	145	407
Standard Error	12	176	8	8	1071	16	49	597	25	66	15	43
Median	7	467	-4	-2	1034	37	-390	759	-185	143	-17	-30
90th percentile	165	1744	111	97	11555	231	109	9325	111	1032	159	211
95th percentile	268	2405	132	156	18226	361	217	16459	169	1226	230	301
Upper 95% CI[a]	65	650	29	32	4075	104	-283	4662	-129	328	27	196
Lower 95% CI[a]	15	-26	-1	-1	-164	40	-479	2242	-230	69	-34	-34
n	87	87	87	87	87	87	87	87	87	87	87	87

a: Upper and lower 95% confidence intervals are bootstrapped confidence intervals with 5000 bootstrapped replicates.

Table 2.6: Soil ingestion rate values for each participant. The table shows the mean, standard deviation (SD), median, and sample size for each participant over the study duration. Soil ingestion rate expressed as mg d^{-1}.

Subject	Al	Ba	Ce	La	Mn	Si	Th	Ti	V	U	Y	Zr
A	18	384	19	16	2346	119	-244	5314	-197	117	16	64
B	-47	-1005	-41	-42	-2820	-19	-790	163	-400	-295	-157	-38
C	152	485	82	92	6665	267	-725	13574	-352	260	78	616
D	32	37	-2	-1	-1697	3	-195	839	-185	-103	-60	-57
E	55	8	-8	0	-875	61	-544	1861	-179	290	27	-109
F	32	394	34	25	10137	58	-406	2343	-252	166	25	-13
H	60	631	13	11	4714	72	-111	5055	-30	582	-15	-30
I	-11	620	-9	-7	1408	87	-349	443	45	54	-34	-38
J	6	1759	10	13	-7612	25	-307	719	-86	601	1	-12
Mean	33	368	11	12	1363	75	-408	3368	-182	186	-13	43
SD	55	725	34	36	5359	84	234	4277	144	292	67	220
Median	32	394	10	11	1408	61	-349	1861	-185	166	1	-30
n	9	9	9	9	9	9	9	9	9	9	9	9

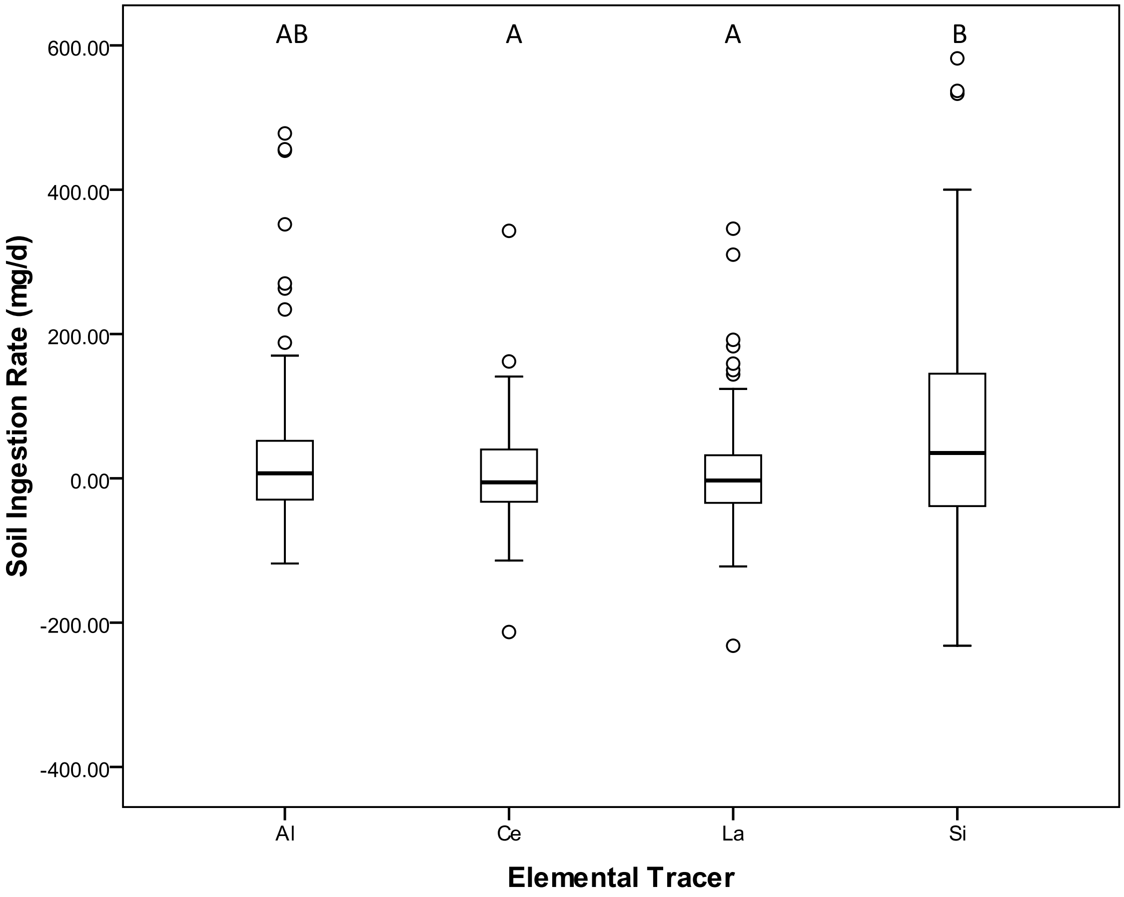

Figure 2.2: Box plot of daily soil ingestion rates determined using the four most reliable tracers. The box limits encompass the interquartile range (i.e., 25th to 75th percentile) with the whiskers indicating the 5th and 95th percentiles, the central line in the box indicates the median. The symbols beyond the whiskers can be regarded as outliers. Welch ANOVA showed significant differences between the tracers ($p = 0.008$). Boxes accompanied by the same letter are not significantly different at $p=0.05$.

Table 2.7: Comparisons of the soil ingestion rates determined in this study with those published by Doyle et al (2012). The Doyle et al. (2012) study examined First Nations inhabitants of in the Nemiah Valley in central British Columbia.

Study and tracer/method	n	**Soil ingestion rate (mg d^{-1})**			
		Mean	**Standard Deviation**	**Median**	**90th Percentile**
Doyle et al. 2012					
• Al, Ce, La, Si	43	74	91	60	193
• Al, Si	43	43	53	38	104
• Al	43	37	52	31	110
• Si	30	49	74	40	145
Current study					
• Al, Ce, La, Si	87	32	88	18	152
• Al, Si	87	52	119	37	220
• Al	87	36	117	7	161
• Si	87	68	152	37	231

Chapter 3 — Cancer risk to First Nations' people from exposure to polycyclic aromatic hydrocarbons near in-situ bitumen extraction in Cold Lake, Alberta

3.1 Abstract

The Alberta oil sands are an important economic resource in Canada but there is growing concern over the environmental and health effects as a result of exposure to contaminants released as a result of the industrialization of this resource. Recent studies have shown a trend of increasing polycyclic aromatic hydrocarbon (PAH) concentrations from the Athabasca oil sands industry but thus far similar studies have not been replicated in Cold Lake, another oil sands region in Alberta. Many PAHs are known mutagens or carcinogens and this study measured soil and atmospheric concentrations of PAHs in the Cold Lake region to assess exposure and lifetime cancer risk to the First Nations' population in the region. Using both deterministic and probabilistic risk assessments, lifetime cancer risk from exposure to PAHs in the Cold Lake region was measured through inhalation and inadvertent soil ingestion. Mean cancer risk to a First Nations' population that practice traditional activities in the Cold Lake region was 0.02 cases per 100,000 people with a 95% risk level of 0.07 cases per 100,000 people. Exposure to PAHs through inhalation had a maximum lifetime cancer risk of less than 0.1 cases per 100,000 people. Lifetime cancer risk below 1 case per 100,000 people is considered negligible and for that reason there was no significant risk calculated for the population in the Cold Lake region as a result of exposure to PAHs.

3.2 Introduction

Canada has the third largest proven oil reserves in the world (CIA 2013). The oil sands in Alberta, Canada represent the vast majority of Canada's oil reserves and are the largest source of crude bitumen in the world. Currently, there is much speculation in the media and within the general public concerning the impact of the Alberta oil sands on the environment and human health. Some areas of the Alberta oil sands have very high background concentrations of contaminants such as polycyclic aromatic hydrocarbons (PAHs) (Akre et al. 2004; Headley et al. 2001, 2002), but there is increasing evidence that there has been significant rises in PAHs and metals as a result of the industrial development of the oil sands (Hall et al. 2012; Kelly et al. 2009, 2010; Kurek et al. 2013; Timoney and Lee 2009, 2011; Wiklund et al. 2012). Additionally, so far unfounded there is concern in the general public and media about increased cancer rates and other health effects downstream of the Athabasca oil sands (CBC News 2013; Financial Post 2013).

There are three major oil sands regions of Alberta: (1) Athabasca (the largest); (2) Cold Lake; and (3) Peace River. The Cold Lake region, the focus of this study, is the largest *in-situ* thermal heavy oil (bitumen) extraction operation in the world producing 54,600 barrels of bitumen each day and is home to the Cold Lake First Nations that encompasses three reserves, as well as the City of Cold Lake (Burrowes et al. 2011). Much of the published research related to PAHs on the Alberta oil sands has so far been focused on the effects of surface mining and bitumen upgrading in the Athabasca region. Consequently, the majority of research on PAHs in the oil sands region has been in regions with open pit mining, and the effects of *in-situ* extraction are relatively unknown. Kelly et al., (2009, 2010) found increased heavy metals and PAH loading in rivers downstream from bitumen upgrading facilities in the Athabasca region, and concentrations of PAHs and heavy metals were found at elevated concentrations up to 50 km away from the upgrading facilities. Kelly et al. (2009) found an estimated annual loading of ~1200 kg of particulate PAH with ~500 kg of dissolved PAH in a 25 km radius from the upgrading facilities. More recently, Kurek et al. (2013) noted that PAH concentrations in lake sediments in the Athabasca region have increased during the same time period as the commercial operations of the Athabasca oil sands. The concentrations in all lakes have increased since development of the region began, with some dated lake sediment showing PAH concentrations increasing tenfold since predevelopment. Other studies in the Athabasca region have suggested that the region has high natural background concentrations that make it hard to differentiate anthropogenic influences from concentrations that would be expected as a result of natural processes such as erosion (Akre et al. 2004; Headley et al. 2001, 2002).

Many PAHs are known mutagens or carcinogens (IARC 1983) that can enter the body through a number of routes of exposure including ingestion, inhalation, and dermal contact (Stowers and Anderson 1985). The larger 4-6 ringed PAHs, which are typically the most carcinogenic, are highly lipophilic and readily adsorb onto particulate matter both in the water and aerosols, indicating that exposure to PAHs can occur via inhalation of atmospheric particulates, ingestion of contaminated particulates (e.g., soil), or dermal contact with contaminated material (e.g., soil, sediment, water) (Boström et al. 2002; Government of Canada 1994; Lima et al. 2005). Inadvertent ingestion of soil is the dominant exposure pathway on contaminated sites for non-volatile and semi-volatile contaminants such as carcinogenic PAHs.

Studies to date on the Athabasca oil sands region suggest there is increased PAH contamination surrounding oil sands industrial development. This study investigated the excess lifetime cancer risk for inhabitants of the Cold Lake region associated with exposure to PAHs through inhalation and the inadvertent ingestion of soil. This study has an unique opportunity to quantitatively evaluate the excess lifetime cancer risk to First Nations people and other residents in the region via a quantitative soil ingestion study (cf. Chapter 2), and simultaneous assessment of PAH contamination in both soil and air. Soil ingestion rates measured in people that follow a traditional lifestyle will be employed, in conjunction with environmental PAH levels, to assess the excess lifetime risk of cancer in the Cold Lake region. Additionally, cancer risk for the inhabitants of the Cold Lake region who live urban lifestyles was also assessed by utilizing the currently recommended methods for Human Health Risk Assessments (HHRA) from Health Canada (Wilson Scientific Consulting Inc. and Meridian Environmental Inc. 2006). We hypothesize that excess risk of cancer for inhabitants of the Cold Lake oil sands extraction region, more specifically, risk associated with potential exposures to carcinogenic PAHs, constitutes an appreciable increase over background.

3.3 Methods

3.3.1 Study area

The study was conducted near Cold Lake, Alberta, Canada at a site approximately 300 km northeast of Edmonton, Alberta and 300 km southeast of the Athabasca oil fields. The Cold Lake region is one of the three major oil sands regions of Alberta along with the Athabasca and Peace River deposit. The region also contains the City of Cold Lake, with a population of approximately 14,000, the Canadian Forces Base Cold Lake, and four native reserves of the Cold Lake First Nations. The aboriginal people of the Cold Lake First Nations belong to the Dene Suline tribe, whose traditional lands ranged from south of Bonnyville to the northernmost point at Peter Pond Lake, Saskatchewan.

In Alberta, *in-situ* extraction of oil sands covers approximately 30 times the area that is available through surface mining with more than twenty companies operating in the Cold Lake region (Burrowes et al. 2011; Moorhouse et al. 2010). *In-situ* extraction methods are economically viable when oil sands deposits are deeper than 100 m, and the deposits in Cold Lake are more than 400 m below the surface (Gosselin et al. 2010; Moorhouse et al. 2010).

3.3.2 Soil sampling and analysis

Soil samples were collected in August 2011 at 18 locations in the Cold Lake region. Sampling involved collecting the top surficial soil horizon with vegetation removed into labeled WhirlPak™ bags with a pre-rinsed spatula. Soil samples were stored in a freezer at -20°C and shipped in an ice packed cooler. The sampling locations were areas where participants of the soil ingestion study (cf. Chapter 2) spent significant portions of their day. Soils were freeze-dried, broken up lightly with mortar and pestle and sieved into 63 μm fraction using an automated sieve shaker (Soil Test Engineering Model Cl-592B, SoilTest, Evanston, IL, USA) for 10 minutes.

To analyze soil samples for PAHs, 2 g of dried soil was mixed with Agilent brand Hydromatrix and each sample was spiked with known concentrations of ^{13}C labeled PAHs (Cambridge Isotope Laboratories). PAHs were extracted from soil using accelerated solvent extraction (ASE 200 Accelerated Solvent Extraction System, Dionex) at 100°C using hexane: dichloromethane followed by 35% acetone: 65% hexane. Liquid-liquid extractions were then used to separate polar extracts from the non-polar extracts which were collected using hexane, 2,2,4-trimethylpentane, and saturated Na_2SO_4. Samples were run through an Agilent 110 Preparative Liquid Chromatograph with Waters Envirogel Columns (USEPA method 3640A and 3630C) to remove pigments and sulphur. Samples were evaporated to 1 mL and the PAH fraction was separated on a (USEPA method 3630C) 60-100 mesh Davisil 635 silica column. The PAH fraction was analyzed with an Agilent 6890 gas chromatograph and Agilent 5973 mass spectrometer by injecting 1 μL with pulsed splitless mode at 280°C on a DB-XLB 30 m x 0.18 μm x 180 μm column. The oven had an initial temperature of 60°C and was held for 2 minutes and then increased at a rate of 6°C per minute up to 300°C and held for 10 minutes. There was a constant flow of helium at a rate of 39 cm s^{-1} for a total runtime of 52 minutes. The mass spectrometer had a transfer line temperature of 280°C, a source temperature of 230°C and quadrupole temperature of 150°C. The 16 US EPA priority PAHs and ^{13}C labeled PAHs were analyzed and quantified with single ion monitoring. The 16 PAH measured and their PEF are listed in Table 3.1. The PAHs were converted to BaP equivalents for assessing carcinogenic potential with PEF (Schneider and Roller 2002).

3.3.3 Air sampling and analysis

A high volume continuous air sampler was setup on the Cold Lake First Nations reserve next to the campsite used during the soil ingestion study (cf. Chapter 2). The air sampler was calibrated with a Dwyer Instruments 2100 magnehelic pressure gauge using a standard high-volume sampler calibration procedure. Flow through the high-volume sampler in m^3 min^{-1} was calibrated against inches of H_2O magnehelic readings. Glass fibre filters (GFF) (Whatman 110 mm 0.7 µm pore size, #1825110) and Supelco large polyurethane foam (PUF) cartridges (#20038) were replaced every 24 hours. There was an average volume of 844.71 m^3 of air sampled each 24 hour period. Prior to use, all GFFs were placed in an oven for 6 hours at 500°C to remove carbon and then pre-weighed and sealed. Pre-rinsed metal tweezers were used to remove GFF and PUF cartridges during sampling. After sampling, GFF and PUF cartridges were kept in a freezer at -20°C and in ice packed coolers during shipping.

Analysis of PAHs was similar as described previously for soil samples with the following differences. GFF filters were weighed again, and placed in ASE cells that were filled with Agilent brand Hydromatrix. PUF cartridges were cut in half, with both top and bottom half analyzed separated to determine breakthrough and then placed in ASE cells that were filled with Agilent brand Hydromatrix.

3.3.4 Risk assessment

The US EPA and Health Canada risk assessment methodology was used for assessing excess lifetime cancer risk from exposure to PAHs (Health Canada 2012; U.S. EPA 2001a). Deterministic risk assessments to assess lifetime cancer risk were conducted using Eq. 1, where Cs is the concentration in the soil (mg kg^{-1}) or air (mg m^{-3}), IR is the soil ingestion rate (mg d^{-1}) or inhalation rate (m^3 d^{-1}), CF is the conversion factor (10^{-6}), EF is the exposure frequency, from 153 d yr^{-1} to 365 d yr^{-1}, ED is the exposure duration, which is 70 years to assess lifetime cancer risk, and BW is body weight . Following the recommendations of the US EPA, 80 kg was used for this study (U.S. EPA 2011). AT is the averaging time of 25550 days (365 d y^{-1} x 70 y) (Kumar et al. 2013), and CSF is the cancer slope factor, which was 7.3 mg kg^{-1} d^{-1} for BaP ingestion and 0.13 mg kg^{-1} d^{-1} for BaP inhalation.

Eq.1 $$Risk = \frac{Cs \times IR \times CF \times EF \times ED}{BW \times AT} \times CSF$$

Monte Carlo simulation was employed to conduct probabilistic risk assessments following the U.S. EPA (2001b) method. Eq. 1 was used with the same inputs as the deterministic risk assessments, except where indicated in the results. Cs, IR, and EF were assumed to have lognormal or uniform distributions, and all Monte Carlo simulations were set for 10,000 trials. Oracle™ Crystal Ball (11.1.2.2.0) software was used for Monte Carlo simulations.

Excess cancer risk comparisons were made with four other oil regions from the literature. Four studies (Bojes & Pope, 2007; Duke & Albert, 2007; Skrbic & Durisic-Mladenovic, 2009; Tiwari et al. 2011) that reported PAH concentrations for the 16 priority PAHs used in this study were selected and concentrations were converted to BaP equivalents using PEF from Table 3.1. Excess lifetime cancer risk was then assessed using equation 1 for the maximum and mean BaP equivalent concentrations using a 20 mg d^{-1} soil ingestion rate.

Health Canada considers increases in excess lifetime cancer risk below 10^{-5} to be negligible, and this threshold level was used in the interpretation of the results in this study (Health Canada 2010, 2012). To assess cancer risk of a population through the ingestion of soil, the dose as well as the cancer slope factor for the exposure pathway needs to be known. Soil ingestion rates used to calculate the dose were determined through a First Nations soil ingestion study conducted over a 13 day period (cf. Chapter 2).

3.4 Results

3.4.1 Soil PAH contamination

Eighteen soil samples were collected and analyzed for 16 priority PAHs and BaP equivalent concentrations at different distances from oil sands facilities in Cold Lake, Alberta and are summarized in Table 3.2 and Figure 3.1. There was no significant relationship between PAH concentration, expressed as BaP equivalents, and distance from an oil sands facility ($F_{(1, 16)} = 1.73$, $p = 0.21$). The maximum concentration of soil PAH was 99.78 ng g^{-1} of BaP equivalents sampled at 6.4 km from the nearest drilling pad. The second highest concentration of 79 ng g^{-1} was collected at the roadside adjacent to one of the oil sands facilities. The mean soil concentration of BaP equivalents was 15.7 ± 29.7 ng g^{-1}, the geometric mean 2.06 ng g^{-1}, and the median concentration was 1.99 ng g^{-1}. Most of the sampled soils had relatively low concentrations with 13 soil samples below 5 ng g^{-1} and 7 samples below 1 ng g^{-1}. The results obtained did not show a relationship between distance to *in-situ* oil sands facilities and soil PAH concentration.

3.4.2 Atmospheric PAH contamination

High-volume air sampling collected particulate and gaseous PAH samples for 10, 24 hour sampling periods (Figure 3.2). The mean concentrations of BaP equivalents were 0.0043 ± 0.013 ng m^{-3} and 0.032 ± 0.007 ng m^{-3} for the particulate and gaseous phases, respectively. The gas phase PAH concentration, expressed as BaP equivalents, is an order of magnitude greater than PAHs on particulates. There was no correlation between particulate and gaseous PAH concentration. The maximum particulate PAH concentration of 0.042 ng g^{-1} was obtained on the fifth day; the maximum gaseous PAH concentration of 0.215 ng g^{-1} was obtained on the tenth day.

3.4.3 Cancer risk via soil ingestion

Our earlier work (see Chapter 2) indicated that the 95th percentile soil ingestion rate is 361 mg d^{-1} (Si as the soil tracer), and the median rate is 37 mg d^{-1} using the mean of Al and Si as the soil tracers. Using a deterministic risk assessment approach, the calculated excess lifetime cancer risk at the maximum PAH concentration observed in this study was 3.29E-06 at the 95th percentile soil ingestion rate, assuming exposure for 365 days per year (Health Canada 2012). In a Canadian climate, daily exposure for a full year seems unlikely because of the relatively harsh winter. A second exposure frequency of 153 days a year was also employed for the risk assessment. This value represents daily exposure for the period from May through September. The excess lifetime cancer risk associated with this exposure at 361 mg d^{-1} soil ingestion rate, and the maximum BaP equivalent soil concentration provided a value of 1.38E-06. There is not a significant increased risk of cancer at this high soil ingestion rate and the median soil ingestion rate the lifetime cancer risk was far lower (i.e., 1.41E-07), substantially below 1E-05. Furthermore, the mean and geometric mean BaP equivalent concentration of the collected Cold Lake soil samples (i.e., 15.7 ng g^{-1} and 2.06 ng g^{-1} respectively) also yielded risk values that are substantially below 1E-05 (i.e., 5.17E-07 and 6.80E-08 at the 95th soil ingestion rate of 361 mg d^{-1} and 365 day yr^{-1} exposure).

Not surprisingly, the soil ingestion rate employed has a large effect on calculated excess lifetime cancer risk. At an assumed 365 days per year exposure (Figure 3.3) a significant increase in cancer risk occurs at a soil concentration of 304 ng g^{-1} BaP equivalents for the 95th percentile soil ingestion rate. If lifetime cancer risk is calculated using the median soil ingestion rate of 37 mg d^{-1}, lifetime cancer risk meets the aforementioned 1 case per 100,000 threshold at a BaP equivalent soil concentration of 2962 ng g^{-1}. The adult soil ingestion rate of 20 mg d^{-1} recommended by Health Canada for HHRA of contaminated sites would require a soil concentration of 5479 ng g^{-1} of BaP equivalents to reach an increased cancer risk of 1 case per 100,000 people. Predictably, with an assumed exposure frequency of 153 days per year (Figure 3.4), which would represent non-winter months from May through September only, excess lifetime cancer risk in excess of 1E-05 would only be observed at a much higher PAH concentrations. Using an EF of 153 day yr^{-1}, at soil ingestion rates of 361 mg d^{-1}, 37 mg d^{-1}, and 20 mg d^{-1}, the 1.0E-05 cancer risk threshold occurs at BaP equivalent concentrations of 724 ng g^{-1}, 7066 ng g^{-1} and 13072 ng g^{-1}.

