

**LONG TERM DYNAMICS AND POTENTIAL REMEDIATION
OF FINE TEXTURED SOILS AND GROUND WATER CONTAMINATED
WITH CHLORINATED ORGANIC COMPOUNDS AND SALTS**

by

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ABSTRACT

Contamination of land, water, and air is a widespread concern often associated with anthropogenic activities. Remediation of contaminated sites is necessary to minimize negative impacts on human health and the environment and allow for safe, productive use of land. Although contaminated sites often have fine textured soils and/or multiple contaminants, research on remediation of such sites is lacking. Fine textured soils can make remediation more difficult by hindering movement of contaminants or amendments and by promoting anaerobic conditions. Contaminant mixtures can complicate remediation since each contaminant has unique characteristics to address and can hamper remediation of other contaminants.

The objective of this research was to characterize a contaminated site with fine textured soils and multiple contaminants and investigate the potential of various remediation techniques. The research site was the former Ellerslie Waste Management Facility which treated laboratory waste from 1972 to 2007 in Edmonton, Alberta. Prior to 1983, a waste water pond cracked and leaked. Contaminant dynamics over 30 years were characterized by identifying soil and ground water contaminants based on Alberta Tier 1 guidelines, identifying temporal trends with Mann Kendall analysis, and identifying spatial trends with two and three dimensional mapping. The potential of anaerobic biostimulation and bioaugmentation treatment of chloroform were investigated in microcosm experiments using soil from the Ellerslie site and a secondary contaminated site. Microcosms were amended with canola oil, acetate, lactate, nitrate, or sulfate. The soil microbial community was characterized by comparing bacterial and fungal communities from various soil depths in, up gradient, and down gradient of the pond. The potential of chemical reduction treatment of chloroform was investigated in an anaerobic bottle experiment using micro scale zero valent iron with and without soil. The potential of leaching treatment of salinity was investigated in soil column experiments using two soil composites of differing textures with and without calcium nitrate amendment.

Of the 18 soil and 37 ground water contaminants identified, chloroform, dichloromethane, and salinity were of greatest concern based on their frequency and magnitude of detection. Temporal contaminant trends were inconsistent, possibly due to heterogeneous contaminant distribution or movement. A clear spatial association with the pond was identified, as expected. There is some evidence of northward chloride and sodium movement with ground water flow. The bioremediation experiments did not yield any meaningful changes in microcosm headspace chloroform or formation of degradation products over time. Biostimulation and bioaugmentation with the inoculation microorganisms used in this research are not likely effective options for the Eilerslie site. This is supported by the lack of chlorinated methane degrading microorganisms enriched in the pond. However, the enrichment of *Geobacter* species in the pond indicated benzene biodegradation may be occurring. Bacterial richness and diversity were decreased in the pond, likely a result of contaminant toxicity. The fungal community was more similar across the Eilerslie site than the bacterial community. Chemical reduction of nearly 2 g L⁻¹ chloroform using zero valent iron was rapid and complete. Headspace chloroform was not detected after 0.5 and 2 days in the soil zero valent iron and zero valent iron treatments, respectively. Dichloromethane which formed during the degradation process was removed by day 2 in both treatments. The leaching experiments showed potential for treatment of fine textured soils. Soil electrical conductivity, sodium adsorption ratio, sodium, chloride, and sulfate were greatly reduced by eight rounds of leaching. Greatest salt removal occurred during the first leaching round. Calcium nitrate increased the saturated hydraulic conductivity of the finer textured soil from $4.12 \pm 0.46 \times 10^{-8}$ to $1.29 \pm 0.14 \times 10^{-7}$ m s⁻¹ and increased chloride leaching.

The soil and ground water contaminant inventory and knowledge of their spatial distribution will aid in development and application of a site remediation plan. Potential remediation methods include chemical reduction with zero valent iron for chlorinated methanes and leaching for salinity. Zero valent iron treatment could be implemented with a permeable reactive barrier, in

situ injection, or ex situ batch reactor. Leaching treatment could be implemented with an interceptor trench or ex situ soil washing. Contaminant maps will be important in guiding soil excavation or placement of in situ remediation infrastructure. Site characterization and remediation methods explored in this research can be applied to similar contaminated sites.

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I. INTRODUCTION

1. CONTAMINATION OVERVIEW

Environmental contamination is a serious problem affecting land, water, and air across the planet. Large scale contamination began in the 1800s when industrialization and the human population increased rapidly and significantly (Petts et al 1997). Urbanization and ignorance of, or disregard for, the consequences of chemical release into the environment increased the severity and distribution of contamination issues (Philp et al 2005). Major industrial activities contributing to land contamination include mineral extraction, chemical synthesis, manufacturing, and waste disposal (Petts et al 1997). Substances considered to be contaminants are numerous and variable. Many contaminated sites contain more than one substance of concern. Some of the most common contaminants include solvents, petroleum products, volatile organic compounds, and trace elements (Singh et al 2009).

In Canada, land is considered contaminated when the concentration of a