The BaP oral slope factor used in this study indicates the risk of gastric (stomach) cancer through ingestion. In Alberta, Canada annual age-standardized new cases of stomach cancer are 7 per 100,000 (Canadian Cancer Society 2012). Therefore, even at the maximum PAH concentration in this study, the median soil ingestion rate of 37 mg d^{-1} does not significantly increase the risk of gastric cancer beyond the current background level (i.e., 7 cases per 100,000). Even at higher soil ingestion rates there is no significant increase in excess cancer risk (Table 3.3). Using the 90th percentile soil ingestion rate of 152 mg d^{-1} and assumption of 365 d yr^{-1} exposure, the risk assessment predicts less than 0.1 new cancer case per 100,000 over existing background. Using the 95th percentile soil ingestion rate of 361 mg d^{-1}, the predicted excess risk is still less than 0.1 additional case per 100,000 people for an EF of 153 d yr^{-1}. Thus, the results obtained, which are summarized in Table 3.3, indicate that the increased risk of gastric cancer associated with the median and mean PAH concentrations found in the soils from the Cold Lake region is low and may be considered negligible.

Excess cancer risk comparisons were made with four other oil regions from the literature using the Health Canada adult soil ingestion rate for HHRA of 20 mg d^{-1}. The results for the Niger Delta, Nigeria; Texas, USA; Vojvodina, Serbia; and Mathura, India and are summarized in Table 3.4. Although these regions had must higher PAH concentrations than the Cold Lake region, there was still negligible excess cancer risk. The Texas, USA location had the greatest cancer risk level with 1.12E-05 for exposure at the maximum measured BaP equivalent concentration.

3.4.4 Probabilistic risk assessment

Deterministic risk assessments (Figure 3.3 and 3.4) have been criticized for being too conservative (i.e., unnecessarily inflate risk). To account for the uncertainty of input variables, probabilistic risk assessments have been recommended (Öberg and Bergbäck 2005; U.S. EPA 2001a). Excess lifetime cancer risk was assessed using the probabilistic Monte Carlo simulation approach (Figure 3.5), the aforementioned BaP equivalent soil concentrations, and the soil ingestion rates from the Cold Lake soil ingestion study (cf. Chapter 2). Calculated lifetime cancer risk in the Cold Lake region is negligible with 95% of the population associated with a risk level almost tenfold below the 10^{-5} threshold (input variables in Table 3.5). Excess lifetime cancer was also calculated using a Monte Carlo simulation, the maximum sampled BaP equivalent soil concentrations (Figure 3.6) and the aforementioned soil ingestion rates from the Cold Lake study (input variables Table 3.6). Again calculated lifetime cancer risk is negligible, below the 10^{-5} threshold.

A sensitivity analysis using Spearman’s rank correlation coefficient was performed on the Monte Carlo risk analysis equation used for the probabilistic risk analysis (Öberg and Bergbäck 2005). The three variables examined were Cs, EF, and IR (Table 3.7). All three variables were positively correlated with lifetime cancer risk, with ingestion rate having the largest impact on lifetime cancer risk, closely followed by Cs, with EF having the smallest influence on risk.

3.4.5 Cancer risk via inhalation

Atmospheric samples of PAHs were collected near the base camp, approximately 10 km from the in-situ mining facilities. Inhalation rates recommended by Health Canada were used to assess excess lifetime cancer risk (Table 3.8) to recorded daily concentrations of BaP equivalents. Using the Health Canada average adult inhalation rate of 16.6 m^3 d^{-1} the calculated excess lifetime cancer risk was far below the aforementioned threshold of 1E-05, even at the maximum measured atmospheric BaP equivalent concentration of 2.15E-04 ng m^{-3}. Health Canada recommends a higher inhalation rate for construction workers. If this inhalation rate of 1.4 m^3 hr^{-1} is assumed for individuals living traditional lifestyles in the Cold Lake region the calculated excess risk is still well below 1E-05, even at the maximum BaP concentration. The calculated excess risk at the maximum PAH concentration is 1.53E-09.

3.5 Discussion

3.5.1 Soil and atmospheric PAH contamination

This study was designed to assess the excess lifetime cancer risk posed by exposures to PAHs in a region of Alberta that contains numerous bitumen extraction facilities. The measured soil BaP equivalent concentrations from this study were all well below BaP guideline levels of 0.7 mg kg^{-1} for lifetime cancer risk (CCME 2010). The highest concentration detected in the soils analyzed in this study (i.e., BaP equivalent 0.0998 mg kg^{-1} and median BaP 0.002 mg kg^{-1}) was observed approximately 6.37 km away from the nearest drilling pad. It was expected that PAH concentrations would follow a similar trend to that seen in Kelly et al. (2009), where PAH concentrations were elevated up to 50 km from the oil sands upgraders in the Athabasca oil sands region. Although there is *in-situ* extraction in the Athabasca region it is dominated by bitumen extraction through open pit mining. Furthermore, the several upgrading facilities in the Athabasca region are suspected of being major sources of atmospheric PAH deposited in the surrounding area (Kelly et al. 2009). The Cold Lake region, despite being a large oil sands region, has a very different industrial landscape compared to the Athabasca region. The Cold Lake oil sands operations extract all bitumen through *in-situ* steam extraction methods and the extracted bitumen is transported to Lloydminster or Edmonton for upgrading and refining. The focus on *in-situ* extraction and lack of upgrading or oil refining in the Cold Lake region may explain the comparatively low PAH concentrations observed in Cold Lake. The PAH concentrations found in the soil from this study suggest that *in-situ* oil sands development in the Cold Lake region do not contribute substantial increases to PAH concentrations in sampled soil. This is contrasted to what previous research has observed in the Athabasca region of Alberta that has bitumen upgrading and open pit mining.

3.5.2 Risk assessment

The results obtained show an increased excess lifetime risk of cancer above the 10^{-5} threshold with the 95th percentile soil ingestion rate of 361 mg d^{-1} (cf. Chapter 2) (Figure 3.3 and Table 3.3) and a 365 day per year EF. This EF will almost certainly contribute to an unrealistically inflated cancer risk due to the likely changes in seasonal soil ingestion rates. An individual with an IR_{soil} at the 95th percentile may quite reasonably be expected to have a much lower IR_{soil} during winter months. Unfortunately, there has never been a soil ingestion study that addresses seasonal variation in ingestion rates. As such the likelihood, magnitude and direction of a seasonal change in IR_{soil} is unknown. It would be reasonable to assume that soil ingestion rates during warmer months, when First Nations' people who follow a traditional lifestyle spend greater amounts of time outside, would be substantially higher than during the winter months. Therefore, an EF of 153 days, encompassing the months May through September, was also used for calculating cancer risk.

The CCME has set soil quality guidelines for BaP at 700 ng g^{-1} for the protection of human health (CCME 2006, 2010). The guidelines have been created with an adult soil ingestion rate of 20 mg d^{-1}, and this ingestion rate may not be applicable to all populations. Although, the 37 mg d^{-1} median soil ingestion rate from the Cold Lake First Nations soil ingestion study is almost twice the recommended IR_{soil} for adults, the risk estimates presented here indicate that the excess lifetime cancer risk associated with levels below the CCME soil quality guidelines, using this higher IR_{soil}, are below 1E-05 and may be regarded as negligible.

Probabilistic risk assessments give a better understanding of the cancer risk to a population instead of just point estimates, at the measured Cold Lake soil ingestion rates (Figure 3.5 and 3.6). The calculated excess lifetime cancer risk associated with exposures to soil PAH levels observed at Cold Lake are well below 1E-05 and can therefore be regarded as negligible.

In terms of increase in cancer rates as a consequence of inadvertent ingestion of contaminated soil, there is no significant risk in the Cold Lake region (Table 3.3). The background incidence of gastric cancer is 7E-05, and even at the 95% IR_{soil} rate and maximum BaP equivalent soil concentrations measured in this study there was not an appreciable increase above background (Canadian Cancer Society 2012). One study that looked at cancer risk from PAH exposure through settled house dust ingestion found much higher excess cancer risk than in this study. At the maximum PAH concentration in the study by Maertens et al. (2008), and a 50 mg d^{-1} dust ingestion rate, they found 27.4 extra cases of cancer per 100,000 people compared to less than 1 excess cases per 100,000 people in this study for the maximum PAH concentration. Williams et al. (2013) investigated the risk of excess cancer from exposure to settled house dust and soil adjacent to parking lots sealed with coal-tar pavement in which PAH is a major constituent and used a distribution for IR with a mean of 27 mg d^{-1}. Exposure to settled house dust had excess cancer risk of 1.8 cases per 100,000 people and 1.2 cases per 10,000 people at the 50th and 95th percentile respectively. Exposure to soil adjacent to parking lots sealed with coal-tar had even a larger cancer risk of 7.3 cases per 100,000 at the 50th percentile and 4.3 cases per 10,000 people at the 95th percentile. Although, this study and the study by Maertens et al. (2008) and Williams et al. (2013) looked at very different environments (soil near oil sands facilities compared to settled house dust and soil in an urban/suburban setting), the cancer risk attributable to PAHs in soils near an *in-situ* oil sands facility might be reasonably regarded to be negligible, whereas there is significant risk associated with ingestion of PAHs in settled house dust and soil particularly if adjacent to coal-tar sealant.

However, a study by Wang et al. (2011) on the lifetime cancer risk from exposure to PAH in urban street dust, the mean cancer risk level from ingestion of dust was 2.51E-6, and thus, considered negligible. The mean BaP equivalent concentration from the study by Wang et al. (2011) was 0.47 mg kg^{-1}, which is much greater than the average for Cold Lake soils of 0.016 mg kg^{-1}. Man et al. (2013) assessed lifetime cancer risk from PAH exposure using 12 different soils from different land use types in Hong Kong. At the median level, no site had cancer risk above the 10^{-5} threshold, and only one site, nearby a car dismantling workshop had excess cancer risk at the 95th percentile of 24 cases per 100,000 people. Among the other sites from the Man et al. (2013) study that may be expected to be have significant lifetime cancer risk but were found to be below the 10^{-5} threshold were an open burning site where large amounts of PAHs would be expected from combustion. Despite only one location having cancer risk above the 10^{-5} threshold, seven of the sampled sites had BaP equivalent concentrations above the average for Cold Lake of 0.016 mg kg^{-1} and ranged from 0.090 mg kg^{-1} to a the highest BaP equivalent concentration of 4.93 mg kg^{-1} near a car dismantling workshop. Although the Cold Lake region may be expected to have excess cancer risk as a result of the surrounding industry like many of these environments from the Wang et al. (2011) and Man et al. (2013) studies, with the exception of the car dismantling workshop risk was below the 10^{-5} level.

Despite there being negligible cancer risk in the deterministic scenarios, the deterministic risk assessments may still inflate the cancer risk over the true lifetime cancer risk. The probability distribution functions (PDF) (Figure 3.5) are likely far more representative of lifetime cancer risk in the Cold Lake region as a PDF accounts for distribution of some or all the variables in a risk assessment calculation (Öberg and Bergbäck 2005). The ingestion rate likely varies across a population, evidenced by the relatively high standard deviations from the soil ingestion study (cf. Chapter 2). Additionally, PAH concentrations are highly variable as well (Figure 3.1 and Table 3.2).

The same risk assessment was run with both a deterministic and probabilistic approach. The deterministic assessment, which used an IR_{soil} of 37 mg d^{-1}, and the mean soil PAH concentration yielded a cancer risk of 2.22E-8, and the probabilistic approach used a distribution for PAH soil concentration and IR_{soil} (Table 3.5), had a very similar mean cancer risk of 3.37E-8. The benefit of the probabilistic assessment and a cumulative probability figure (Figure 3.5) is the ability to discern that the mean risk level of 3.39E-8 accounts for more than 80% of the population. The actual cancer risk level to most of the population is even lower than the risk level calculated using the deterministic equation (Öberg and Bergbäck 2005). Without analyzing risk through the use of a PDF, the risk calculated may only represent a small part of the population.

There have been few studies in the literature that report PAH soil concentrations in petroleum extraction and refining regions, however, the concentrations that have been published are much greater than found in the Cold Lake oil sands region (Table 3.4). The mean BaP equivalent concentrations observed at other sites are at least an order of magnitude greater than those observed in this study. Only the mean concentration from a site in India, that reported a BaP equivalent concentration of 0.34 mg kg^{-1}, was below the Canadian CCME guidelines value of 0.7 mg kg^{-1} (Tiwari et al. 2011). The cancer risk estimates for the locations reported from the literature are also much greater than those calculated for Cold Lake, but still below the Health Canada threshold. There is not a significant cancer risk in any of the locations using 1E-05 excess cancer risk threshold. (Table 3.4) (Bojes and Pope 2007; Duke and Albert 2007; Skrbic and Durisic-Mladenovic 2009). It is important to note that the US EPA typically uses a 1E-06 cancer risk threshold, and under this jurisdiction there is a significant excess lifetime cancer risk at the Texas location. However, the industrial landscape at Cold Lake is different from the locations reported in the literature. With the exception of the Texas location, which had contaminated soils as a result of faulty oil storage facilities, all the other locations had an oil refinery in close proximity to soil samples of high PAH concentrations. This suggests that atmospheric deposition as a result of oil refining is contributing to regional soil contamination.

The sensitivity analysis (Table 3.7) using Spearman's rank correlation coefficient show soil ingestion rate had the largest effect on cancer risk (U.S. EPA 2001b). Sensitivity analysis data in other published studies is rare, but one study that also assessed cancer risk from PAHs, but through inhalation and dermal exposure also found EF to have the lowest influence on lifetime cancer risk in comparison to other variables (Chen and Liao 2006). Williams et al. (2013) also conducted a sensitivity analysis on the Cs and IR (dust and soil) variables and found for adults that the Cs parameter had the greatest influence when exposed to soil or house dust adjacent to a pavement where coal-tar was not used. If instead, coal-tar sealant was used, than IR_{soil} had a larger influence on cancer risk. This is similar to this study where both IR and Cs were found to have similar influence on lifetime cancer risk.

Although soil ingestion rates reported in the Cold Lake study (cf. Chapter 2) are higher than the current rates recommended to be used in HHRA, there is negligible cancer risk in the Cold Lake region. Even still, there is potential for increased cancer risk to a population that has a higher soil ingestion rate than the 20 mg d^{-1} recommended rate (Figure 3.6) if the higher soil ingestion rates are considered. Therefore it is advised that the potential for higher soil ingestion in different regions needs be taken into account when PAH concentrations are compared to the current soil quality guidelines of 0.7 mg kg^{-1} for BaP.

3.5.3 Cancer risk via inhalation

The atmospheric concentrations of PAHs are also below the threshold cancer risk value of 10^{-5}. The maximum BaP equivalent concentration was 0.21 ng m^{-3}, with a median atmospheric concentration of 0.0071 ng m^{-3}. One published study reported rural BaP equivalent concentrations in Canada as a median of 0.14 ng m^{-3}, with major cities such as Toronto and Winnipeg having median atmospheric BaP equivalent concentrations of 0.76 ng m^{-3} and 0.14 ng m^{-3} respectively (Government of Canada et al. 1994). The World Health Organization recommends a threshold of atmospheric BaP concentrations of 0.12 ng m^{-3} for 1 cancer case per 100,000 people (Boström et al. 2002; World Health Organization 2000). Current guidelines from the Alberta Government require an annual average BaP atmospheric concentration of 0.30 ng m^{-3} (Alberta Government 2013), and the Ontario Ministry of the Environment (MOE), specify an Ambient Air Quality guideline of 1.1 ng m^{-3} over a 24 hour period (WBK & Associates Inc. 2004). Concentrations above guideline values over the averaging period indicate increased risk of adverse health effects. Atmospheric BaP concentrations measured in the Cold Lake region are typically lower than what is seen in the major urban centres of Alberta. WBK & Associates Inc. (2004) report the annual average BaP concentrations in Calgary and Edmonton during this time period, with a 7 year median concentration of 0.12 ng m^{-3} in Calgary and 0.14 ng m^{-3} in Edmonton. The maximum annual average concentrations were much greater than this at 0.37 ng m^{-3} and 0.41 ng m^{-3} in Calgary and Edmonton, respectively. The median concentration found in Cold Lake is much lower than typical urban concentrations, with only one day during the study providing a concentration of similar magnitude to typical urban BaP concentrations. This atmospheric concentration in Cold Lake is fairly representative of a rural region, with the median concentration far below air quality guideline levels for a 24 hour period and even below the annual average during the sampling period, recommended by the Alberta Government (Boström et al. 2002; Butler et al. 1993). This indicates that where the air sampler was located there is little to no atmospheric contamination during the sampling period.

The very low PAH concentrations that are typical of a rural environment make the lifetime cancer risk from inhalation of atmospheric PAHs negligible at all concentrations using the Health Canada daily inhalation rate for adults (Table 3.8). Even if a construction worker daily inhalation rate is used for a 24 hour period and the maximum PAH concentration, the excess risk is 1.34E-09, still far below the 10^{-5} value threshold. Although, this value is likely not an accurate representation as a typical construction worker inhalation rate is used for only 8 hour periods (or time spent working). It is difficult to know how representative these concentrations are of the region as it is from a 10 day period in one location, but the results obtained suggest a negligible cancer risk. Furthermore, other studies that compared excess cancer risk from PAH inhalation exposures to ingestion exposures have noted that inhalation exposures are relatively small contributions to risk (Butler et al. 1993; Wang et al. 2011).

3.6 Conclusions

This study conducted a lifetime cancer risk assessment to evaluate risks posed by exposure to PAHs in soil and air in an area in close proximity to in situ bitumen extraction facilities, and the risk analyses employed empirically measured soil ingestion rates determined at the time of environmental sample collection. The deterministic risk assessment and PDF associated with IR_{soil} from the First Nations soil ingestion study (cf. Chapter 2) both indicate that cancer risk is below the 10^{-5} threshold, and this would be considered negligible in most jurisdictions. Moreover, the PDF showed that 95% of the population is below the risk threshold of 1E-05. Additionally, the risk associated with inhalation exposure to PAHs measured during this study was considered to be negligible. The hypothesis that there is an increased excess lifetime cancer posed to First Nations people in the Cold Lake region from exposure to PAH is not supported by this study.

3.7 References

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Table 3.1: List of the 16 priority PAHs measured for this study and their Potency Equivalency Factors (Schneider et al. 2002).

PAH Compound	Potency Equivalency Factor
Naphthalene	0
Acenaphthylene	0.01
Acenaphthene	0
Fluorene	0
Phenanthrene	0
Anthracene	0.01
Fluoranthene	0.01
Pyrene	0
Benz[a]anthracene	0.1
Chrysene	0.01
Benzo[b]fluoranthene	1
Benzo[k]fluoranthene	0.1
Benzo[a]pyrene	1
Indeno[1,2,3-cd]pyrene	0.1
Dibenz[a,h]anthracene	1
Benzo[g,h,i]perylene	0.01

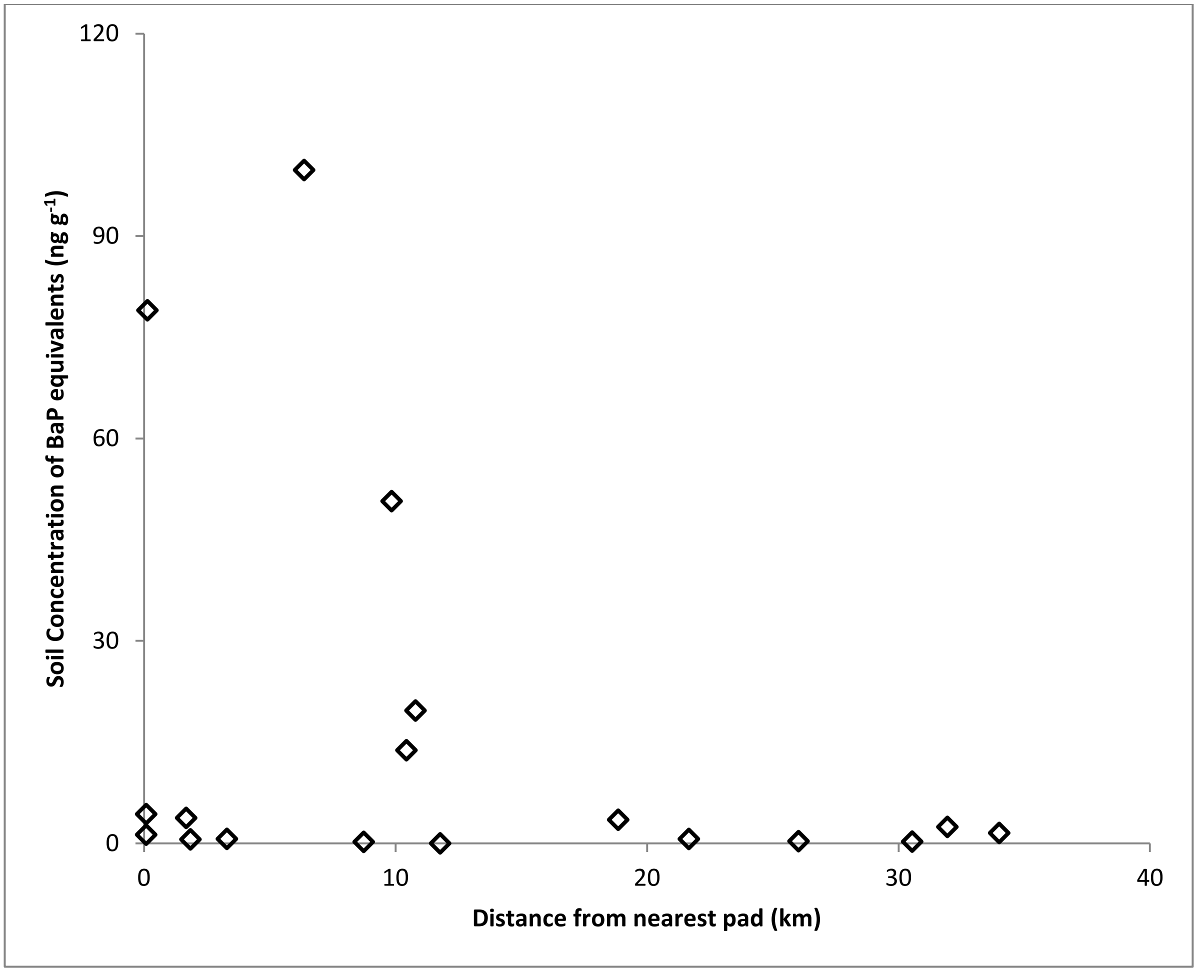

Figure 3.1: The concentration of PAHs, expressed as BaP equivalents, in soil samples collected at Cold Lake, Alberta as a function of distance from the nearest oil pad.

Table 3.2: The 16 priority PAH concentrations, ΣPAH, and BaP equivalents, in soil samples collected in Cold Lake, Alberta. Detected concentrations in samples that were below the minimum detection limit had a value substituted with half the detection limit. Values that were not detected are assumed 0 and marked with a -.

Sample #	2	3	4	5	7	8	10	12	13	14	15	17	21	22	23	24	26	28
Distance from nearest pad (km)	21.7	0.1	11.8	0.1	1.9	3.3	32	30.6	8.7	34	10.8	18.9	1.7	6.4	9.9	26	10.4	0.1
	PAH Concentration (ng g^{-1})																	
Naphthalene	-	1.9	5.1	-	27.5	-	26.4	-	-	-	100.5	-	-	-	-	-	-	-
Acenaphthylene	-	-	-	38.2	39.9	30.4	-	24.7	22.5	22.3	15.9	22.4	20.5	6.3	17.5	30.9	17.2	93.5
Acenaphthene	-	-	-	1.1	1.9	-	-	-	0.5	3.8	-	-	18.1	8.7	-	11	39.7	-
Fluorene	-	0.6	-	-	20.5	-	0.4	-	-	-	0.1	1	4.9	2.7	-	-	2.4	1.4
Phenanthrene	-	5.9	-	-	23.5	-	1.4	-	-	-	1.9	12.5	29.2	35.4	4.3	-	33.1	5.4
Anthracene	-	-	-	-	7.8	-	-	-	-	-	3.4	-	-	-	0.4	2	6.1	0.6
Fluoranthene	-	11.8	-	0.7	7.6	-	4.2	0.8	-	0.7	13.8	5.8	5.9	154.2	18.4	-	14	2
Pyrene	-	46.5	-	-	-	-	6.1	2	-	-	13.5	2.6	7	66.1	18.6	-	15.3	6.2
Benz[a]anthracene	-	4.4	-	-	-	-	0.6	-	-	0.6	5.2	0.6	-	5.3	9	-	3.8	0.1
Chrysene	0.7	122.1	0.3	1	4.3	0.7	5.2	1.1	-	2.7	10.8	6	6	33	19.8	-	8.1	1.2
Benzo[b]fluoranthene	0.6	46.2	-	0.9	-	0.4	1.7	-	-	1	10.5	2.9	3.5	52.9	25.3	-	8.6	3.3
Benzo[k]fluoranthene	-	11.8	-	-	-	-	-	-	-	-	4.2	0.3	-	196.5	9.5	-	1.9	-
Benzo[a]pyrene	-	18.8	-	-	-	-	0.6	-	-	0.6	7.1	0.6	-	17.9	19.1	-	3.8	-
Indeno[1,2,3-cd]pyrene	-	26.2	-	-	-	-	-	-	-	-	6.5	-	-	66.8	16.3	-	3.3	-
Dibenz[a,h]anthracene	-	8	-	-	-	-	-	-	-	-	-	-	-	-	2.3	-	-	-
Benzo[g,h,i]perylene	1.9	35.4	-	-	-	-	-	-	-	-	5.7	-	-	20.9	16.4	-	4.8	2.6
ΣPAH	3.2	339.6	5.4	41.9	133	31.5	46.6	28.6	23	31.7	199.1	54.7	95.1	666.7	176.9	43.9	162.1	116.3
BaP Equivalents	0.7	79.0	3.0E-03	1.3	0.6	0.7	2.4	0.3	0.2	1.5	19.7	3.5	3.8	99.8	50.7	0.3	13.8	4.3

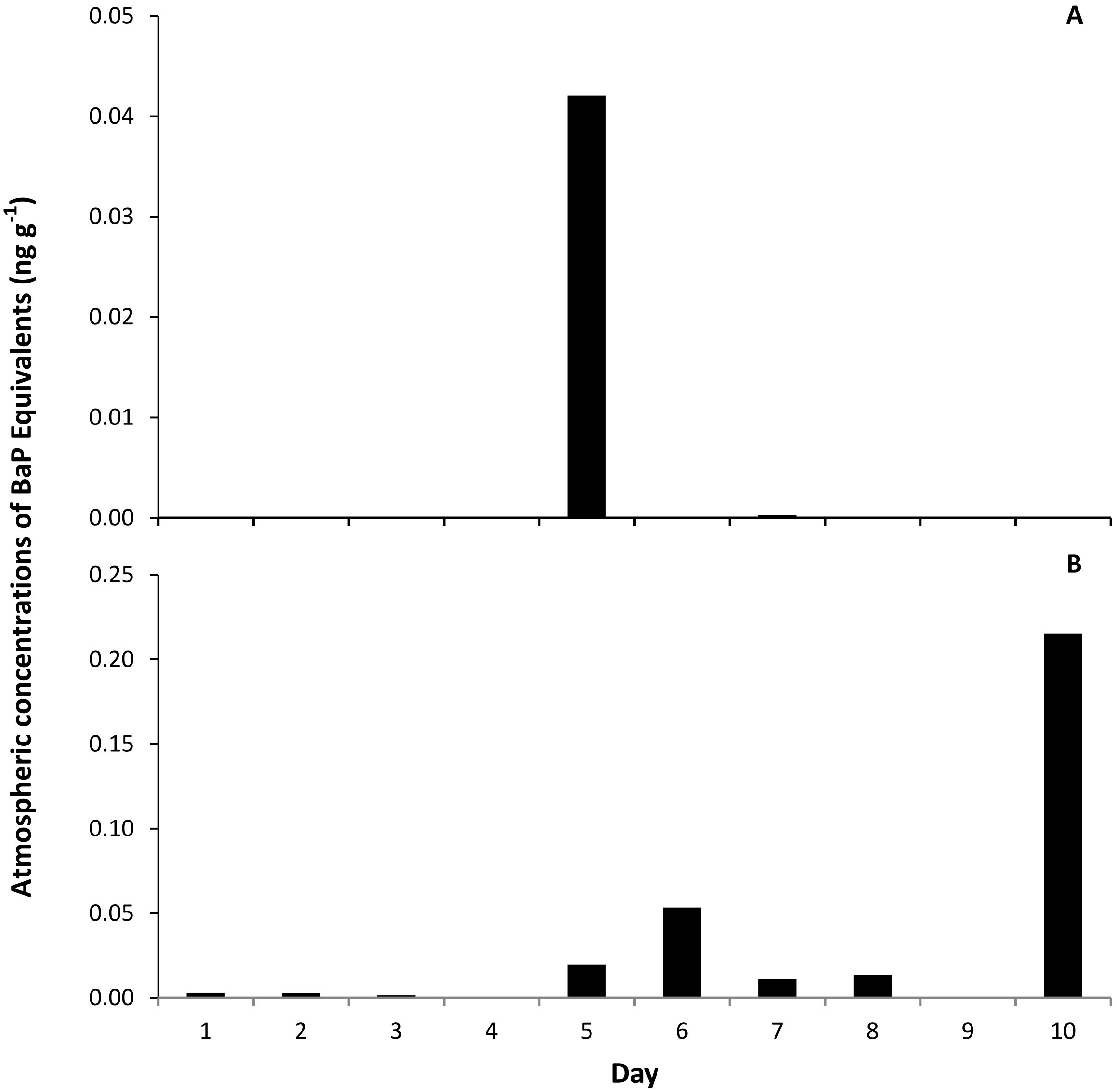

Figure 3.2: Atmospheric concentrations of PAHs expressed as Benzo[a]pyrene equivalents. Panel A shows particulate PAH concentrations, and panel B shows gas-phase PAH concentrations. For individual PAH concentrations see Appendix H and Appendix I.

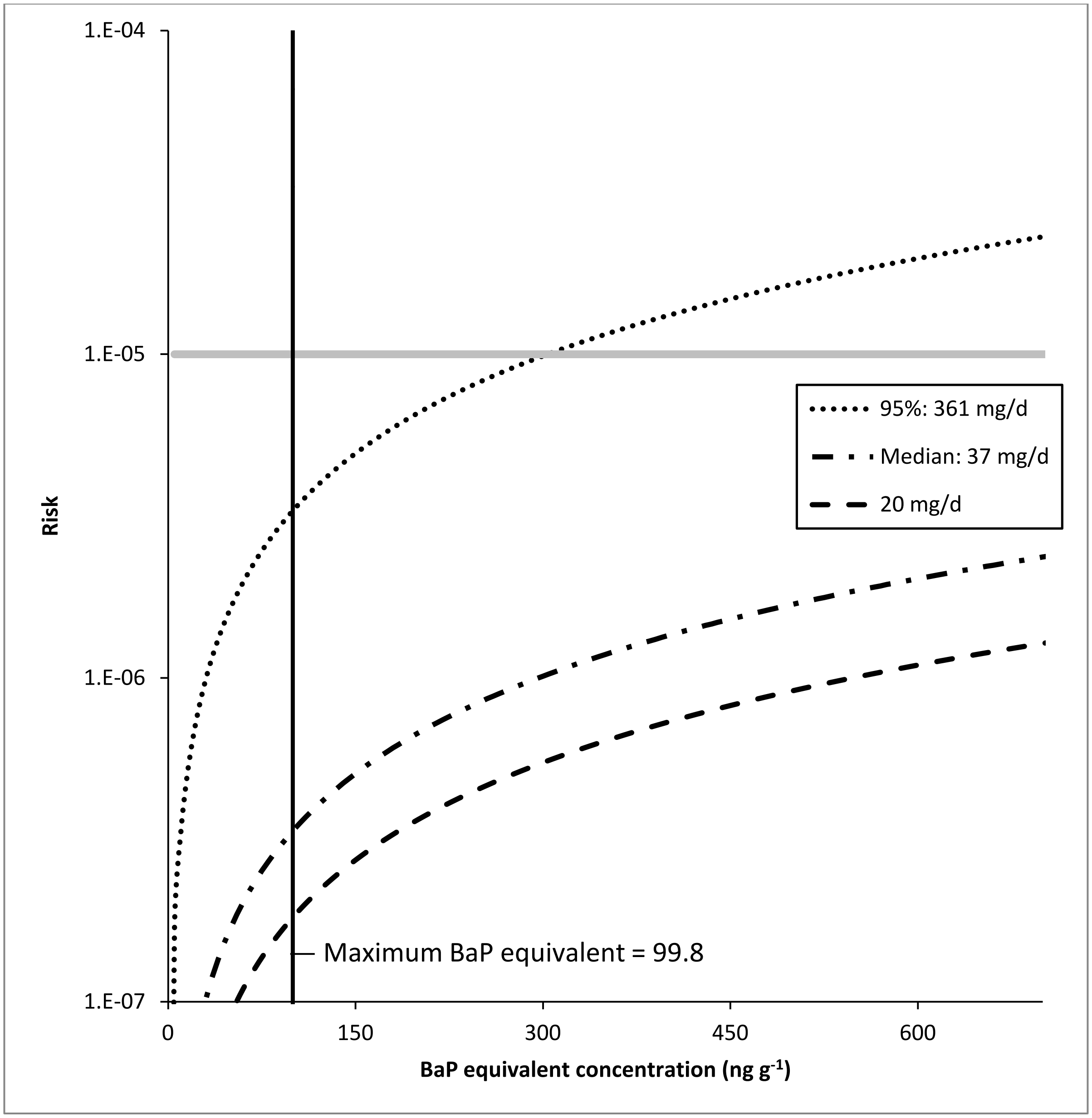

Figure 3.3: Calculated excess life time cancer risk as a function of soil PAH concentration (expressed as BaP equivalents) for different soil ingestion rates. Assumed exposure frequency of 365 days per year. The solid grey line denotes an excess lifetime risk of 1 extra cancer case per 100,000 people. This 1E-05 risk occurs at a BaP concentration of 304 ng g^{-1}, 2962 ng g^{-1}, and 5479 ng g^{-1} for soil ingestion rates of 361 mg d^{-1}, 37 mg d^{-1}, and 20 mg d^{-1}, respectively. The solid black vertical line denotes the maximum BaP equivalent concentration sampled in Cold Lake region. The three chosen soil ingestion rates are the 95th percentile (361 mg d^{-1}) and the median rate (37 mg d^{-1}) obtained in the Cold Lake soil ingestion study. The ingestion rate value of 20 mg d^{-1} is the value recommended by Health Canada for adult HHRA of contaminated sites.

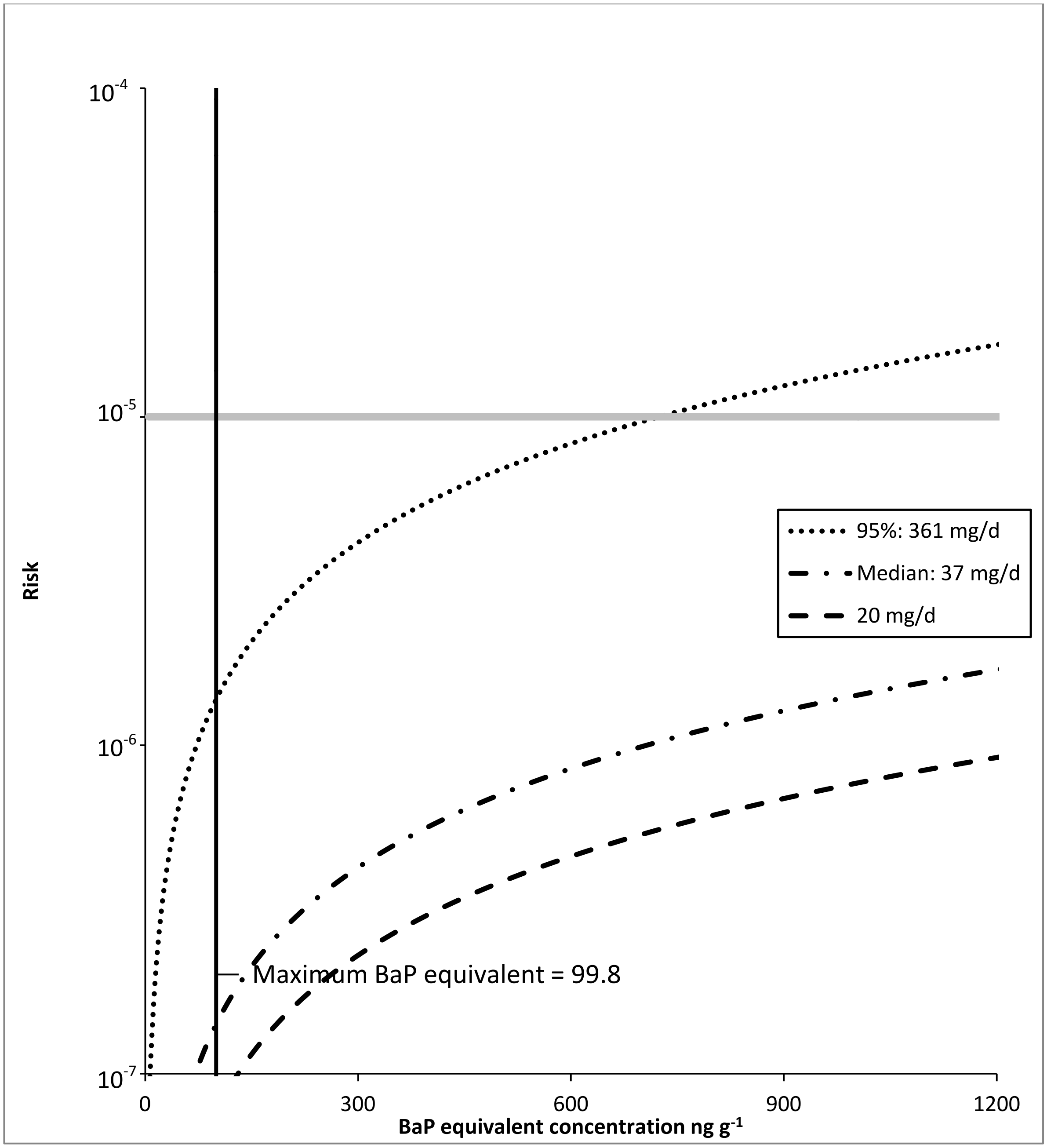

Figure 3.4: Calculated excess life time cancer risk as a function of soil PAH concentration (expressed as BaP equivalents) for different soil ingestion rates. Assumed exposure frequency of 153 days per year. The solid grey line denotes an excess lifetime risk of 1 extra cancer case per 100,000 people. This 1E-05 risk occurs at a BaP concentration of 724 ng g^{-1}, 7066 ng g^{-1}, and 13072 ng g^{-1} for soil ingestion rates of 361 mg d^{-1}, 37 mg d^{-1}, and 20 mg d^{-1}, respectively. The solid black vertical line denotes the maximum BaP equivalent concentration sampled in Cold Lake region. The three chosen soil ingestion rates are the 95th percentile (361 mg d^{-1}) and the median rate (37 mg d^{-1}) obtained in the Cold Lake soil ingestion study. The ingestion rate value of 20 mg d^{-1} is the value recommended by Health Canada for adult HHRA of contaminated sites.

Table 3.3: Rates of gastric cancer (per 100, 000 people) in Alberta, and the calculated increases in cancer incidence associated with exposure to contaminated soils in Cold Lake, Alberta. Calculated cases per 100,000 are the sum of the current background rates and the additional cases associated with exposure to contaminated soil.

Stomach Cancer	New cases per 100,000
Age-standardized incidence[a]	7
Age-standardized mortality[a]	4
Maximum PAH exposure 361 mg/d and 365 days/yr exposure frequency	7
Maximum PAH exposure 361 mg/d and 153 days/yr exposure frequency	7
Maximum PAH exposure 152 mg/d and 365 days/yr exposure frequency	7
Maximum PAH exposure 152 mg/d and 153 days/yr exposure frequency	7
Maximum PAH exposure 37 mg/d and 365 days/yr exposure frequency	7

a: (Canadian Cancer Society 2012)

Table 3.4: Comparison of BaP equivalent concentrations and lifetime cancer risk estimates for different locations reported in the literature. Risk calculations employed the Health Canada recommended adult soil ingestion rate of 20 mg d^{-1}. Cancer risk was assessed using the Maximum Cs, an ED of 70 years, an EF of 153 days, BW of 80 kg, an AT of 25550 days, and CSF_{oral} of 7.3 (mg kg^{-1} d^{-1})$^{-1}$.

		BaP Equivalent[a] (mg kg^{-1})		Lifetime Cancer Risk	
Location	n	Maximum	Mean	Maximum	Mean
Niger Delta, Nigeria[b]	8	5.3	2.0	4.05E-06	1.50E-06
Texas, USA[c]	3	14.7	11.0	1.12E-05	8.41E-06
Vojvodina, Serbia[d]	7	8.2	2.6	6.28E-06	1.98E-06
Mathura, India[e]	29	-	0.34	-	2.59E-07

a: PAH concentrations converted to BaP equivalent concentrations using toxic equivalency factors (Schneider et al. 2002)

b: Duke & Albert, 2007

c:Bojes & Pope, 2007

d:Skrbic & Durisic-Mladenovic, 2009

e:Tiwari et al. 2011

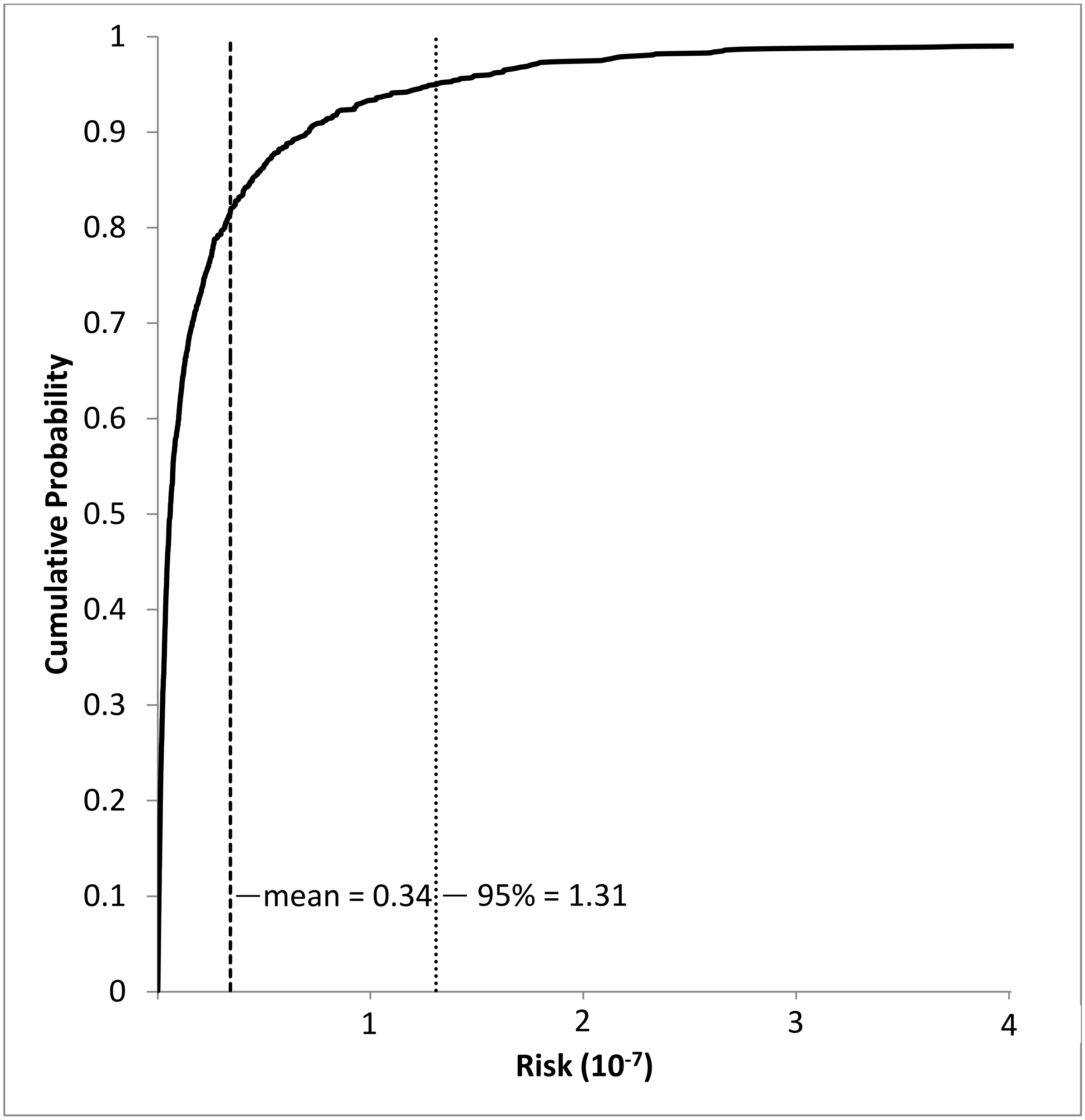

Figure 3.5: Cumulative probability distribution of a Monte Carlo simulation using 10000 trials and the input variables from Table 3.5. The soil concentration was the distribution of BaP equivalents measured at Cold Lake (Figure 3.1), the IR used is from Cold Lake soil ingestion study (cf. Chapter 2), and the exposure frequency is 153 days. The mean risk level was 3.42E-8, while the 95% risk level was 1.31E-7.

Table 3.5: Input variables for Monte Carlo simulation displayed in Figure 3.5.

Input Variable	Distribution	Parameters
Cs (mg/kg d.w. BaP equivalent)	lognormal	mean=0.0157, SD=0.0297
IR (mg/d)	lognormal	mean=52, SD=119
EF d/yr	uniform	min=max=153
ED yr	uniform	min=max=70
BW kg	uniform	min=max=80
AT	uniform	min=max=25550
CSF_{oral} (mg/kg-day)	uniform	min=max=7.3

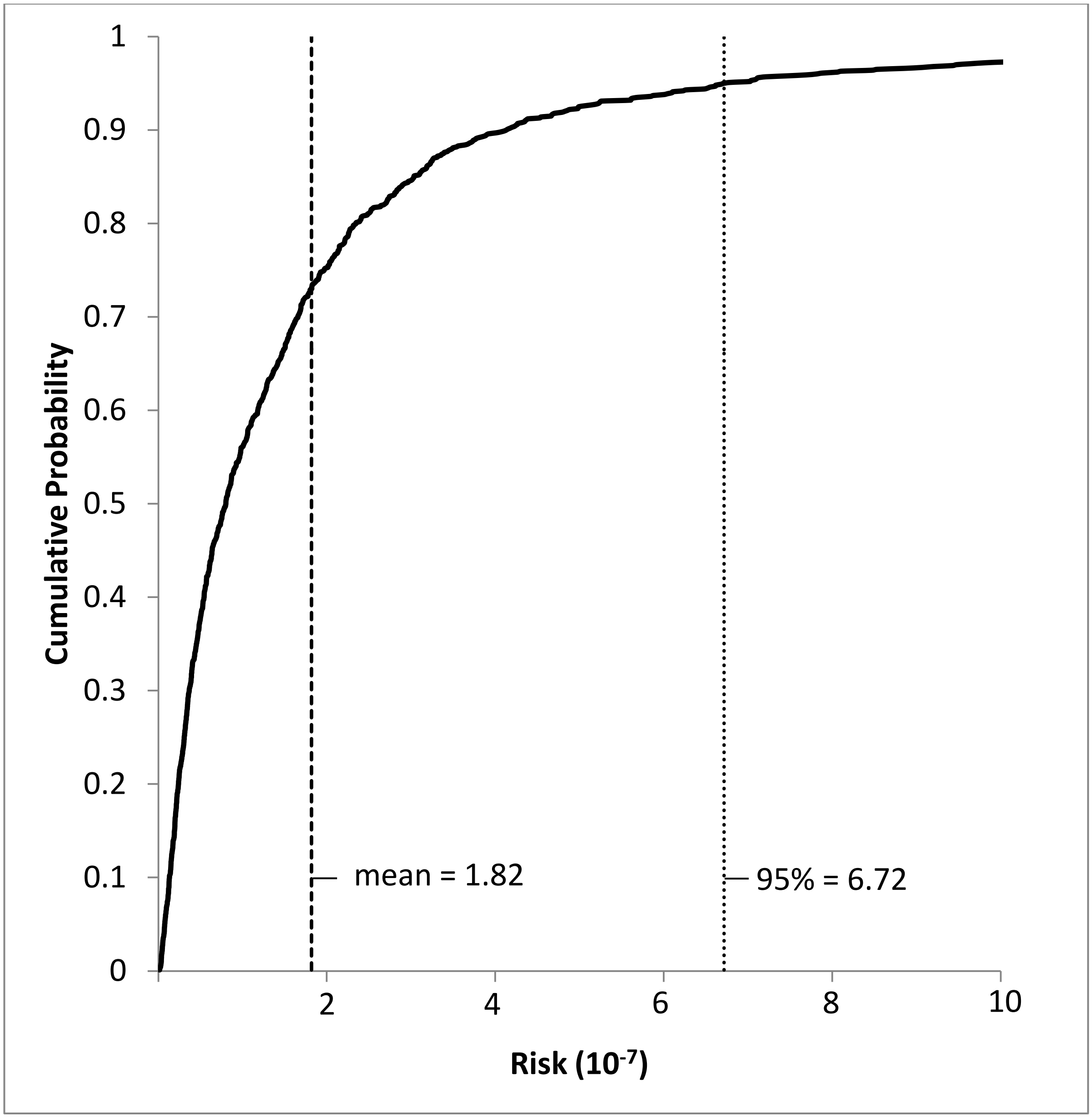

Figure 3.6: Cumulative probability distribution of a Monte Carlo simulation using 10000 trials and the input variables from Table 3.6. The soil concentration was the highest BaP equivalent concentration measured at Cold Lake (Figure 3.1), IR used were from Cold Lake soil ingestion study (cf. Chapter 2), and an exposure frequency of 153 days. The mean risk level was 1.82E-7, while the 95% risk level was 6.72E-7.

Table 3.6: Input variables for Monte Carlo simulation displayed in Figure 3.6.

Input Variable	Distribution	Parameters
Cs (mg/kg d.w. BaP equivalent)	uniform	min=max= 0.0998
IR (mg/d)	lognormal	mean=52 SD=119
EF d/yr	uniform	min=max=153
ED yr	uniform	min=max=70
BW kg	uniform	min=max=80
AT	uniform	min=max=25550
CSF_{oral} (mg/kg-day)	uniform	min=max=7.3

Table 3.7: Sensitivity analysis coefficient values using a Spearman's rank correlation coefficient.

Input Variable	Coefficient
Cs mg kg^{-1}	0.61
EF d yr^{-1}	0.11
IR mg d^{-1}	0.71

Table 3.8: Cancer risk estimates associated with inhalation of atmospheric PAH in Cold Lake, Alberta for different inhalation rates. Cancer risk below 1 extra case per 100, 000 people (1.0E-05) is considered negligible.

Inhalation Rate $m^3\ d^{-1}$ / BaP equivalent PAH concentration $\mu g\ m^{-3}$	16.6[a]	33.6[b]
2.15E-04 (maximum)	7.55E-10	1.53E-09
1.32E-08 (minimum)	4.64E-14	9.40E-14
3.68E-05 (mean)	1.27E-10	2.57E-10
7.08E-06 (median)	2.48E-11	5.03E-11

a: Inhalation rate of a typical adult.

b: Construction worker inhalation rate for 24 hours at (1.4 $m^3\ hr^{-1}$).

Chapter 4 — General Conclusions

4.1 Conclusions

First Nations' people practicing traditional activities are in closer contact with soil than typical urban or suburban residents. This study found that First Nations' people in the Cold Lake region of Alberta who engage in traditional activities have a higher inadvertent soil ingestion rate than that ordinarily employed for HHRA. This study followed the First Nations' pilot soil ingestion study in the Nemiah Valley, BC that also followed subjects who engage in traditional wilderness activities (Doyle et al. (2012). The latter study also noted that soil ingestion rates in these individuals are typically greater than those routinely employed for HHRAs of contaminated sites. The current study was the largest soil ingestion study on a Canadian population, and to my knowledge, the first combined soil ingestion risk assessment study in a region that might reasonably be expected to be contaminated with environmental carcinogens released by bitumen extraction.

The soil ingestion rates reported in this study for the mean of Al and Si were 52 mg d^{-1} with a 37 mg d^{-1} median respectively. Although, these are similar to the soil ingestion rates recommended by US EPA of 50 mg d^{-1}, it is larger than the Health Canada recommended rate of 20 mg d^{-1}. Furthermore, Wilson Scientific Consulting Inc. and Meridian Environmental Inc. (2006) in a report that reviewed and recommended soil ingestion rates to be used Health Canada in HHRA found a 95th percentile rate of 330 mg d^{-1}, smaller than the 361 mg d^{-1} Si soil ingestion rate determined from this study. Nevertheless, the qualitative recommendations for First Nations' individuals from Harper et al. (2007) (i.e., 400 mg d^{-1}) is greater than the rates reported in this study.

It is important to emphasize that the soil ingestion rates determined in this project are much lower than previously predicted or anticipated. Soil ingestion rates for First Nations people were expected to be much greater (from 400 mg d^{-1} upwards to grams per day) than soil ingestion rates based on studies on urban and suburban populations. However, the results obtained did not support this contention, both for this study and in the earlier Nemiah Valley study of Doyle et al (2012). This study made a concerted effort to ensure that the traditional activities performed during this study were typical of a traditional lifestyle. Perhaps it should be noted that some of the study participants were not from the Cold Lake region, and thus their typical daily routine had to be adjusted to suit the study environment, and this "activity adjustment" could have resulted in different soil ingestion rates than the subjects would typically experience. Furthermore, the traditional activities undertaken in this study might be considered to be moderate soil contact activities. Other traditional activities that are likely to contribute to higher soil contact, and that might be considered an integral part of many traditional lifestyles, include subsistence agriculture, collection and consumption of wild plants and roots, and participation in outdoor recreational activities such as rodeos. Additionally, traditional methods of food preparation such as the smoking or drying of meats and fish out of doors may be a significant contributor to soil ingestion rate through the adhesion of soil to meats and fish. Engagement in these activities might be expected to yield higher soil ingestion rates than those found in this study. The soil ingestion rates from this study can be used for HHRA of contaminated areas near First Nations' communities in wilderness areas, but further work may be necessary to encompass different lifestyles and activities.

The mass balance tracers used in this study are the same as those used in previous soil ingestion studies (Calabrese et al. 1989, 1997; Davis et al. 1990; Doyle et al. 2012; Stanek et al. 1995, 1997). Al and Si, which have previously been found to be the most reliable tracers, were also found to be the most reliable tracers in this study (Doyle et al. 2010; Stanek et al. 2011). Al and Si have high concentrations in all soils that were collected, low variability (with low CVs of 7% and 9% respectively), are not easily absorbed in the gastrointestinal tract, and are found in low concentrations in foods compared to other tracers (Calabrese et al. 1989, 1997; Doyle et al. 2012; Stanek and Calabrese 1991). However, as with previous soil ingestion studies, the rates determined in this study still show relatively high variability in ingestion rates that are difficult to eliminate due the assumptions employed in mass balance soil ingestion studies. All mass balance soil ingestion studies assume a 24 hour tracer transit time that allows for the daily determination of soil ingestion rates. However, the transit time of a tracer may vary between each subject as well as vary day to day and is dependent on other factors such as diet, age, and gender (Doyle et al. 2012; Stanek and Calabrese 1991; Madsen and Jensen 1989). In this study, a controlled diet was used to minimize transit time variability. Additionally, there is an assumed one to one correspondence between the ingestion of a tracer and fecal output which will increase the soil ingestion rate intra- and inter-subject variability. Study duration, or an increased number of participants can minimize some of the inter-subject or daily variabilty in soil ingestion rates by increased statistical power (Doyle et al. 2010). This study had a longer continuous study duration than previous soil ingestion studies which can minimize transit time misalignment as food tracer intake should balance with fecal tracer output (Stanek and Calabrese 1991).

This study was the largest aboriginal soil ingestion study, with 9 different participants followed over a 13 day period, resulting in 87 soil ingestion estimates, and it is one of the largest soil ingestion studies conducted to date. The results obtained are consistent with the aforementioned aboriginal soil ingestion study by Doyle et al (2012), and moreover, confirms the need to employ a separate soil ingestion rates for a rural or wilderness population considered in HHRAs of contaminated sites, and in the development of risk management activities (e.g., site remediation and soil clean-up criteria).

Human health risk assessments provide regulators and decision makers (e.g., contaminated site custodians) with the knowledge required to ensure that an area does not, or no longer, pose a risk to nearby inhabitants, and moreover, provide information to determine the need for risk management interventions (i.e., site clean-up, containment, access restriction). There has been recent speculation that inhabitants of areas located in close proximity to oil sands operations may be at increased risk of cancer due to increased exposures to contaminants such as metals and PAHs (CBC News 2013; Financial Post 2013). Although, most of the concerns relate to increased cancer risk in the surface mining areas such as the Athabasca oil sands region, it would be prudent to assess excess lifetime cancer risk posed to a First Nations' population in *in-situ* bitumen extraction areas such as those located in the Cold Lake region. In addition to the soil ingestion rate determination, this study also measured PAH concentrations in soil and air, and moreover, assessed the excess lifetime cancer risk posed to individuals in the area who engage in traditional wilderness activities. The results obtained indicate that the excess lifetime cancer risk is below the Health Canada cancer risk threshold of 10^{-5} for exposures from both inhalation and inadvertent soil ingestion, and far below the background Alberta cancer rates for the target organ (Canadian Cancer Society 2012). Although PAH exposures might be expected to increase the risk of other health effects, for the purposes of this study, cancer risk was deemed to be the adverse health effect since very high PAH concentrations would be required for acute effects. The measured concentrations of PAHs in air and soil collected in the Cold Lake region are typical of a rural area, and this suggests that unlike the Athabasca region, where atmospheric deposition of PAHs has been found to be elevated above historical background levels, and moreover, empirically linked to the oil sands industry, the *in situ* bitumen extraction at Cold Lake does not contribute to elevated PAH contamination (Kelly et al. 2009; Kurek et al. 2013). The hypothesis that exposures to contaminants from oil sands operations in the Cold lake region contribute to an increase in cancer risk was not supported by this study. It is reasonable to hypothesize that the absence of bitumen upgrading and refining activities in the Cold Lake region are critical determinants of the calculated negligible increase in risk associated with exposures to PAHs in air and soil.

4.2 Future research

The large variability in reported soil ingestion rates and the unknown effects of seasonality, lifestyle variability, or degree of urbanization, combined with the importance of inadvertent soil ingestion for HHRA, make this an area in need of further research. Additionally, soil is the most common contaminated medium in the federal contaminated sites inventory, and the dominant pathway that people are exposed to many non-volatile and semi-volatile contaminants on federal contaminated sites. There is a need to focus on different populations that may not be adequately protected by risk management measures that are based on an urban lifestyle soil ingestion rate. Military personnel, farmers, as well as First Nations' populations that engage in activities not covered in this study or the previous study by Doyle et al. (2012) may all be at an increased risk of adverse health effects due to the use of inaccurate soil ingestion rates. Some occupations such as construction workers may have an increased soil contact and may be an increased risk of harmful exposure through inadvertent soil ingestion. Determination of accurate occupational soil ingestion rates would be a laudable research objective for future studies. The current occupational soil ingestion rate for construction workers is 100 mg d^{-1}, but this value is based on an extrapolation of the aforementioned urban/suburban studies.

There is a lot of room in the field for novel studies as well as the development of new methods to more accurately assess inadvertent ingestion of soil. Mass balance soil ingestion studies are the most popular method of measuring soil ingestion as they produce soil ingestion rates that are representative of a population and have relatively simple methods and equipment requirements. However, mass balance studies have a number of concerns. All mass balance studies have relatively high variability, require a number of assumptions, a high investment of time, money and the need for strict subject compliance. Mechanistic methods, suggested by Wilson et al. (2013) calculate soil ingestion rate through an equation that accounts for adhesion of soil to hands, mouthing behaviour, amount dissolved in saliva and exposure time. This method still requires a number of assumptions for the equation but may be useful to calculate soil ingestion when a mass balance soil ingestion study is not possible. Particularly it may be useful for determination of activity specific soil ingestion rates because of the exposure time variable duration.

The results obtained did not show any significant increase in excess lifetime cancer risk in the Cold Lake region from exposure to PAHs in air and soil, and thus there is no indication that a larger scale risk assessment is necessary for this region. However, the bitumen extraction, processing and refining activities in the Athabasca region have been linked to far greater concentrations of PAHs than those reported here, and it would seem prudent to conduct a similar risk assessment for First Nations' people following traditional lifestyles in the Athabasca oil sands region.

4.3 References

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Appendix A - Consent form signed by soil ingestion study participants

Assessing Ingestion of Contaminated Soils in People Following Traditional Lifestyles near the Alberta Oil Sands

Jamie Doyle, Graham Irvine, and Jules Blais*, Department of Biology, Faculty of Science, (613) 562-5800 Ext 6650, Jules.Blais@uottawa.ca)*

Invitation to Participate: Volunteers will be invited to participate in the abovementioned research study conducted by Jamie Doyle and Jules Blais; six volunteers for the soil ingestion study and 15 volunteers for the ethno-cultural survey.

Purpose of the Study: The purpose of our research is to determine if soil ingestion estimates currently used in risk assessments of contaminated sites are adequately protective of people following traditional or subsistence lifestyles.

Participation: My participation will consist essentially of providing daily fecal samples and three daily urine samples for radiochemical/chemical analysis in a soil ingestion study; and/or participate in an ethno-cultural survey to determine their traditional food consumption and participation in traditional activities. Volunteers participating in the soil ingestion study will be provided food rations during the study (rations will be provided by the researcher).

Risks: No physical or emotional risks are anticipated from this research.

Benefits: My participation in this study will improve soil ingestion rate measurements and contribute to better estimates of soil exposure in areas where soil contamination is a concern.

Confidentiality and anonymity: I have received assurance from the researcher that the information I will share will remain strictly confidential. I understand that the contents will be used only for research purposes and that my confidentiality and anonymity will be protected by removing my name from any publications, and by keeping results securely in the possession of the two investigators.

Conservation of data: The data collected (fecal samples, analytical results, responses to interview questions) will be kept in a secure manner by storing in the offices of the two investigators (Jamie Doyle and Jules Blais) until publication within 5 years (i.e. until ca. 2017).

Compensation: There will be compensation for the participant of this study that has been negotiated with the Tribal Chiefs Association.

Voluntary Participation: I am under no obligation to participate and if I choose to participate, I can withdraw from the study at any time and/or refuse to answer any questions, without suffering any negative consequences. If I choose to withdraw, all data gathered until the time of withdrawal will be destroyed.

Acceptance: I, (participant)*,* agree to participate in the above research study conducted by Graham Irvine (M Sc candidate, Department of Biology, University of Ottawa*),* whose research is under the supervision of Jules Blais.

If I have any questions about the study, I may contact the researcher or his supervisor.

If I have any questions regarding the ethical conduct of this study, I may contact the Protocol Officer for Ethics in Research, University of Ottawa, Tabaret Hall, 550 Cumberland Street, Room 154, Ottawa, ON K1N 6N5

Tel.: (613) 562-5387

Email: ethics@uottawa.ca

There are two copies of the consent form, one of which is mine to keep.

Participant's signature: Date:

Researcher's signature: Date:

Appendix B - Fecal handling safe operating procedure

Radionuclide Analysis of Fecal Samples through Gamma Spectrometry

Fecal Sample Preparation Safe Operating Instruction (SOP)

Fecal Sample Preparation for Soil Ingestion Mass Balance Calculations

Last updated: July 20 2011

Scope:

The objective of this SOP is to prepare human fecal samples collected in soil ingestion study analysis with gamma spectrometry. The individual samples will be contained in pre-weighed 19” X 25” biohazard bags closed with a zip ties (sample bags). Several individual samples will be bagged within larger Biohazards bags closed with the zip tie (secondary containment bag). The larger bags will be contained within a regular plastic camping cooler. The samples should be frozen.

Hazards and Precautions

- Safety precautions must be taken in order to guard against the risk of infection from pathogens that may be present in un-sterilized feces. Wear PPE as required by the procedure. Dispose of wipes, gloves etc. in biohazard bags as per lab procedures.
- Wash hands with antibacterial soap and rinse well with water after working with these samples. Wipe down all equipment that could have been exposed to samples with 70% ethanol.
- Follow all existing lab and Emergency Response Procedures as required.
- Remember muffle furnace 2 requires PID settings to be reset before each sample run.*

Personal Protection Equipment (PPE) Required

- Lab coat
- Safety glasses
- Dust mask
- Nitrile gloves
- Biohazard bags and disposal
- Oven mitts

Materials

- Biohazard bags

- Analytical balance
- Evaporation dishes
- Oven in fumehood
- Crucibles and tongs
- Muffle furnace
- Mortar and pestle
- Falcon tubes
- Plastic spatulas
- Paintbrushes
- Compaction stand and piston
- Compaction press (machine shop – contact Hervé ext 6778)
- 8mL centrifuge tubes and PEFlon septa
- Epoxy and epoxy gun
- Gamma spectrometer

Procedure:

In lab	
1. Unseal outer container (camping cooler) and check for abnormalities (i.e., leaks in containment). If leak observed stop work and obtain guidance.	
2. Remove secondary containment bag with the individual samples and transfer into freezer – if required, unseal outer containment bag to deflate the bag to permit the bag to fit in the freezer.	
3. Leave samples in freezer for at least 24 hours to allow samples to fully freeze.	
4. Transfer samples to fume hood.	
In fume hood - drying	
5. Remove sample bags from the secondary containment bag and put empty secondary containment bag into biohazard disposal.	
6. Weigh each full sample bag and record weight and sample number.	
7. Cut sample bag seal and place sample bag in <u>pre-weighed</u> evaporation dish and fold over bag to allow moisture to evaporate from sample (be careful not to fold bag such that there is exposed sample outside of the evaporation dish).	
8. Weigh each dish with sample, record wet weight and sample number.	
9. Transfer evaporation dish into drying oven.	
10. Dry sample for 3 days at 95ºC.	
11. Weigh each dish with sample, record dry weight and sample number.	

12. Transfer dry sample to pre-weighed large crucibles (this may require the sample to be divided up into smaller chunks and record dry weight and sample number.	
In lab - Ashing	
13. Transfer crucibles to muffle furnace and ash at 550ºC using Program 4**	
14. Remove cool crucibles with tongs and record dry weight and sample number.	
15. Lightly de-aggregate ashed sample with glass pestle and transfer (using spatula and paintbrush) to pre-weighed falcon tubes labeled with sample number.	
16. Weigh falcon tubes and record ashed weight and sample number.	
17. Store ashed fecal sample in Falcon tube.	
18. Transfer sub-sample (~1.0g) to labeled and etched glass scintillation vial for independent ICPMS metals analysis, as deemed necessary.	
Compacting	
19. Compact samples into pre-weighed container: a. 8mL centrifuge tube – compact to maximum 4cm high, using piston and press located in machine shop (Macdonald basement, contact Hervé) OR compact by hand using piston	
20. Record packed sample weights and heights.	
Sealing	
21. Seal 8ml centrifuge tube with PEFlon septa and epoxy. Record sealing date.	
22. Wait until the sample's radionuclides of interest reach secular equilibrium before analyzing with gamma spectrometry for 82800s count time (Room 422). For soil ingestion analysis, equilibrium is reached between 222Rn, 214Pb and 226Ra after 21 days.	

*Muffle Furnace PID settings	**Muffle Furnace Program 4
G.SP: 750	Hb; band
Pb: 3	HbU: 25
ti: 365	Ramp to 200 ºC over 5 hours
td: 60	Ramp to 550 ºC over 6 hours

Pb2: 3 ti2: 217 td2: 36	Dwell over 10 hours. End

Appendix C - Table summarizing food portions for each participant for each meal/day of the Cold Lake soil ingestion study.

C.1 Liquids

ID	Meal	Water	Coffee	Tea	Apple Juice	Muskeg Tea	Muskeg and tamrac Tea	Mint Tea	Oasis Juice	V8-Fruit Juice	Nestea Iced Tea	5-Alive juice	Cream	Milk
		mL	mL	mL	mL	mL	mL	mL	mL	mL	mL	mL	mL	mL
		1	**200**	**200**	**200**	**200**	**200**	**200**	**200**	200	710	200	15	5
		Number of portions consumed												
A10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack		1											
A11	Bkfst	1255.682	1											
	Lunch													
	Dinner													
	Snack		1	1										
A12	Bkfst	1255.682	2										1	
	Lunch													
	Dinner													
	Snack		3										1	
A13	Bkfst	1255.682	2										2	
	Lunch		3										3	
	Dinner													
	Snack		2										2	40

A14	Bkfst	1255.682	1										1	
	Lunch													
	Dinner		3			1	1					1	3	
	Snack													
A15	Bkfst	1255.682	1										1	
	Lunch				1									
	Dinner													
	Snack			1				1						40
A16	Bkfst	1255.682												
	Lunch		1										1	
	Dinner													
	Snack		3	1									3	
A17	Bkfst	1255.682	1										1	
	Lunch		2										2	
	Dinner													
	Snack						1							
A21	Bkfst	1255.682	2										2	
	Lunch													
	Dinner													
	Snack		1										1	
A22	Bkfst	1255.682	2										2	
	Lunch													
	Dinner											1.25		
	Snack							1						
A23	Bkfst	1255.682	3										3	
	Lunch		10										10	
	Dinner													40
	Snack							1						
A24	Bkfst	1255.682	2										2	
	Lunch		2									1.25	2	

	Dinner		2									1.25	2	
	Snack		2					1					2	
A25	Bkfst	1255.682	1									1.25	1	
	Lunch													
	Dinner		2											
	Snack													
A26	Bkfst	1255.682	2										2	
	Lunch													
	Dinner											1.25		
	Snack		3										3	
B10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack		1	3										
B11	Bkfst	1255.682	2									1		
	Lunch													
	Dinner								1					
	Snack											2		
B12	Bkfst	1255.682	3						1					
	Lunch													
	Dinner													
	Snack													
B13	Bkfst	1255.682	3											
	Lunch		2						1					
	Dinner													
	Snack		2										2	
B14	Bkfst	1255.682	2										2	
	Lunch													
	Dinner		1						2				1	
	Snack													

B15	Bkfst	1255.682	2						1				2	
	Lunch				2									
	Dinner				1									
	Snack			1	2			1						
B16	Bkfst	1255.682	4										4	
	Lunch				2									
	Dinner				2			3						
	Snack													
B17	Bkfst	1255.682	2										2	
	Lunch											1		
	Dinner		2				2						2	
	Snack													
B21	Bkfst	1255.682	2										2	
	Lunch		3										3	
	Dinner		2										2	
	Snack		1				2						1	
B22	Bkfst	1255.682	2		1								2	
	Lunch													
	Dinner			2								1.25		
	Snack				3			3						
B23	Bkfst	1255.682	3		1							1.25	3	
	Lunch				1									
	Dinner			1								1.25		
	Snack		2									2.5	2	
B24	Bkfst	1255.682	3									1.25	3	
	Lunch													
	Dinner		1					3					1	
	Snack													
B25	Bkfst	1255.682	3									2.5	3	
	Lunch													

	Dinner							3				2.5		
	Snack													
B26	Bkfst	1255.682	3									1.25	3	
	Lunch													
	Dinner													
	Snack											2.5		
C10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack		1									1		
C11	Bkfst	1255.682	3									1		
	Lunch											1		
	Dinner			2					2					
	Snack		3							1				
C12	Bkfst	1255.682	1											
	Lunch													
	Dinner		9											
	Snack							1						
C13	Bkfst	1255.682	3											
	Lunch		2											
	Dinner								0.5					
	Snack		2											
C14	Bkfst	1255.682							2					
	Lunch													
	Dinner		2			1								
	Snack					1								
C15	Bkfst	1255.682	1.5											
	Lunch		2		1									
	Dinner													
	Snack		3	0.5				0.5						

C16	Bkfst	1255.682	1											
	Lunch							2						
	Dinner		1					3						
	Snack													
C17	Bkfst	1255.682	1											
	Lunch				1									
	Dinner				1									
	Snack						2							
C21	Bkfst	1255.682	1											
	Lunch													
	Dinner		1											
	Snack		1											
C22	Bkfst	1255.682	1											
	Lunch			1	1									
	Dinner			1								3.75		
	Snack							2						
C23	Bkfst	1255.682	3											
	Lunch											3.75		
	Dinner		2	1										
	Snack							1						
C24	Bkfst	1255.682	2											
	Lunch		1											
	Dinner													40
	Snack							1						
C25	Bkfst	1255.682	2											40
	Lunch			1										
	Dinner		3											40
	Snack							1				1		
C26	Bkfst	1255.682	2									1		
	Lunch			1										

	Dinner		3											
	Snack													
D10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack			1								2		
D11	Bkfst	1255.682										1		
	Lunch													
	Dinner													
	Snack		1									2	1	1
D12	Bkfst	1255.682												40
	Lunch													
	Dinner													
	Snack							3						40
D13	Bkfst	1255.682												
	Lunch													80
	Dinner		1											40
	Snack		1									1		
D14	Bkfst	1255.682										1		
	Lunch											1		
	Dinner		1									1	1	
	Snack						1							
D15	Bkfst	1255.682												80
	Lunch				1									
	Dinner													
	Snack			1										40
D16	Bkfst	1255.682			1									
	Lunch													
	Dinner							2						
	Snack				1									

D17	Bkfst	1255.682										1.25		
	Lunch				1									
	Dinner				1									
	Snack						4							
D21	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack						1							
D22	Bkfst	1255.682			1									
	Lunch				1									
	Dinner											2.5		
	Snack			1				2						
D23	Bkfst	1255.682												
	Lunch				1									
	Dinner											1.25		40
	Snack							1						
D24	Bkfst	1255.682										1		
	Lunch													40
	Dinner							2						80
	Snack													
D25	Bkfst	1255.682												40
	Lunch													
	Dinner													
	Snack													
D26	Bkfst	1255.682												
	Lunch													
	Dinner		1									1.25		
	Snack											1.25		
E10	Bkfst	1255.682												
	Lunch													

	Dinner													
	Snack											1		
E11	Bkfst	1255.682												
	Lunch													
	Dinner										1	2		
	Snack		2.5	2										
E12	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack		4					2			1			
E13	Bkfst	1255.682												
	Lunch										1			
	Dinner													
	Snack		3											
E14	Bkfst	1255.682	2									1		
	Lunch													
	Dinner													
	Snack		2								1			
E15	Bkfst	1255.682	3											
	Lunch										0.5			
	Dinner		1	4										
	Snack													
E16	Bkfst	1255.682	3		1									
	Lunch				1									
	Dinner							1						
	Snack			3										
E17	Bkfst	1255.682	2											
	Lunch			3	1									
	Dinner					1								
	Snack		5											

E21	Bkfst	1255.682	2	1										
	Lunch			5										
	Dinner													
	Snack													
E22	Bkfst	1255.682	2											
	Lunch											1.25		
	Dinner			3								1.25		
	Snack													
E23	Bkfst	1255.682	2	2										
	Lunch													
	Dinner													
	Snack													
E24	Bkfst	1255.682	1											
	Lunch				1									
	Dinner													
	Snack													
E25	Bkfst	1255.682										1.25		
	Lunch				1									
	Dinner											2.5		
	Snack		2	2								1.25		
E26	Bkfst	1255.682	1											
	Lunch													
	Dinner													
	Snack													
F10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack		1	2								1		
F11	Bkfst	1255.682												
	Lunch											1		

	Dinner													
	Snack		3	1										
F12	Bkfst	1255.682		3										
	Lunch													
	Dinner													
	Snack													
F13	Bkfst	1255.682	2											
	Lunch		2											
	Dinner													
	Snack		1											
F14	Bkfst	1255.682	2											
	Lunch								3.936					
	Dinner		3						3.936					
	Snack		1	1										
F15	Bkfst	1255.682	3											
	Lunch		1											
	Dinner													
	Snack			1	1			1						
F16	Bkfst	1255.682	1											
	Lunch				1									
	Dinner							3						
	Snack													
F17	Bkfst	1255.682	1											
	Lunch				1									
	Dinner				1									
	Snack		2			1								
F21	Bkfst	1255.682	1											
	Lunch													
	Dinner													
	Snack			1										

F22	Bkfst	1255.682	1											
	Lunch													
	Dinner		1											
	Snack		4					2						
F23	Bkfst	1255.682	2											
	Lunch													
	Dinner		1											
	Snack		2					1						
F24	Bkfst	1255.682	2											
	Lunch													
	Dinner							2						
	Snack													
F25	Bkfst	1255.682	2											
	Lunch		2											
	Dinner		2					2						
	Snack													
F26	Bkfst	1255.682		3										
	Lunch		1									1		
	Dinner													
	Snack													
H10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack											1		
H11	Bkfst	1255.682												
	Lunch											2		
	Dinner													
	Snack								5			1		
H12	Bkfst	1255.682		1										1
	Lunch													

	Dinner								2					
	Snack							3	3					
H13	Bkfst	1255.682		2										100
	Lunch													
	Dinner													50
	Snack								1					
H14	Bkfst	1255.682							2					
	Lunch													80
	Dinner													
	Snack													40
H15	Bkfst	1255.682		1										1
	Lunch													40
	Dinner													
	Snack			0.5	2			0.5						
H16	Bkfst	1255.682												
	Lunch				0.5									
	Dinner			2										
	Snack													
H17	Bkfst	1255.682			1									100
	Lunch													
	Dinner													
	Snack													
H21	Bkfst	1255.682		1										1
	Lunch													
	Dinner													
	Snack			1										1
H22	Bkfst	1255.682		0.5										1
	Lunch											1.25		
	Dinner											5		
	Snack							1						

H23	Bkfst	1255.682		2										
	Lunch				1									
	Dinner							1				2.5		
	Snack													
H24	Bkfst	1255.682		1										100
	Lunch													
	Dinner							3				3.75		
	Snack													
H25	Bkfst	1255.682										1.25		
	Lunch													
	Dinner			2										
	Snack													
H26	Bkfst	1255.682										2.5		
	Lunch											2.5		
	Dinner													
	Snack													
I10	Bkfst	1255.682												
	Lunch													
	Dinner													
	Snack			1								1		
I11	Bkfst	1255.682												
	Lunch											1		
	Dinner			1										
	Snack		3									1		
I12	Bkfst	1255.682	1	1										
	Lunch													
	Dinner													
	Snack													
J13	Bkfst	1255.682												
	Lunch													

	Dinner													
	Snack													
J14	Bkfst	1255.682	2	1					2					2
	Lunch													
	Dinner													
	Snack													
J15	Bkfst	1255.682	2.5											2
	Lunch				2									40
	Dinner													
	Snack													
J16	Bkfst	1255.682	2											2
	Lunch													
	Dinner													
	Snack			0.5				0.5						
J17	Bkfst	1255.682	1											
	Lunch													
	Dinner													
	Snack		2											
J21	Bkfst	1255.682	3									1		2
	Lunch													
	Dinner													
	Snack			2										
J22	Bkfst	1255.682	2											
	Lunch													
	Dinner			1								1.25		
	Snack													

C.2 Protein

		Protein																										
		Hamburger - lean ground beef	Breakfast sausages	Bologna -sliced	Rabbit Stew	Packaged bologna	Moose meat stew	Red Horse Sucker	Boil White Fish head	Meat balls - lean ground beef	Garlic Sausage	Garlic Sausage slices	Pork Roast	Kielbasa Sausage Slices	Kielbasa Sausage chunk	Steak	Chicken Rotini soup/stew	Pork chops - no bone	Moose Ribs	Bacon	Hot Dogs	Harvest Thick Sliced Bacon	egg - whole	Smoked Ham	BBQ Chicken	Beef Roast	Ham	Beef Stew - Canned
		116.17 g	38.81 g	26.44 g	168 g	27.78 g	168 g	160.5 g	244 g	11.83 g	68 g	10.33 g	59 g	11 g	68 g	148 g	115 g	139.41 g	132 g	8 g	37.5 g	41.67 g	52 g	21 g	387 g	62.63 g	21 g	152 g
ID	Meal	Number of portions consumed																										
A10	Bkfst																											
	Lunch	2																										
	Dinner																											
	Snack																						3					
A11	Bkfst			1																								
	Lunch																	1										
	Dinner																											
	Snack																						3					
A12	Bkfst					1																						
	Lunch																	1										
	Dinner																											
	Snack																			5			2					
A13	Bkfst							1																				
	Lunch																											

	Dinner																											
	Snack		1																			2					
A14	Bkfst			2																								
	Lunch								1																			
	Dinner																											
	Snack																					2					
A15	Bkfst			1																								
	Lunch									6																		
	Dinner																											
	Snack																					2				3	
A16	Bkfst																											4
	Lunch												1															
	Dinner																											
	Snack													4														
A17	Bkfst																											
	Lunch															1												
	Dinner																											
	Snack																			7			2					
A21	Bkfst			2																								
	Lunch				1																							
	Dinner																											
	Snack																			2			2					
A22	Bkfst					1																						
	Lunch																									3		
	Dinner																											
	Snack																											
A23	Bkfst					1																						
	Lunch																											
	Dinner																											

	Snack																			7			2					
A24	Bkfst			1																								
	Lunch															1												
	Dinner																											
	Snack		1																				3					
A25	Bkfst					1																						
	Lunch										1																	
	Dinner																											
	Snack																			7			2					
A26	Bkfst	1																									2	
	Lunch															1												
	Dinner																											
	Snack																											
B10	Bkfst																											
	Lunch	1																										
	Dinner																											
	Snack		3																				2					
B11	Bkfst			2																								
	Lunch																	3										
	Dinner																											
	Snack																						2					
B12	Bkfst			1		1																						
	Lunch																	2										
	Dinner																											
	Snack																			7			2					
B13	Bkfst						1																					
	Lunch																											
	Dinner																				1							
	Snack		5																				2					

B14	Bkfst																											
	Lunch								1																			
	Dinner																											
	Snack																			6			3					
B15	Bkfst			1										3														
	Lunch									8																		
	Dinner																											
	Snack																						3				5	
B16	Bkfst											4																3
	Lunch												4															
	Dinner																											
	Snack		5									5																
B17	Bkfst																											
	Lunch															3												
	Dinner																											
	Snack																			6			3					
B21	Bkfst											2					3											
	Lunch				4																							
	Dinner																											
	Snack		3																	3			3					
B22	Bkfst																											
	Lunch																									5		
	Dinner																											
	Snack					2																	2				2	
B23	Bkfst																									1		
	Lunch																											
	Dinner																											
	Snack																			12			5					
B24	Bkfst																											

	Lunch															2												
	Dinner																											
	Snack		4																				3					
B25	Bkfst					4																						
	Lunch	3																										
	Dinner																											
	Snack																			7			2					
B26	Bkfst																										2	
	Lunch	2														2												
	Dinner																											
	Snack																											
C10	Bkfst																											
	Lunch	1																										
	Dinner																											
	Snack		4																				4					
C11	Bkfst			1																								
	Lunch																	3										
	Dinner																											
	Snack		1																				2					
C12	Bkfst																											
	Lunch																	1										
	Dinner																											
	Snack																			6			1					
C13	Bkfst						1																					
	Lunch						2																					
	Dinner																				1							
	Snack		2																				2					
C14	Bkfst																											
	Lunch							1	1																			

	Dinner																											
	Snack																			6			3					
C15	Bkfst														2.00													
	Lunch									6																		
	Dinner																											
	Snack																						2				2	
C16	Bkfst										1.5																	2
	Lunch												2															
	Dinner																											
	Snack																											
C17	Bkfst																											
	Lunch															1												
	Dinner															1												
	Snack																			6			2					
C21	Bkfst					1																						
	Lunch				2																							
	Dinner																											
	Snack		2																	4			2					
C22	Bkfst																											
	Lunch																									2		
	Dinner																											
	Snack					4																	2				2	
C23	Bkfst																											
	Lunch																									1		
	Dinner																											
	Snack																			6			4					
C24	Bkfst																											
	Lunch															1												
	Dinner																											

	Snack		2																				3					
C25	Bkfst																											1
	Lunch	1																										
	Dinner																											
	Snack		2																				4					
C26	Bkfst																											
	Lunch	2																										
	Dinner																											
	Snack																											
D10	Bkfst																											
	Lunch	2																										
	Dinner																											
	Snack		4																				1					
D11	Bkfst			1																								
	Lunch																2											
	Dinner																											
	Snack		2																									
D12	Bkfst					2																						
	Lunch																2											
	Dinner																											
	Snack																		6			1						
D13	Bkfst						0.25																					
	Lunch																											
	Dinner																			2								
	Snack		4																			1						
D14	Bkfst					2																						
	Lunch																											
	Dinner																											
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D15	Bkfst					1																						
	Lunch									10																		
	Dinner																											
	Snack																							1				1	
D16	Bkfst										2																	4
	Lunch												3															
	Dinner																											
	Snack		2								2			1														
D17	Bkfst																											
	Lunch															1												
	Dinner																											
	Snack																			10			1					
D21	Bkfst										2																	
	Lunch				2																							
	Dinner																											
	Snack		2																	3			1					
D22	Bkfst					2																						
	Lunch																									4		
	Dinner																											
	Snack					4																						
D23	Bkfst																									1		
	Lunch																									2		
	Dinner																											
	Snack																			7			1					
D24	Bkfst																											
	Lunch															1												
	Dinner																											
	Snack		4																									
D25	Bkfst					2																						

	Lunch	1																										
	Dinner																											
	Snack	1	1																	9								
D26	Bkfst																										1	
	Lunch															1												
	Dinner																											
	Snack																											
E10	Bkfst																											
	Lunch	3																										
	Dinner																											
	Snack		4																				4					
E11	Bkfst			3																								
	Lunch																	3										
	Dinner																											
	Snack		4																				4					
E12	Bkfst			1		1								#VALUE!														
	Lunch																	3										
	Dinner																											
	Snack																			6			2					
E13	Bkfst								1																			
	Lunch						2																					
	Dinner																				2							
	Snack		5																				3					
E14	Bkfst	2																										
	Lunch							1																				
	Dinner																											
	Snack																						3					
E15	Bkfst							2													4							
	Lunch									12																		
	Dinner																											
	Snack																						3				5	
E16	Bkfst										2																	3

	Lunch												4															
	Dinner																											
	Snack										2			10									3					
E17	Bkfst																								0.25			
	Lunch															2												
	Dinner																											
	Snack																			6			4					
E21	Bkfst										1																	3
	Lunch				2																							
	Dinner																											
	Snack		5																	3			3					
E22	Bkfst				3	5																						
	Lunch																									3		
	Dinner																											
	Snack					3																	3				1	
E23	Bkfst																									2		
	Lunch																		1									
	Dinner																											
	Snack																			6			3					
E24	Bkfst																											
	Lunch																											
	Dinner																											
	Snack		4																				4					
E25	Bkfst					3																						2
	Lunch											6																
	Dinner																											
	Snack	1	2																	5			4					
E26	Bkfst																											
	Lunch																											
	Dinner																											
	Snack																											
F10	Bkfst																											
	Lunch	1																										

	Dinner																											
	Snack																					2					
F11	Bkfst			1																								
	Lunch																2										
	Dinner																											
	Snack		2																			2					
F12	Bkfst					1																						
	Lunch																2										
	Dinner																											
	Snack																			9			2					
F13	Bkfst						1																					
	Lunch																											
	Dinner																											
	Snack		4																			2					
F14	Bkfst					4																						
	Lunch								2																			
	Dinner																											
	Snack																			6			3					
F15	Bkfst							2																				
	Lunch									7																		
	Dinner																											
	Snack																					3				3	
F16	Bkfst										1																	2
	Lunch												2															
	Dinner																											
	Snack		4									4																
F17	Bkfst																							0.25			
	Lunch															2												
	Dinner																											
	Snack																			6			3					
F21	Bkfst			2																								
	Lunch				4																							
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	Snack		4																	5			3					
F22	Bkfst																											
	Lunch																									2		
	Dinner																											
	Snack					2																	3					
F23	Bkfst																									2		
	Lunch																									2		
	Dinner																											
	Snack																			6			4					
F24	Bkfst																											
	Lunch															2												
	Dinner																											
	Snack		3																				3					
F25	Bkfst					1																						1
	Lunch	2																										
	Dinner																											
	Snack		4																	3			2					
F26	Bkfst	1																									1	
	Lunch																											
	Dinner																											
	Snack																											
H10	Bkfst																											
	Lunch	1																										
	Dinner																											
	Snack		5																				1					
H11	Bkfst			1																								
	Lunch																											
	Dinner																											
	Snack		3																				1					
H12	Bkfst														2													
	Lunch																	3										

	Dinner																											
	Snack																											
H13	Bkfst																											
	Lunch																											
	Dinner						1																					
	Snack		12																				1					
H14	Bkfst							0.5																				
	Lunch																											
	Dinner																											
	Snack																			11								
H15	Bkfst																											
	Lunch									20																		
	Dinner																											
	Snack																						1				3	
H16	Bkfst										0.5																	4
	Lunch												4															
	Dinner																											
	Snack		2									4											1					
H17	Bkfst																											
	Lunch															1												
	Dinner																											
	Snack																			10			1					
H21	Bkfst																											
	Lunch				1																							
	Dinner																											
	Snack		4.5																	4			1					
H22	Bkfst																											
	Lunch																									3		
	Dinne																											

	r																											
	Snack																					1						
H23	Bkfst																								1			
	Lunch																								2			
	Dinner																											
	Snack																											
H24	Bkfst																											
	Lunch															2												
	Dinner																											
	Snack		5																									
H25	Bkfst																											
	Lunch	3																										
	Dinner																											
	Snack		3																	5			1.5					
H26	Bkfst	1																										
	Lunch															1												
	Dinner																											
	Snack																											
I10	Bkfst																											
	Lunch	1																										
	Dinner																											
	Snack		3																				2					
I11	Bkfst			2																								
	Lunch																	2										
	Dinner																											
	Snack		2																				1					
I12	Bkfst																											
	Lunch																											
	Dinner																											
	Snack																											
J13	Bkfst																											

	Lunch							1													1							
	Dinner																											
	Snack		2																				1					
J14	Bkfst																											
	Lunch							0.25																				
	Dinner																											
	Snack																			4			2					
J15	Bkfst																											
	Lunch									6																		
	Dinner																											
	Snack																						1				3	
J16	Bkfst																											1
	Lunch												1															
	Dinner																											
	Snack		2								2																	
J17	Bkfst																								0.25			
	Lunch																											
	Dinner																											
	Snack																			4			2					
J21	Bkfst																											
	Lunch				1																							
	Dinner																											
	Snack																			3			1					
J22	Bkfst																											
	Lunch																									2		
	Dinner																											
	Snack																											

C.3 Starch/Carbohydrates

		Carbohydrates/Starch								
		Cherrioes	Corn Flakes	Bannock	bread	Hot Dog buns	Hamburger Buns	Whole Wheat Buns	Spaghetti	Potatoes
ID	Meal	32	28	99.83	32	32	45.67	60	125	91
		Number of portions consumed								
A10	Bkfst									
	Lunch									
	Dinner									
	Snack				1					1
A11	Bkfst				2					
	Lunch									2
	Dinner									
	Snack				1					
A12	Bkfst				2					
	Lunch									
	Dinner									
	Snack									1
A13	Bkfst									
	Lunch									
	Dinner	1								
	Snack				0.5					1
A14	Bkfst				4					
	Lunch									2
	Dinner									
	Snack									
A15	Bkfst				2					
	Lunch								1	
	Dinner	1								
	Snack				1					1
A16	Bkfst				1					
	Lunch									1
	Dinner									
	Snack									1

A17	Bkfst									
	Lunch									1
	Dinner									
	Snack				1					2
A21	Bkfst				2					
	Lunch			1						
	Dinner									
	Snack									
A22	Bkfst				2					
	Lunch									3
	Dinner									
	Snack									
A23	Bkfst				2					
	Lunch	1								
	Dinner									
	Snack									1
A24	Bkfst				2					
	Lunch									2
	Dinner									
	Snack									
A25	Bkfst				2					
	Lunch									
	Dinner									
	Snack									1
A26	Bkfst				2					
	Lunch									1
	Dinner									
	Snack									
B10	Bkfst									
	Lunch						1			2
	Dinner									
	Snack				1					1
B11	Bkfst				4					
	Lunch									2
	Dinner									
	Snack				2					1
B12	Bkfst				4					
	Lunch									1
	Dinner									

	Snack									1
B13	Bkfst				3					
	Lunch									
	Dinner					1				
	Snack				1					1
B14	Bkfst									
	Lunch									
	Dinner									
	Snack				1					1
B15	Bkfst				4					
	Lunch								3	
	Dinner									
	Snack				2					1
B16	Bkfst				2					
	Lunch									2
	Dinner									
	Snack									2
B17	Bkfst							2		
	Lunch									2
	Dinner									
	Snack				1					1
B21	Bkfst				2					
	Lunch			3						
	Dinner									
	Snack				1					
B22	Bkfst									
	Lunch				1					2
	Dinner									
	Snack									
B23	Bkfst				2					
	Lunch				2					
	Dinner									
	Snack									
B24	Bkfst									
	Lunch									2
	Dinner									
	Snack						1			
B25	Bkfst				4					
	Lunch						3			

	Dinner									
	Snack									1
B26	Bkfst				4					
	Lunch						2			1
	Dinner									
	Snack									
C10	Bkfst									
	Lunch						1			2
	Dinner									
	Snack									
C11	Bkfst				2					
	Lunch									3
	Dinner									
	Snack									
C12	Bkfst									
	Lunch				2					2
	Dinner									
	Snack									
C13	Bkfst									
	Lunch					1				
	Dinner				1					
	Snack				1					
C14	Bkfst									
	Lunch									
	Dinner									
	Snack				1					
C15	Bkfst				1					
	Lunch								1	
	Dinner									
	Snack				1					
C16	Bkfst						2			
	Lunch						2			
	Dinner									
	Snack									
C17	Bkfst							1		
	Lunch									2
	Dinner				1					
	Snack				1					2
C21	Bkfst				4					

	Lunch			4						
	Dinner									
	Snack				1					
C22	Bkfst									
	Lunch				2					2
	Dinner									
	Snack									
C23	Bkfst									
	Lunch				2					
	Dinner									
	Snack									2
C24	Bkfst									
	Lunch									2
	Dinner									
	Snack						1			
C25	Bkfst									
	Lunch						1			
	Dinner									
	Snack									2
C26	Bkfst									
	Lunch						2			
	Dinner									
	Snack									
D10	Bkfst									
	Lunch									1
	Dinner									
	Snack				2					1
D11	Bkfst				2					
	Lunch									2
	Dinner									
	Snack	1			2					
D12	Bkfst				4					
	Lunch									2
	Dinner									
	Snack				2					1
D13	Bkfst									
	Lunch	1								
	Dinner					2				
	Snack				1					2

D14	Bkfst				2					
	Lunch									3
	Dinner									
	Snack	2			2					
D15	Bkfst				2					
	Lunch								3	
	Dinner	1								
	Snack				1					
D16	Bkfst				3					
	Lunch						1			2
	Dinner									
	Snack									
D17	Bkfst							1		
	Lunch									2
	Dinner									
	Snack				1					1
D21	Bkfst				2					
	Lunch			2						
	Dinner									
	Snack				1					
D22	Bkfst				2					
	Lunch									2
	Dinner									
	Snack									
D23	Bkfst				2					
	Lunch	1			2					
	Dinner									
	Snack				1					1
D24	Bkfst									
	Lunch									1
	Dinner									
	Snack	1								
D25	Bkfst				2					
	Lunch						1			
	Dinner									
	Snack						1			1
D26	Bkfst				2					
	Lunch									1
	Dinner									

	Snack									
E10	Bkfst									
	Lunch						2			3
	Dinner									
	Snack				2					0.5
E11	Bkfst				6					
	Lunch									2
	Dinner									
	Snack				3					0.5
E12	Bkfst				4					
	Lunch									2
	Dinner									
	Snack				1					1
E13	Bkfst				1					
	Lunch				1					
	Dinner				1					
	Snack				3					2
E14	Bkfst						1			
	Lunch									
	Dinner									
	Snack				2					1
E15	Bkfst					2				
	Lunch								4	
	Dinner									
	Snack				2					0.5
E16	Bkfst				3					
	Lunch									2
	Dinner									
	Snack				2					0.75
E17	Bkfst									
	Lunch									2
	Dinner									
	Snack				2					2
E21	Bkfst				2					
	Lunch			2						
	Dinner									
	Snack				3					
E22	Bkfst				3					
	Lunch				1					

	Dinner									
	Snack				2					
E23	Bkfst				2					
	Lunch				1					
	Dinner									
	Snack				3					
E24	Bkfst									
	Lunch									
	Dinner									
	Snack						1.5			
E25	Bkfst				3					
	Lunch									
	Dinner									
	Snack						1			1
E26	Bkfst									
	Lunch									
	Dinner									
	Snack									
F10	Bkfst									
	Lunch						1			2
	Dinner									
	Snack				1					2
F11	Bkfst				2					
	Lunch									3
	Dinner									
	Snack									1
F12	Bkfst				2					
	Lunch									2
	Dinner									
	Snack									2
F13	Bkfst									
	Lunch				1					
	Dinner									
	Snack									1
F14	Bkfst				4					
	Lunch									2
	Dinner									
	Snack									1
F15	Bkfst									

	Lunch								2	
	Dinner									
	Snack				1					1
F16	Bkfst						1			
	Lunch									1
	Dinner									
	Snack									2
F17	Bkfst							1		
	Lunch									
	Dinner									
	Snack									2
F21	Bkfst				4					
	Lunch			3						
	Dinner									
	Snack									
F22	Bkfst									
	Lunch									1.5
	Dinner									
	Snack									
F23	Bkfst				2					
	Lunch				1					
	Dinner									
	Snack									2
F24	Bkfst									
	Lunch									2
	Dinner									
	Snack						1			
F25	Bkfst				2					
	Lunch						2			
	Dinner									
	Snack									1
F26	Bkfst				1		1			1
	Lunch									
	Dinner									
	Snack									
H10	Bkfst									
	Lunch						1			1
	Dinner									
	Snack				1					1

H11	Bkfst				2					
	Lunch									1
	Dinner									
	Snack				2					
H12	Bkfst				4					
	Lunch									1
	Dinner									
	Snack									
H13	Bkfst									
	Lunch									
	Dinner				1					
	Snack				1					1
H14	Bkfst									
	Lunch									
	Dinner		1							
	Snack				2					1
H15	Bkfst		1							
	Lunch								2	
	Dinner									
	Snack				1					
H16	Bkfst						2			
	Lunch						2			1
	Dinner									
	Snack									
H17	Bkfst									
	Lunch				2					
	Dinner									
	Snack				1					0.5
H21	Bkfst									
	Lunch			3						
	Dinner									
	Snack				1					
H22	Bkfst									
	Lunch									2
	Dinner									
	Snack				3					
H23	Bkfst				2					
	Lunch									
	Dinner									

	Snack									
H24	Bkfst									
	Lunch									4
	Dinner									
	Snack						1			
H25	Bkfst									
	Lunch									
	Dinner						3			
	Snack				2					1
H26	Bkfst									
	Lunch						1			
	Dinner									
	Snack									
I10	Bkfst									
	Lunch						1			1
	Dinner									
	Snack				1					1
I11	Bkfst				4					
	Lunch									2
	Dinner									
	Snack				1					
I12	Bkfst									
	Lunch									
	Dinner									
	Snack									
J13	Bkfst									
	Lunch				1					
	Dinner									
	Snack				1					2
J14	Bkfst									
	Lunch									1
	Dinner									
	Snack									2
J15	Bkfst	2								
	Lunch								1.5	
	Dinner									
	Snack									
J16	Bkfst						1			
	Lunch									2

	Dinner									
	Snack									2
J17	Bkfst									
	Lunch									1
	Dinner									
	Snack									1
J21	Bkfst									
	Lunch			3						
	Dinner									
	Snack									
J22	Bkfst									
	Lunch									1
	Dinner									
	Snack									

C.4 Fruits and vegetables

		Fruit and vegetables																												
		Tomato Slice	Onions	Pickles	Fruit Salad	Cherry Tomatoes	Cauliflower	Broccoli	Cherries	Blueberry	Blueberry	Pineapple	Parsnips	Button Mushrooms	Strawberries	Grapes	Raspberry	Watermelon	Blackberry	Beans	Banana	Carrots	Plums	Orange	Mandarin Orange	Salad	Lettuce	Apple	Jam	Pasta Sauce
		23.36 g	20 g	6.3 g	112.7 g	10.1 g	10.1 g	10.1 g	11 g	910 g	64 g	136.5 g	52 g	81 g	54 g	6 g	128 g	62.92 g	164 g	89 g	91.15 g	52 g	64.5 g	162 g	79.67 g	53 g	53 g	50 g	5 g	125 g
ID	Meal	Number of portions consumed																												
A10	Bkfst																													
	Lunch	5	0.5																	3										
	Dinner																													
	Snack																			1										
A11	Bkfst																										1			
	Lunch					3	2	1														0.78								
	Dinner																											1		
	Snack	1																		1										
A12	Bkfst																													
	Lunch																													
	Dinner					3	1				1											0.97			5					
	Snack																													
A13	Bkfst										2																			

	Lunch																													
	Dinner																													
	Snack																		1											
A14	Bkfst																							1		2				
	Lunch																			1										
	Dinner																													
	Snack																													
A15	Bkfst																						1							
	Lunch																													1
	Dinner																													
	Snack																													
A16	Bkfst																													
	Lunch											0.5									0.5				1					
	Dinner																													
	Snack																		1											
A17	Bkfst															1														
	Lunch												1	1		1														
	Dinner																													
	Snack																													
A21	Bkfst																						1			1				
	Lunch																													
	Dinner																													
	Snack																						1							
A22	Bkfst									2																1				
	Lunch																													
	Dinn															1														

	er																													
	Snack																													
A23	Bkfst																						1						
	Lunch																													
	Dinner																													
	Snack																													
A24	Bkfst																									1			
	Lunch																													
	Dinner																													
	Snack																													
A25	Bkfst										2																1			
	Lunch										1																			
	Dinner																													
	Snack																													
A26	Bkfst																								1				
	Lunch	1																											
	Dinner														4				4											
	Snack																													
B10	Bkfst																													
	Lunch		4																											
	Dinner																													
	Snack																			1										
B11	Bkfst	2																									2			
	Lunch			1	1																									
	Dinner																				1							1		
	Snack	2																		1										
B1	Bkfst										1												2							

2																														
	Lunch			1	1																									
	Dinner																													
	Snack																													
B13	Bkfst										1																			
	Lunch																													
	Dinner																													
	Snack																			1										
B14	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
B15	Bkfst																													
	Lunch																													3
	Dinner																													
	Snack																													
B16	Bkfst								5			0.5																		
	Lunch												0.5							1		0.50								
	Dinner																													
	Snack																			1										
B17	Bkfst														1															
	Lunch													2																
	Dinner														1	6														
	Snack																													
B21	Bkfst																				1			1						
	Lunch																													

	Dinner																													
	Snack																			1										
B22	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																		1											
B23	Bkfst																	1						2						
	Lunch																													
	Dinner																		13											
	Snack																							0.25						
B24	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
B25	Bkfst										1										1									
	Lunch		3																											
	Dinner																	2												
	Snack																													
B26	Bkfst																									1				
	Lunch	2	2																											
	Dinner																													
	Snack																													
C10	Bkfst																													
	Lunch		1	6																1										
	Dinner																				0.75									

	Snack																			2										
C11	Bkfst																				1						1			
	Lunch				1																									
	Dinner										0.2																			6
	Snack	0.5																		2										
C12	Bkfst																													
	Lunch																												2	
	Dinner																													
	Snack																													
C13	Bkfst																													
	Lunch																													
	Dinner																													
	Snack	1																		1										
C14	Bkfst																													
	Lunch																													
	Dinner																													
	Snack			1																										
C15	Bkfst																							1						
	Lunch																													1
	Dinner																													
	Snack																													
C16	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
C1	Bkfst																1													

7																														
	Lunch														2	8														
	Dinner																													
	Snack																													
C21	Bkfst																							1						
	Lunch																													
	Dinner																													
	Snack																			2										
C22	Bkfst																				1									
	Lunch																													
	Dinner																													
	Snack																													
C23	Bkfst									0.25						12		3	0.3		1			1						
	Lunch																													
	Dinner																													
	Snack			4																										
C24	Bkfst									0.25																				
	Lunch																	0.5												
	Dinner																													
	Snack			3																										
C25	Bkfst																	1												
	Lunch		1																											
	Dinner																													
	Snack			2																										
C26	Bkfst																													
	Lunch	2	2																											

	Dinner																													
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D10	Bkfst																													
	Lunch		2																1											
	Dinner																													
	Snack																		1											
D11	Bkfst																									1				
	Lunch				2			2													0.58									
	Dinner			1																1							1			
	Snack																		0.5									2		
D12	Bkfst																				0.78	2					1			
	Lunch				1																									
	Dinner																													
	Snack																		1									2		
D13	Bkfst																			1										
	Lunch																			1										
	Dinner																													
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D14	Bkfst																					1								
	Lunch																													
	Dinner																			2										
	Snack																			1								2		
D15	Bkfst																													
	Lunch																													3
	Dinner																													

	Snack																													
D16	Bkfst																													
	Lunch																								1				
	Dinner																													
	Snack																		1										
D17	Bkfst																1												
	Lunch													1	7	12													
	Dinner																													
	Snack																													
D21	Bkfst																			1									
	Lunch																													
	Dinner																													
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D22	Bkfst										1									1									
	Lunch																													
	Dinner																													
	Snack																		1										
D23	Bkfst														6			2											
	Lunch																								2				
	Dinner																													
	Snack																			1									
D24	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
D25	Bkfst									0.25											1									

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	Snack																													
D26	Bkfst																				1					1				
	Lunch																													
	Dinner																													
	Snack																													
E10	Bkfst																													
	Lunch	2		4																3										
	Dinner																				1									
	Snack																			0.5										
E11	Bkfst										1																3	1		
	Lunch				3				12									1												
	Dinner																													
	Snack																			1										
E12	Bkfst																						1		2					
	Lunch				0.5																									
	Dinner																													
	Snack																			2										
E13	Bkfst								15												1				1					
	Lunch																													
	Dinner																													
	Snack																			1										
E14	Bkfst	1																		1.5	1			2						
	Lunch																													

	Dinner																													
	Snack																			1.5										
E15	Bkfst			2																1	1									
	Lunch																													4
	Dinner																													
	Snack																			1										
E16	Bkfst								12			0.5												1						
	Lunch												0.5							1		0.50								
	Dinner																													
	Snack												0.5							1		0.50								
E17	Bkfst								12			0.5												1						
	Lunch													1	5															
	Dinner																													
	Snack																													
E21	Bkfst									1																				
	Lunch																												2	
	Dinner																													
	Snack																			2										
E22	Bkfst															7														
	Lunch																													
	Dinner																													
	Snack																			1										
E23	Bkfst									0.5								1												
	Lunch								12																	2				
	Dinner																													

	Snack																							1						
E24	Bkfst														12			4			2									
	Lunch																													
	Dinner																													
	Snack																													
E25	Bkfst																				1									
	Lunch									2						12														
	Dinner																													
	Snack																													
E26	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
F10	Bkfst																													
	Lunch		7																		4										
	Dinner																													
	Snack																				1										
F11	Bkfst	1																				1						1			
	Lunch				1															1	1									
	Dinner																													
	Snack	2																												
F12	Bkfst										0.5												2							
	Lunch				1																							1		
	Dinner																													
	Snack																			1										
F13	Bkfst										1																			

	Lunch																													
	Dinner																													
	Snack																		1											
F14	Bkfst															6					1									
	Lunch																													
	Dinner																													
	Snack																													
F15	Bkfst																				2									
	Lunch																													2
	Dinner																													
	Snack																													
F16	Bkfst																													
	Lunch											0.5										0.50				1				
	Dinner								10																					
	Snack																		1											
F17	Bkfst								12						1															
	Lunch												1	0.5	12															
	Dinner																													
	Snack																													
F21	Bkfst																													
	Lunch																												1	
	Dinner																													
	Snack																		2											
F22	Bkfst																													
	Lunch																				1									
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	er																													
	Snack																		2											
F23	Bkfst																1													
	Lunch																													
	Dinner								8							10														
	Snack																													
F24	Bkfst								12																					
	Lunch																													
	Dinner																													
	Snack																													
F25	Bkfst								12																					
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	Dinner																													
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F26	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
H10	Bkfst																													
	Lunch	1	1																1											
	Dinner																													
	Snack																		0.5											
H11	Bkfst	1																									1			
	Lunch							1													0.58									
	Dinner																													
	Snack	1																										1		

H12	Bkfst	2																			0.78			1					
	Lunch																												
	Dinner																												
	Snack																												
H13	Bkfst																												
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	Dinner																												
	Snack																												
H14	Bkfst																												
	Lunch																												
	Dinner																												
	Snack																											2	
H15	Bkfst																												
	Lunch																												2
	Dinner																												
	Snack																												
H16	Bkfst																												
	Lunch																								1				
	Dinner																												
	Snack																												
H17	Bkfst																												
	Lunch																												
	Dinner																												
	Snack																												
H21	Bkfst																												
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	h																													
	Dinner																													
	Snack																													
H22	Bkfst															10														
	Lunch																													
	Dinner																													
	Snack																												2	
H23	Bkfst															12														
	Lunch																									2				
	Dinner																													
	Snack																													
H24	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
H25	Bkfst																													
	Lunch																													
	Dinner		2																											
	Snack																									1				
H26	Bkfst	1	1																											
	Lunch																													
	Dinner																													
	Snack																													
I10	Bkfst																													
	Lunch		1	7																										
	Dinner																				1									
	Snack																			1										

I11	Bkfst			1																							2			
	Lunch			1																										
	Dinner																													
	Snack																				1					1					
I12	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
J13	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																				2										
J14	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													
J15	Bkfst																													
	Lunch																													1.5
	Dinner																													
	Snack											1						2												
J16	Bkfst																													
	Lunch												0.25									0.25								
	Dinner																													
	Snack																			1										
J17	Bkfst											2												1						
	Lunch													1																
	Dinner																													
	Snack																													
J21	Bkfst																													

	Lunch																												1	
	Dinner																													
	Snack																		1											
J22	Bkfst																													
	Lunch																													
	Dinner																													
	Snack																													

C.5 Condiments and other

		Condiments and other														
		Jalapeno Havarti cheese slices	Kraft singles - Swiss	Salted butter	Margarine	Mustard	Mountain steak sauce	HP sauce	Ketchup	Sundried Tomato Dressing	Sour Cream	Honey - Moose mountain	Ranch Dressing	BBQ Sauce	Dad's cookies	Blueberry rhubarb dessert
		15.3	20.83	5	5	5	5	5	5	5	5	5	5	5	10	200
ID	Meal	Food portions consumed														
A10	Bkfst															
	Lunch															
	Dinner															
	Snack			1												
A11	Bkfst		1		1										2	
	Lunch															
	Dinner														2	1.5
	Snack				1											
A12	Bkfst	1				1										
	Lunch				2											
	Dinner															
	Snack								1							
A13	Bkfst														3	
	Lunch															
	Dinner											2				
	Snack			1								1				
A14	Bkfst	2				2									3	
	Lunch															1
	Dinner															
	Snack															
A15	Bkfst					1										
	Lunch															
	Dinner															
	Snack				1											
A16	Bkfst				1											
	Lunch				1								1			
	Dinner															
	Snack								2							

A17	Bkfst															
	Lunch						1	1								
	Dinner															
	Snack			1					2							
A21	Bkfst		1			1									5	
	Lunch				1											
	Dinner															
	Snack															
A22	Bkfst		1			1									3	
	Lunch															
	Dinner															
	Snack															
A23	Bkfst		1			1									15	
	Lunch															
	Dinner															
	Snack															
A24	Bkfst		1			1										
	Lunch			4				1								
	Dinner															
	Snack															
A25	Bkfst		1			1									13	
	Lunch															
	Dinner															
	Snack								1							
A26	Bkfst		1			1				1						
	Lunch			2											3	
	Dinner															
	Snack															
B10	Bkfst															
	Lunch										2					
	Dinner															
	Snack				1				1							
B11	Bkfst		2		2	2									3	
	Lunch															
	Dinner															
	Snack				1											
B12	Bkfst				2										3	
	Lunch															
	Dinner															

	Snack								1							
B13	Bkfst				1											
	Lunch					1										
	Dinner															
	Snack															
B14	Bkfst															
	Lunch															
	Dinner															1
	Snack				1											
B15	Bkfst				1	2										
	Lunch															
	Dinner											1				
	Snack				1				1							
B16	Bkfst				1	1										
	Lunch															
	Dinner															
	Snack															
B17	Bkfst				2											
	Lunch			1				3								
	Dinner															
	Snack				1											
B21	Bkfst				1	1										
	Lunch				3											
	Dinner															
	Snack				1											
B22	Bkfst															
	Lunch					1										
	Dinner															
	Snack															
B23	Bkfst				2										5	
	Lunch														2	
	Dinner															
	Snack															
B24	Bkfst															
	Lunch			1				2								
	Dinner															
	Snack															
B25	Bkfst		2			2									6	
	Lunch		3		3											

	Dinner															
	Snack															
B26	Bkfst		2	2									1		5	
	Lunch		2													
	Dinner															
	Snack															
C10	Bkfst															
	Lunch							1			2					
	Dinner															
	Snack															
C11	Bkfst		1		1											
	Lunch														6	
	Dinner															
	Snack															
C12	Bkfst															
	Lunch				1											
	Dinner															
	Snack															
C13	Bkfst															
	Lunch															
	Dinner					1										
	Snack			1												
C14	Bkfst															
	Lunch														5	
	Dinner															1
	Snack				1											
C15	Bkfst															
	Lunch															
	Dinner														4	
	Snack				1											
C16	Bkfst				1											
	Lunch				1											
	Dinner															
	Snack															
C17	Bkfst				1											
	Lunch															
	Dinner				1											
	Snack															
C21	Bkfst															

	Lunch															
	Dinner															
	Snack				1											
C22	Bkfst														6	
	Lunch			1												
	Dinner															
	Snack															
C23	Bkfst															
	Lunch				1										10	
	Dinner															
	Snack															
C24	Bkfst															
	Lunch			3				3							1	
	Dinner															
	Snack			1												
C25	Bkfst														10	
	Lunch		1	1												
	Dinner															
	Snack															
C26	Bkfst															
	Lunch			2												
	Dinner															
	Snack															
D10	Bkfst															
	Lunch	2									1			2		
	Dinner														2	
	Snack				1											
D11	Bkfst		1		1	1									1	
	Lunch												1			
	Dinner														3	
	Snack				1				2							
D12	Bkfst	2				2									3	
	Lunch			1										1		
	Dinner														3	
	Snack				1				2							
D13	Bkfst													2	4	
	Lunch															
	Dinner													1		
	Snack				1				2							

D14	Bkfst	1				1									3	
	Lunch													3	3	
	Dinner															1
	Snack				1											
D15	Bkfst					1										
	Lunch															
	Dinner															
	Snack								2							
D16	Bkfst		1		1	2										
	Lunch									1				1		
	Dinner															
	Snack															
D17	Bkfst				1											
	Lunch													1		
	Dinner															
	Snack															
D21	Bkfst													1		
	Lunch				2											
	Dinner															
	Snack															
D22	Bkfst					1									5	
	Lunch															
	Dinner															
	Snack															
D23	Bkfst													1	4	
	Lunch									2						
	Dinner															
	Snack															
D24	Bkfst														5	
	Lunch			1										4		
	Dinner															
	Snack								1							
D25	Bkfst		1			1									5	
	Lunch		1			1			1							
	Dinner															
	Snack		1			1			1							
D26	Bkfst		1			1				1					1	
	Lunch			1										1		
	Dinner															

	Snack															
E10	Bkfst															
	Lunch					2			2							
	Dinner															
	Snack				1				1							
E11	Bkfst					3									3	
	Lunch															
	Dinner															
	Snack				2				1							
E12	Bkfst				4	4									4	
	Lunch				2									3		
	Dinner															
	Snack								1							
E13	Bkfst															
	Lunch															
	Dinner															
	Snack			1	2											
E14	Bkfst														4	
	Lunch															1
	Dinner															
	Snack				2											1
E15	Bkfst								2							
	Lunch															
	Dinner															
	Snack				2											
E16	Bkfst															
	Lunch								1							
	Dinner															
	Snack				2											
E17	Bkfst															
	Lunch				1									2		
	Dinner															
	Snack															
E21	Bkfst			2												
	Lunch															
	Dinner															
	Snack				2											
E22	Bkfst				3											
	Lunch			1												

	Dinner															
	Snack				2											
E23	Bkfst				2										5	
	Lunch				1					2					6	
	Dinner															
	Snack				3											
E24	Bkfst														12	
	Lunch															
	Dinner															
	Snack			1												
E25	Bkfst				3										3	
	Lunch															
	Dinner															
	Snack								2							
E26	Bkfst															
	Lunch															
	Dinner															
	Snack															
F10	Bkfst															
	Lunch															
	Dinner															
	Snack				1				1							
F11	Bkfst				1	1									3	
	Lunch															
	Dinner															
	Snack															
F12	Bkfst					1									3	
	Lunch															
	Dinner															
	Snack															
F13	Bkfst															
	Lunch				1										4	
	Dinner															
	Snack															
F14	Bkfst															
	Lunch														5	
	Dinner															
	Snack															
F15	Bkfst															

	Lunch															
	Dinner															
	Snack				1											
F16	Bkfst				1											
	Lunch												1			
	Dinner															
	Snack															
F17	Bkfst				1											
	Lunch				1			2								
	Dinner															
	Snack															
F21	Bkfst															
	Lunch				3											
	Dinner															
	Snack															
F22	Bkfst														5	
	Lunch															
	Dinner															
	Snack															
F23	Bkfst				1										12	
	Lunch				1											
	Dinner														5	
	Snack															
F24	Bkfst															
	Lunch			1												
	Dinner															
	Snack			1												
F25	Bkfst				1										8	
	Lunch															
	Dinner															
	Snack															
F26	Bkfst															
	Lunch														5	
	Dinner															
	Snack															
H10	Bkfst															
	Lunch	1												1		
	Dinner														1	
	Snack				1				3							

H11	Bkfst		1												3	
	Lunch												3			1
	Dinner														7	
	Snack				1											
H12	Bkfst	2			1										5	
	Lunch			1										3		
	Dinner															
	Snack															
H13	Bkfst															
	Lunch															
	Dinner															
	Snack			1					3							
H14	Bkfst															
	Lunch															
	Dinner														17	1
	Snack				1				1			1				1
H15	Bkfst											1				
	Lunch															
	Dinner											1				
	Snack				1				3							
H16	Bkfst	2			1											
	Lunch				1				1			1	1	1		
	Dinner															
	Snack								2							
H17	Bkfst														18	
	Lunch													2		
	Dinner															
	Snack				1							1				
H21	Bkfst														1	
	Lunch				2											
	Dinner															
	Snack				1							0.5				
H22	Bkfst														3	
	Lunch															
	Dinner															
	Snack				1				1							
H23	Bkfst		1											1	8	
	Lunch												2		3	
	Dinner															

	Snack															
H24	Bkfst														10	
	Lunch			2					8							
	Dinner															
	Snack								1			2				
H25	Bkfst														13	
	Lunch		3				1					2		1		
	Dinner				2											
	Snack								1				1		4	
H26	Bkfst													1		
	Lunch		1													
	Dinner															
	Snack															
I10	Bkfst															
	Lunch	1							1							
	Dinner															
	Snack				1											
I11	Bkfst					2									3	
	Lunch															
	Dinner															
	Snack				1											
I12	Bkfst															
	Lunch															
	Dinner															
	Snack															
J13	Bkfst															
	Lunch								1							
	Dinner															
	Snack															
J14	Bkfst															
	Lunch															
	Dinner															
	Snack															
J15	Bkfst											2				
	Lunch															
	Dinner															
	Snack															
J16	Bkfst				1											
	Lunch							1								

	Dinner															
	Snack															
J17	Bkfst															
	Lunch							1								
	Dinner															
	Snack															
J21	Bkfst															
	Lunch				1											
	Dinner															
	Snack															
J22	Bkfst															
	Lunch															
	Dinner															
	Snack															

Appendix D - Calculated daily tracer intake for each subject during the Cold Lake soil ingestion study

		elemental tracers (µg)										
Participant	Day	Al	Ba	Ce	La	Mn	Si	Ti	V	U	Y	Zr
A10	1	720.24	173.16	0.64	0.34	1091.31	35,223	48.15	8.72	0.15	0.03	6.98
A11	2	5,067.84	735.20	6.35	1.83	8734.21	32,143	397.34	24.68	1.40	1.00	18.37
A12	3	2,437.95	620.22	2.45	1.52	1905.86	19,028	278.40	11.99	0.69	0.91	20.86
A13	4	1,820.71	545.42	2.91	1.62	3596.05	18,938	273.71	13.91	0.73	1.11	9.99
A14	5	3,578.96	866.16	6.13	1.98	7053.93	38,704	376.82	27.19	1.80	1.34	17.65
A15	6	2,581.70	605.12	2.21	1.29	3317.13	31,024	179.90	10.94	0.52	2.13	10.34
A16	7	4,099.80	455.20	2.68	1.40	1859.54	20,471	278.55	17.68	1.21	0.82	18.88
A17	8	2,180.76	250.96	1.90	1.01	1737.17	23,057	178.93	14.39	0.94	0.50	12.34
A21	9	2,588.93	1165.09	2.70	1.39	2382.00	25,536	241.16	15.17	1.05	0.82	11.97
A22	10	2,529.68	617.31	3.73	1.75	2649.96	20,665	275.43	18.24	1.34	1.79	19.49
A23	11	1,208.00	567.91	2.27	1.41	3430.69	27,642	262.05	20.60	0.77	1.53	9.13
A24	12	2,512.55	452.93	4.12	1.89	1672.21	18,414	265.66	24.16	1.38	1.99	16.73
A25	13	1,279.72	411.76	2.77	1.28	2932.00	18,562	190.11	11.06	0.41	0.97	16.29
A26	14	4,671.73	1640.71	4.19	2.25	5901.59	44,698	392.71	21.23	1.37	1.49	37.00
B10	1	3,221.42	199.97	1.17	0.61	2110.04	19,385	75.10	7.60	0.77	0.52	3.62
B11	2	3,759.31	788.75	5.22	2.47	3969.04	35,240	328.85	26.68	1.75	2.32	35.57
B12	3	2,653.16	562.29	3.22	1.71	3723.04	33,055	237.36	16.91	1.35	1.05	18.49
B13	4	2,010.53	1034.37	2.86	1.56	1964.74	24,348	228.81	13.08	0.82	0.87	8.31
B14	5	2,630.92	680.77	4.78	1.58	5447.15	37,654	252.57	18.77	0.86	0.90	11.69
B15	6	5,146.06	1293.59	5.62	2.98	5810.05	50,339	316.58	31.14	1.20	4.13	17.69
B16	7	6,182.77	912.63	6.03	3.16	8273.58	30,179	482.05	40.97	2.13	4.37	29.19
B17	8	3,562.56	524.12	4.09	2.09	2797.02	38,739	332.09	26.33	2.01	1.48	26.30
B21	9	4,539.12	3321.62	4.85	2.48	3696.07	52,764	414.12	23.42	0.99	1.23	23.54
B22	10	4,221.23	590.09	4.29	2.04	2484.04	22,015	236.34	25.57	1.22	4.11	19.68
B23	11	6,855.22	4644.65	7.66	3.74	14403.33	43,495	539.82	29.37	1.30	4.45	43.57
B24	12	2,730.85	376.48	3.31	1.54	992.15	18,796	241.94	19.53	0.98	2.09	16.58
B25	13	3,119.29	797.34	6.98	3.21	3567.85	63,540	310.68	27.74	1.08	4.33	34.19
B26	14	4,146.90	605.16	5.65	2.54	2535.66	48,328	378.88	23.59	1.41	2.48	28.57

C10	1	1,285.97	238.91	1.81	0.82	1187.11	28,516	122.01	11.72	0.80	0.74	7.37
C11	2	8,866.18	1254.02	8.95	4.46	6130.34	59,435	620.35	39.97	1.92	3.89	42.33
C12	3	1,871.27	414.76	2.37	1.22	1914.38	19,279	220.97	22.38	1.17	0.83	10.97
C13	4	2,299.18	2155.08	2.55	1.38	1376.54	35,041	234.86	12.69	0.41	0.63	7.74
C14	5	3,092.08	927.84	5.59	1.97	6769.30	36,141	287.34	17.41	0.55	0.94	13.11
C15	6	2,728.62	664.57	2.75	1.44	2813.59	34,328	263.96	18.07	0.59	1.29	16.38
C16	7	2,883.82	449.73	3.51	1.71	1680.78	20,654	229.59	18.80	0.90	2.82	19.28
C17	8	2,382.50	295.73	2.54	1.36	1900.46	20,446	168.81	14.41	1.02	1.42	9.62
C21	9	3,578.49	1974.80	3.88	1.90	3379.70	30,910	299.68	15.18	1.16	1.17	13.50
C22	10	4,149.78	603.31	5.01	2.14	3432.74	23,526	225.21	22.41	1.07	3.46	22.25
C23	11	3,477.33	737.35	5.52	2.42	4275.52	19,948	362.98	20.16	0.75	2.86	29.36
C24	12	2,587.72	417.85	3.35	1.71	2534.63	22,837	274.42	19.90	1.52	1.22	14.66
C25	13	2,463.80	449.17	2.64	1.39	2388.58	33,978	166.11	14.18	0.52	1.43	13.45
C26	14	2,819.82	372.40	2.63	1.28	1848.73	41,373	201.80	14.75	1.02	0.96	11.66
D10	1	1,810.75	216.08	1.99	0.82	1201.44	33,056	70.17	9.49	0.54	1.03	9.74
D11	2	3,289.12	639.99	4.59	2.19	4409.20	35,233	291.54	22.87	1.56	2.11	28.32
D12	3	3,409.95	641.25	3.70	2.25	4329.22	41,939	272.50	21.48	1.68	2.98	21.30
D13	4	2,135.21	599.47	2.89	1.58	3104.30	28,802	233.19	11.24	0.79	1.78	10.48
D14	5	3,900.72	705.11	7.12	2.23	6877.91	35,149	391.95	29.82	1.96	2.21	20.75
D15	6	4,127.89	1164.30	3.77	2.39	7997.44	65,966	343.51	16.38	0.91	3.54	12.37
D16	7	3,250.02	568.39	3.62	1.76	1991.34	28,045	216.93	24.85	1.40	2.48	42.70
D17	8	3,803.09	493.43	4.29	2.27	3336.32	42,164	249.78	21.73	1.29	2.17	25.99
D21	9	2,923.34	1633.44	2.90	1.44	2249.74	31,850	234.31	10.82	0.65	0.73	13.87
D22	10	3,050.67	556.01	4.41	1.95	2726.85	20,045	222.24	20.12	1.12	3.37	21.03
D23	11	2,776.18	624.52	3.60	2.04	4076.39	38,755	223.20	13.24	0.82	2.77	49.63
D24	12	2,096.93	320.08	2.77	1.41	1403.42	16,120	157.46	13.94	0.84	1.87	10.68
D25	13	1,132.76	379.04	2.48	1.44	3913.99	35,404	209.46	8.92	0.57	1.34	15.43
D26	14	2,693.78	351.17	3.64	1.62	1565.16	28,739	249.34	15.72	1.03	1.66	33.70
E10	1	2,081.15	388.79	2.59	1.22	2232.51	56,548	163.21	18.32	1.21	0.93	11.80
E11	2	6,987.15	871.61	5.48	2.63	8342.89	46,200	410.73	23.18	2.00	2.28	39.98
E12	3	5,757.47	801.93	4.71	2.45	4533.25	44,115	378.63	27.21	2.02	2.13	38.51
E13	4	7,648.15	3470.94	8.69	4.40	5827.62	75,329	530.15	20.95	1.24	2.28	26.83

E14	5	9,024.37	2871.32	12.82	5.38	11477.78	92,876	721.38	33.12	1.77	3.43	41.50
E15	6	10,751.15	2110.26	9.57	3.86	15407.30	73,480	638.38	37.26	1.14	2.29	25.67
E16	7	8,624.82	1010.50	5.73	3.03	10025.53	36,804	552.65	34.91	1.95	3.11	34.95
E17	8	7,134.92	919.51	4.89	2.94	10387.92	46,633	458.89	32.13	1.77	2.74	33.85
E21	9	8,403.11	2175.86	7.62	3.72	10246.84	39,089	482.13	20.95	1.42	2.28	46.13
E22	10	6,281.02	2527.62	5.45	2.55	4147.93	53,506	337.80	17.60	0.79	2.33	25.09
E23	11	5,153.18	728.49	5.68	2.91	6299.13	29,154	449.06	13.43	0.80	1.70	58.97
E24	12	3,686.70	683.76	3.32	2.14	4862.36	42,717	304.11	11.18	0.73	1.45	17.27
E25	13	4,052.98	1150.98	12.74	5.75	12759.35	26,548	665.10	25.92	1.20	5.03	78.50
E26	14	1,386.80	183.13	1.14	0.58	633.85	23,538	114.33	5.42	0.50	0.27	5.48
F10	1	2,968.45	423.92	2.15	0.98	2790.22	27,996	99.70	15.07	0.85	0.88	8.84
F11	2	3,977.80	609.10	3.64	1.72	4510.06	28,632	294.48	26.78	2.05	1.25	20.54
F12	3	4,720.83	378.06	2.25	1.13	3931.22	26,317	193.59	14.32	1.33	0.82	18.06
F13	4	2,004.17	952.92	2.30	1.16	2022.03	23,090	210.31	15.00	0.91	0.48	8.01
F14	5	4,787.85	1485.63	6.33	3.95	4763.82	67,690	304.01	34.17	2.21	2.69	18.50
F15	6	4,844.83	1237.36	4.93	2.51	6010.28	33,411	401.35	24.60	0.72	1.90	14.80
F16	7	3,863.60	475.26	3.51	1.83	1623.00	21,237	330.44	23.32	1.32	2.47	19.91
F17	8	2,524.00	385.24	2.74	1.49	2003.27	29,585	263.47	22.05	1.21	2.26	17.28
F21	9	5,052.02	3079.80	4.27	2.13	3546.24	45,800	311.47	15.85	1.07	1.22	13.41
F22	10	1,978.89	461.64	2.21	1.13	2044.41	24,107	221.45	20.65	0.88	1.16	10.53
F23	11	1,868.34	587.17	2.21	1.14	3167.76	28,972	232.66	15.89	0.57	0.90	10.21
F24	12	2,568.29	329.76	2.43	1.23	1311.29	16,007	248.12	20.29	1.45	1.28	10.54
F25	13	1,954.49	501.19	2.72	1.45	2522.53	46,384	252.35	16.39	0.78	1.52	12.67
F26	14	4,475.80	314.48	2.56	1.20	2794.06	28,666	191.44	11.30	0.96	1.17	10.77
H10	1	869.85	185.92	1.41	0.61	876.50	21,019	63.60	7.32	0.51	0.63	6.32
H11	2	4,030.52	1156.02	8.21	3.40	7107.14	45,492	332.06	26.61	1.22	3.24	24.70
H12	3	5,225.07	1058.48	5.53	3.46	3499.97	45,043	296.74	25.21	1.58	3.49	35.04
H13	4	2,412.48	841.23	1.42	0.98	1460.95	14,826	51.05	2.76	0.13	0.88	3.10
H14	5	3,195.72	858.91	5.43	2.05	7014.67	40,736	328.86	19.17	0.75	1.42	14.86
H15	6	4,915.87	883.81	5.70	1.93	7888.57	55,229	334.58	22.22	0.75	2.16	14.82
H16	7	5,198.64	491.66	3.16	1.49	2691.54	24,722	270.62	14.27	1.24	1.30	22.05
H17	8	1,417.80	322.14	1.58	0.92	2035.78	21,182	115.06	8.19	0.31	0.94	7.94

H21	9	3,480.70	949.75	1.96	0.96	2576.18	15,448	125.13	4.73	0.32	0.61	6.79
H22	10	2,648.03	468.18	5.36	2.07	1396.52	18,594	206.10	17.19	0.87	3.38	26.99
H23	11	3,485.31	526.17	3.67	1.61	3145.07	21,823	169.54	12.01	0.53	2.55	17.20
H24	12	3,510.37	480.85	4.91	2.21	2407.25	19,196	192.88	26.05	1.44	3.49	21.45
H25	13	3,297.11	409.51	2.72	1.40	3142.89	58,160	152.77	10.90	0.55	1.30	14.13
H26	14	2,293.08	413.94	4.63	1.86	1503.95	29,537	201.25	13.27	0.66	2.49	24.49
I10	1	1,724.53	164.46	1.43	0.64	1175.76	25,371	90.39	7.11	0.53	0.71	6.99
I11	2	3,736.44	557.95	3.95	1.80	3005.61	24,680	265.11	20.17	1.58	1.65	23.25
I12	3	1,419.77	231.57	0.76	0.40	1114.72	10,131	78.92	4.06	0.19	0.35	6.00
J13	4	853.09	265.72	1.23	0.61	1222.85	3,604	82.09	2.41	0.08	0.25	2.87
J14	5	2,685.33	553.54	2.60	1.47	2514.62	21,058	142.57	17.11	1.19	0.99	6.40
J15	6	2,698.80	719.15	2.83	1.69	4460.03	43,211	258.72	19.34	1.17	2.63	9.44
J16	7	3,848.45	652.91	3.05	1.81	12821.25	13,241	263.78	17.21	1.14	1.45	12.25
J17	8	3,143.09	1146.26	3.55	2.27	24392.66	20,547	243.24	26.19	1.45	2.42	21.86
J21	9	3,594.75	1009.90	2.62	1.19	2609.82	15,825	155.95	9.98	0.49	0.96	9.43
J22	10	1,764.68	218.64	1.58	0.66	1098.26	10,237	97.06	7.71	0.42	0.71	7.87

Appendix E - Calculated daily tracer concentrations for each subject's fecal samples during the Cold Lake soil ingestion study

		elemental tracers (µg/g)										
Participant	Day	Al	Ba	Ce	La	Mn	Si	Ti	V	U	Y	Zr
A10	1											
A11	2	1100	150	1.70	0.86	830.00	32000	6300.00	3.00	0.49	0.58	10
A12	3	980	210	1.60	0.76	870.00	35800	7100.00	3.00	0.45	0.53	10
A13	4	1000	260	1.60	0.69	1200.00	27600	6000.00	2.00	0.40	0.50	10
A14	5	1100	270	1.70	0.70	1800.00		4400.00	2.00	0.42	0.42	14
A15	6	1700	250	1.10	0.49	2200.00	13300	680.00	1.00	0.19	0.34	3
A16	7	1100	230	1.10	0.45	1900.00	15100	300.00	1.00	0.25	0.37	3
A17	8	820	210	1.40	0.42	1400.00	14100	440.00	1.00	0.32	0.37	3
A21	9	950	220	1.50	0.51	1200.00		1600.00	2.00	0.32	0.38	4
A22	10	1000	240	1.20	0.43	1200.00	14600	2400.00	2.00	0.36	0.41	6
A23	11	780	250	0.76	0.31	1100.00	16100	1100.00	2.00	0.25	0.25	5
A24	12	730	300	0.60	0.33	1200.00	19600	270.00	2.00	0.29	0.28	4
A25	13	490	270	0.54	0.39	1300.00	12800	160.00	2.00	0.26	0.46	2
A26	14	530	230	0.62	0.40	1200.00	11700	100.00	2.00	0.31	0.28	3
B10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
B11	2	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
B12	3	1500	480	1.30	0.64	1900.00	26400	1300.00	3.00	0.50	0.50	6
B13	4	830	250	1.80	0.90	1900.00	10500	690.00	3.00	0.28	0.40	10
B14	5	1500	240	1.00	0.55	2100.00	15400	240.00	3.00	0.28	0.31	14
B15	6	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
B16	7	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
B17	8	890	240	1.50	0.52	1200.00	13500	52.00	3.00	0.39	0.42	6
B21	9	890	280	1.50	0.66	1300.00	9100	65.00	2.00	0.30	0.37	5
B22	10	720	280	0.69	0.31	1000.00		55.00	2.00	0.33	0.20	7
B23	11	860	350	0.73	0.41	1400.00	20700	50.00	5.00	0.34	0.37	6
B24	12	1100	530	0.86	0.55	1200.00	12600	54.00	6.00	0.35	0.42	8
B25	13	1100	300	1.40	0.67	790.00	9900	85.00	2.00	0.49	0.36	8
B26	14	750	270	1.40	0.62	1200.00	10900	64.00	2.00	0.59	0.36	6
C10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C11	2	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C12	3	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C13	4	1400	240	1.30	0.72	1000.00	16700	3800.00	3.00	0.32	0.56	8
C14	5	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C15	6	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C16	7	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C17	8	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C21	9	2000	210	2.10	1.00	1100.00	15200	2800.00	2.00	0.28	0.60	4
C22	10	2200	220	2.20	0.97	1300.00	21400	3400.00	2.00	0.34	0.74	5
C23	11	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
C24	12	1700	240	1.40	0.71	1200.00	23100	3600.00	2.00	0.33	0.59	5
C25	13	1100	240	0.87	0.51	910.00	15600	2800.00	2.00	0.25	0.39	37
C26	14	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
D10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
D11	2	1400	330	0.99	0.57	800.00	11600	310.00	2.00	0.25	0.37	6
D12	3	720	230	0.68	0.36	1000.00		3000.00	2.00	0.19	0.23	4
D13	4	500	170	0.60	0.30	1300.00	6000	1000.00	1.00	0.12	0.26	2
D14	5	870	180	0.85	0.41	1300.00	10900	900.00	2.00	0.15	0.38	3
D15	6	2200	210	2.00	0.92	1400.00	30500	1800.00	2.00	0.26	0.66	5
D16	7	1700	180	1.90	0.68	1500.00		1100.00	2.00	0.27	0.55	4

D17	8	770	190	0.90	0.41	780.00	13900	170.00	1.00	0.30	0.34	2
D21	9	1200	240	1.10	0.58	1200.00	11800	250.00	3.00	0.29	0.47	4
D22	10	970	250	1.00	0.46	1100.00	19100	130.00	2.00	0.40	0.36	6
D23	11	370	130	0.38	0.19	510.00		78.00	2.00	0.22	0.16	3
D24	12	660	200	0.68	0.40	850.00	16700	61.00	6.00	0.29	0.39	3
D25	13	6100	100	4.20	2.00	480.00	34500	250.00	2.00	0.56	1.60	12
D26	14	2200	160	1.90	0.85	790.00		180.00	2.00	0.47	0.68	7
E10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
E11	2	1300	190	1.50	1.50	920.00	5500	150.00	6.00	0.40	1.30	2
E12	3	980	390	1.00	0.74	1200.00	15200	360.00	3.00	0.40	0.76	3
E13	4	970	280	0.76	0.46	1600.00	16500	3100.00	2.00	0.36	0.40	6
E14	5	1000	230	0.82	0.41	1700.00	11200	1900.00	2.00	0.28	0.41	5
E15	6	1000	260	1.20	0.58	1400.00	12500	530.00	2.00	0.21	0.42	4
E16	7	1900	270	1.70	0.55	1800.00	10900	2100.00	2.00	0.22	0.42	4
E17	8	2200	280	1.90	0.48	1800.00	13100	1000.00	2.00	0.32	0.35	3
E21	9	1300	300	1.20	0.46	1100.00	14600	210.00	2.00	0.34	0.37	3
E22	10	1100	240	0.97	0.45	1100.00	16500	86.00	2.00	0.33	0.37	4
E23	11	1100	290	0.95	0.50	1500.00	17000	65.00	2.00	0.47	0.45	5
E24	12	1200	370	0.84	0.51	1400.00		73.00	3.00	0.41	0.43	3
E25	13	2400	310	0.88	0.47	1600.00	20400	61.00	3.00	0.44	0.42	5
E26	14	3400	270	0.93	0.43	1300.00	13700	57.00	4.00	0.45	0.41	7
F10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
F11	2	1700	240	1.70	1.10	1300.00	11700	720.00	3.00	0.89	0.87	4
F12	3	1200	280	2.10	0.69	2300.00	19500	3400.00	2.00	0.36	0.44	5
F13	4	1200	240	1.30	0.68	2600.00		1300.00	2.00	0.61	0.74	4
F14	5	2100	280	0.62	0.34	3500.00	13000	120.00	2.00	0.45	0.32	3
F15	6	1000	280	1.40	0.67	4400.00	21500	380.00	2.00	0.20	0.38	3
F16	7	1200	210	1.80	0.55	1300.00	17500	1200.00	2.00	0.24	0.46	3
F17	8	790	270	1.60	0.57	980.00		1000.00	2.00	0.31	0.37	3
F21	9	970	300	1.60	0.68	1200.00	17600	1100.00	3.00	0.44	0.41	3
F22	10	1200	350	1.40	0.62	1900.00	14900	550.00	2.00	0.23	0.37	3
F23	11	920	310	0.66	0.37	1500.00		1500.00	1.00	0.19	0.36	3
F24	12	900	310	0.49	0.31	1400.00	18800	1200.00	1.00	0.23	0.26	2
F25	13	760	300	0.85	0.34	1000.00	17000	820.00	0.50	0.22	0.22	2
F26	14	910	280	1.40	0.49	970.00	7100	4400.00	2.00	0.20	0.31	5
H10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
H11	2	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
H12	3	770	240	0.77	0.33	1100.00	7200	2800.00	4.00	0.36	0.35	3
H13	4	780	190	0.86	0.36	1700.00	5500	1600.00	4.00	0.21	0.33	2
H14	5	1000	150	1.20	0.62	680.00	11700	120.00	2.00	0.49	0.39	2
H15	6	2800	180	1.30	0.31	1600.00	7200	72.00	1.00	0.11	0.25	1
H16	7	1100	280	1.10	0.67	1100.00	13000	130.00	3.00	0.34	0.46	2
H17	8	1100	180	1.30	0.35	1100.00	11500	3900.00	3.00	0.36	0.39	4
H21	9	833	167	0.79	0.35	790.00	10767	4600.00	2.00	0.40	0.31	3
H22	10	1100	220	0.96	0.45	1200.00	15600	400.00	3.00	0.73	0.39	2
H23	11	910	320	0.63	0.52	1100.00	21600	150.00	2.00	0.41	0.37	2
H24	12	1000	220	0.91	0.54	1100.00	23800	7300.00	3.00	0.53	0.42	5
H25	13	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
H26	14	620	130	0.51	0.30	610.00	7000	920.00	1.00	0.28	0.19	2
I10	1	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
I11	2	710	200	0.71	0.36	840.00	15900	110.00	7.00	0.29	0.23	3
I12	3	660	240	0.64	0.27	850.00	16500	470.00	3.00	0.40	0.22	3
J13	4	0	0	0.00	0.00	0.00	0	0.00	0.00	0.00	0.00	0
J14	5	1300	450	0.56	0.44	1100.00	8300	760.00	5.00	0.53	0.34	3
J15	6	1000	680	0.82	0.67	1000.00		650.00	4.00	0.63	0.39	3

J16	7	1100	1000	0.75	0.60	1200.00	6700	710.00	4.00	0.84	0.50	4
J17	8	750	740	0.77	0.49	940.00	6200	510.00	3.00	0.75	0.45	3
J21	9	760	560	1.40	0.64	960.00	7200	360.00	2.00	0.72	0.49	3
J22	10	590	350	0.85	0.42	870.00	12000	210.00	2.00	0.45	0.37	2
J23	11	760	280	0.85	0.83	930.00		110.00	1.00	0.30	0.33	2

Appendix F - Daily soil ingestion rate for each subject during the Cold Lake soil ingestion study

		Soil ingestion rate (mg/d)										
Participant	Day	Al	Ba	Ce	La	Si	Th	Ti	V	U	Y	Zr
A10	1	92	764	118	124	301	163	14053	77	1020	184	269
A11	2	14	807	53	98	533	-101	21699	-144	644	159	305
A12	3	67	1267	109	81	386	-267	16771	-27	765	133	231
A13	4	63	939	73	47	-58	1164	9453	-112	530	47	364
A14	5	36	-194	-51	-19	4	-574	885	-477	-654	-25	-62
A15	6	45	565	42	21	94	-83	559	-135	271	-47	21
A16	7	-66	-199	-8	-28	6	-458	226	-317	-375	-17	-113
A17	8	-16	169	9	-7	-70	-464	1267	-221	-230	8	-52
A21	9	-33	-1466	-23	-33	-24	-204	1404	-252	-330	-25	-36
A22	10	14	640	-12	-19	133	-13	2195	-202	-183	-61	-8
A23	11	59	1608	14	10	220	-822	593	-205	381	-7	75
A24	12	-53	-531	-70	-64	-36	-1121	-99	-457	-664	-135	-120
A25	13	12	619	-12	3	64	-388	80	-84	346	-1	-36
B10	1											
B11	2	-118	-610	-85	-85	-62	-816	684	-599	-996	-168	-246
B12	3	27	982	91	92	46	-396	1562	-61	-31	61	197
B13	4	101	-44	24	29	122	-499	415	-9	190	33	388
B14	5											
B15	6											
B16	7											
B17	8	-6	856	26	13	-15	-700	-49	-373	-478	-9	-68
B21	9	-80	-4804	-68	-75	-160	-952	-174	-399	-244	-70	-101
B22	10	-6	1705	-17	-6	222	-248	-3	-51	183	-185	71
B23	11	-58	-4220	-76	-58	21	-1327	-172	-106	54	-212	-89
B24	12	-41	-178	-37	-36	-30	-525	-88	-350	-285	-136	-75
B25	13	-27	-159	-60	-61	-107	-925	-77	-444	240	-260	-149
C10	1											
C11	2											
C12	3	188	2548	99	144	400	-2047	28400	-614	369	196	369
C13	4	-54	-3603	-46	-52	-106	-759	-125	-250	-216	-48	-59
C14	5											
C15	6											
C16	7											
C17	8	47	-602	31	18	2	-1203	7096	-814	-592	-194	-183
C21	9	454	524	343	310	582	813	18654	111	1267	500	272
C22	10											
C23	11	5	-396	-74	-49	191	-1339	8498	-658	-163	-277	-231

C24	12	270	4437	141	183	537	184	18923	112	896	289	3528
C25	13											
D10	1	234	4273	114	150	196	-20	1348	144	831	160	293
D11	2	-18	276	-40	-35	-107	-533	5432	-313	-478	-100	-118
D12	3	-50	-333	-39	-56	-80	-51	1238	-372	-732	-177	-128
D13	4	-20	-551	-29	-37	-38	-390	594	-162	-304	-93	-51
D14	5	-14	-652	-74	-32	32	-512	1228	-529	-838	-94	-108
D15	6	-73	-1766	-48	-75	-200	-181	168	-299	-396	-246	-79
D16	7	-40	-315	-33	-36	-1	-291	65	-450	-429	-138	-293
D17	8	-13	258	-24	-27	-31	-495	226	-269	-270	-69	-125
D21	9	-16	-1769	-11	-15	37	-299	34	-123	143	7	-4
D22	10	-16	461	-36	-28	-61	-231	147	-144	155	-180	-19
D23	11	19	761	1	5	156	-131	57	378	398	-51	-274
D24	12	352	-67	162	159	244	370	289	-164	391	200	176
D25	13	77	-99	24	10	-108	228	80	-97	197	1	-6
E10	1	162	1542	140	346	-56	402	464	455	828	616	37
E11	2	-49	1803	-9	41	90	-606	739	-161	1	117	-188
E12	3	-26	906	-19	-9	107	-675	7716	-347	-154	-16	-67
E13	4	-112	-4687	-114	-122	-130	-451	2650	-299	-227	-83	-101
E14	5	-62	-2018	-93	-63	-39	-869	1421	-401	-225	-57	-112
E15	6	78	-188	54	8	22	-547	7929	-443	259	62	13
E16	7	154	1540	133	11	163	-518	3377	-416	136	-53	-120
E17	8	-76	-33	-23	-59	-9	-1327	91	-515	-398	-125	-203
E21	9	-37	-1150	-29	-35	192	-456	27	-169	332	1	-174
E22	10	-26	-1947	-18	-8	80	-566	-17	-162	757	-17	2
E23	11	-17	1071	-47	-39	-89	-346	-95	-46	380	-9	-359
E24	12	263	2070	39	29	254	-81	39	146	1067	88	110
E25	13	456	1190	-114	-106	206	-1034	-145	33	1017	-169	-259
F10	1	150	1498	130	192	110	-109	2054	28	2157	300	88
F11	2	36	1135	109	55	185	-440	8165	-347	-208	59	19
F12	3	41	1534	86	96	-80	77	3632	-69	1050	243	15
F13	4	170	467	8	13	104	-503	169	-122	573	71	37
F14	5	24	231	32	-2	173	-650	1011	-445	-560	-37	-31
F15	6	-1	-664	41	-12	111	-671	2340	-327	127	-4	-28
F16	7	-70	-298	-32	-45	-64	-297	409	-416	-522	-158	-131
F17	8	30	1312	63	44	119	-467	2142	-204	268	-51	-43
F21	9	-12	-2925	19	9	33	-197	946	-163	-102	14	-24
F22	10	23	887	-2	2	-73	-630	2436	-344	-143	0	-5
F23	11	34	936	-7	0	123	-517	2239	-240	148	5	-10
F24	12	-1	1104	7	-4	122	-200	1308	-368	-385	-42	-23
F25	13	-12	-89	-9	-25	-106	-673	3611	-260	-243	-78	-35

H10	1											
H11	2	78	2050	-25	-18	32	-444	15730	175	1131	-10	17
H12	3	11	549	14	-31	-15	-272	6057	79	-24	-83	-135
H13	4	26	-529	50	45	79	154	196	84	847	37	38
H14	5	478	1092	99	21	60	47	147	-212	92	52	-24
H15	6	-22	208	-32	18	-26	189	71	-225	251	-39	-50
H16	7	-63	-130	-3	-26	5	79	4629	-145	-221	-31	-101
H17	8	41	520	25	15	60	11	9239	-12	639	17	25
H21	9	27	-43	37	35	152	7	827	155	1463	79	19
H22	10	36	1678	-45	12	245	-609	257	-158	544	-128	-130
H23	11	7	518	-4	17	208	-265	14669	-12	793	-73	5
H24	12											
H25	13	38	1032	24	37	-9	-123	3786	-59	889	16	7
I10	1	10	728	13	17	68	-104	127	274	183	-1	22
I11	2	-32	512	-30	-32	105	-595	759	-185	-76	-66	-98
J13	4	160	3994	37	75	138	0	2342	534	1626	135	100
J14	5	-40	211	-32	-30	-64	-250	270	-258	-296	-46	-30
J15	6	-17	1807	-27	-23	-95	-346	542	-239	183	-132	-14
J16	7	-75	22	-43	-52	-23	-285	104	-286	-249	-80	-76
J17	8	57	4918	121	91	97	-497	1269	-228	2033	89	-22
J21	9	-7	1588	39	44	156	-426	543	24	1085	86	18
J22	10	-36	-225	-24	-16	-31	-348	-34	-146	-174	-47	-57

Appendix G - Soil concentrations for each of the 16 priority PAHs.

Samples that were below the minimum detection limit, value was substituted with half the detection limit. Values that were not detected are assumed 0 and marked with a -.

		PAH (ng/g)															
Soil sample	Distance from facilities (km)	Naphthalene	Acenaphthylene	Acenaphthene	Fluorene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Benz[a]anthracene	Chrysene	Benzo[b]fluoranthene	Benzo[k]fluoranthene	Benzo[a]pyrene	Indeno[1,2,3-cd]pyrene	Dibenz[a,h]anthracene	Benzo[g,h,i]perylene
2	21.7	-	-	-	-	-	-	-	-	-	0.7	0.6	-	-	-	-	1.9
3	0.1	1.9	-	-	0.6	5.9	-	11.8	46.5	4.4	122.1	46.2	11.8	18.8	26.2	8.0	35.4
4	11.8	5.1	-	-	-	-	-	-	-	-	0.3	-	-	-	-	-	-
5	0.1	-	38.2	1.1	-	-	-	0.7	-	-	1.0	0.9	-	-	-	-	-
7	1.9	27.5	39.9	1.9	20.5	23.5	7.8	7.6	-	-	4.3	-	-	-	-	-	-
8	3.3	-	30.4	-	-	-	-	-	-	-	0.7	0.4	-	-	-	-	-
10	32.0	26.4	-	-	0.4	1.4	-	4.2	6.1	0.6	5.2	1.7	-	0.6	-	-	-
12	30.6	-	24.7	-	-	-	-	0.8	2.0	-	1.1	-	-	-	-	-	-
13	8.7	-	22.5	0.5	-	-	-	-	-	-	-	-	-	-	-	-	-
14	34.0	-	22.3	3.8	-	-	-	0.7	-	0.6	2.7	1.0	-	0.6	-	-	-
15	10.8	100.5	15.9	-	0.1	1.9	3.4	13.8	13.5	5.2	10.8	10.5	4.2	7.1	6.5	-	5.7
17	18.9	-	22.4	-	1.0	12.5	-	5.8	2.6	0.6	6.0	2.9	0.3	0.6	-	-	-
21	1.7	-	20.5	18.1	4.9	29.2	-	5.9	7.0	-	6.0	3.5	-	-	-	-	-
22	6.4	-	6.3	8.7	2.7	35.4	-	154.2	66.1	5.3	33.0	52.9	196.5	17.9	66.8	-	20.9
23	9.9	-	17.5	-	-	4.3	0.4	18.4	18.6	9.0	19.8	25.3	9.5	19.1	16.3	2.3	16.4
24	26.0	-	30.9	11.0	-	-	2.0	-	-	-	-	-	-	-	-	-	-
26	10.4	-	17.2	39.7	2.4	33.1	6.1	14.0	15.3	3.8	8.1	8.6	1.9	3.8	3.3	-	4.8
28	0.1	-	93.5	-	1.4	5.4	0.6	2.0	6.2	0.1	1.2	3.3	-	-	-	-	2.6

Appendix H- Atmospheric particulate concentrations for each of the 16 priority PAHs analyzed from GFF filters.

Samples that were below the minimum detection limit, value was substituted with half the detection limit. Values that were not detected are assumed 0 and marked with a -.

	PAH (pg/m^3)															
Day	Naphthalene	Acenaphthylene	Acenaphthene	Fluorene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Benz[a]anthracene	Chrysene	Benzo[b]fluoranthene	Benzo[k]fluoranthene	Benzo[a]pyrene	Indeno[1,2,3-cd]pyrene	Dibenz[a,h]anthracene	Benzo[g,h,i]perylene
1	0.73	6.33	-	-	3.93	-	0.55	0.43	-	-	-	-	-	-	-	-
2	8.33	4.84	2.81	-	1.90	-	0.15	0.28	-	-	-	-	-	-	-	-
3	0.73	1.37	-	-	2.01	-	0.16	0.26	-	-	-	-	-	-	-	-
4	-	0.93	-	-	0.16	-	0.07	-	-	-	-	-	-	-	-	-
5	26.10	16.26	-	-	16.09	-	-	2.74	-	1.09	-	-	41.90	-	-	-
6	3.42	1.10	-	-	1.02	-	0.21	0.06	-	-	-	-	-	-	-	-
7	2.38	0.12	-	-	0.68	-	-	0.55	-	-	0.18	-	-	0.95	-	1.82
8	-	-	-	-	0.76	-	0.25	0.24	-	-	-	-	-	-	-	-
9	0.73	-	-	-	-	-	-	0.14	-	-	-	0.21	-	-	-	0.70
10	0.73	-	-	-	-	-	-	0.35	-	-	-	-	-	-	-	-

Appendix I - Atmospheric gaseous concentrations for each of the 16 priority PAHs analyzed from PUF cartridges.

Samples that were below the minimum detection limit, value was substituted with half the detection limit. Values that were not detected are assumed 0 and marked with a -.

	PAH (ng/m3)															
Day	Naphthalene	Acenaphthylene	Acenaphthene	Fluorene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Benz[a]anthracene	Chrysene	Benzo[b]fluoranthene	Benzo[k]fluoranthene	Benzo[a]pyrene	Indeno[1,2,3-cd]pyrene	Dibenz[a,h]anthracene	Benzo[g,h,i]perylene
1	3.7	3.2	2.5	9.3	49.5	-	5.9	9.8	0.3	3.3	0.6	-	1.3	-	-	-
2	-	5.2	1.2	8.3	48.2	-	6.0	4.1	2.4	1.3	-	-	1.3	-	-	-
3	-	0.7	1.2	10.6	96.3	10.1	16.0	17.4	1.3	4.1	0.2	-	1.3	-	-	-
4	-	0.2	1.2	6.5	41.2	0.8	7.1	6.2	-	2.2	-	-	-	-	-	-
5	39.9	41.4	8.4	6.4	45.1	2.6	1.6	6.4	4.0	1.5	-	-	18.6	-	-	-
6	318	136.3	52.1	181.6	1,525.8	440.4	15.3	248.4	54.7	154.5	40.4	-	-	-	-	-
7	-	59.0	9.0	15.4	108.7	18.5	9.1	16.0	2.3	5.8	0.7	-	9.0	-	-	-
8	-	186.0	21.5	37.7	383.5	77.6	3.7	55.4	14.4	27.2	9.1	2.2	-	-	-	-
9	-	-	1.2	8.3	1.6	-	0.1	-	-	-	-	-	-	-	-	-
10	-	-	1.2	12.6	81.9	14.7	3.4	14.2	3.0	6.5	1.6	-	213.0	-	-	-

Appendix J - Method detection limits for soils, GFF, and PUF cartridges.

Table J.1: PAH method detection limit and calculation for soil samples. Ce is analyte concentration, Ve, is injection analyte volume, Vs is average sample mass measured, R% is the average recovery rate, and MDL is the calculated method detection limit.

PAH	Ce' (ng/mL)	Ve (mL)	Vs (g)	R%	MDL ng/g
Naphthalene	0.343683	1	1.717341	24.42561	0.82
Acenaphthylene	0.076227	1	1.717341	24.33039	0.18
Acenaphthene	0.659067	1	1.717341	23.65808	1.62
Fluorene	0.21459	1	1.717341	40.199	0.31
Phenanthrene	0.155418	1	1.717341	33.65142	0.27
Anthracene	0.038777	1	1.717341	30.02193	0.08
Fluoranthene	0.099717	1	1.717341	45.36169	0.13
Pyrene	0.066034	1	1.717341	36.19563	0.11
Benz[a]anthracene	0.264983	1	1.717341	49.09347	0.31
Chrysene	0.152506	1	1.717341	57.78152	0.15
Benzo[b]fluoranthene	0.232417	1	1.717341	49.21396	0.27
Benzo[k]fluoranthene	0.266377	1	1.717341	61.1021	0.25
Benzo[a]pyrene	0.965446	1	1.717341	46.92114	1.20
Ind[123cd[pyrene	1.284471	1	1.717341	48.80843	1.53
Dibenz(a,h)anthracene	4.111251	1	1.717341	51.69556	4.63
Benzo[ghi]perylene	1.006194	1	1.717341	44.78616	1.31

Table J.2: Method detection limit and calculation for particulate PAH samples measured on GFFs. Ce is analyte concentration, Ve, is injection analyte volume, Vs is average sample mass measured, R% is the average recovery rate, and MDL is the calculated method detection limit.

PAH	Ce' (ng/mL)	Ve (mL)	Vs (m3)	R%	MDL pg/m^3
Naphthalene	0.343682864	1	844.7095	28.04	1.45
Acenaphthylene	0.076226958	1	844.7095	38.25333	0.24
Acenaphthene	0.659066901	1	844.7095	52.68333	1.48
Fluorene	0.214590366	1	844.7095	52	0.49
Phenanthrene	0.155418091	1	844.7095	59.05167	0.31
Anthracene	0.038777257	1	844.7095	39.76833	0.12
Fluoranthene	0.099717419	1	844.7095	84.84	0.14
Pyrene	0.066033818	1	844.7095	63.11333	0.12
Benz[a]anthracene	0.264983112	1	844.7095	56.19	0.56
Chrysene	0.152505823	1	844.7095	72.34333	0.25
Benzo[b]fluoranthene	0.232417364	1	844.7095	75.84667	0.36
Benzo[k]fluoranthene	0.266376841	1	844.7095	75.49	0.42
Benzo[a]pyrene	0.965446267	1	844.7095	69.29	1.65
Ind[123cd[pyrene	1.284470887	1	844.7095	80.34	1.89
Dibenz(a,h)anthracene	4.111250603	1	844.7095	77.28333	6.30
Benzo[ghi]perylene	1.00619433	1	844.7095	84.56333	1.41

Table J.3: Method detection limit and calculation for gaseous PAH samples measured on PUF cartridges. Ce is analyte concentration, Ve, is injection analyte volume, Vs is average sample mass measured, R% is the average recovery rate, and MDL is the calculated method detection limit.

PAH	Ce' (ng/mL)	Ve (mL)	Vs (m3)	R%	MDL pg/m^3
Naphthalene	0.343682864	1	844.7095	25.07259	1.62
Acenaphthylene	0.076226958	1	844.7095	26.59438	0.34
Acenaphthene	0.659066901	1	844.7095	33.33813	2.34
Fluorene	0.214590366	1	844.7095	45.62938	0.56
Phenanthrene	0.155418091	1	844.7095	47.37313	0.39
Anthracene	0.038777257	1	844.7095	30.82563	0.15
Fluoranthene	0.099717419	1	844.7095	137.7863	0.09
Pyrene	0.066033818	1	844.7095	52.20188	0.15
Benz[a]anthracene	0.264983112	1	844.7095	54.00063	0.58
Chrysene	0.152505823	1	844.7095	64.02375	0.28
Benzo[b]fluoranthene	0.232417364	1	844.7095	66.36938	0.41
Benzo[k]fluoranthene	0.266376841	1	844.7095	63.55438	0.50
Benzo[a]pyrene	0.965446267	1	844.7095	43.1256	2.65
Ind[123cd[pyrene	1.284470887	1	844.7095	65.83667	2.31
Dibenz(a,h)anthracene	4.111250603	1	844.7095	67.61267	7.20
Benzo[ghi]perylene	1.00619433	1	844.7095	58.67226	2.03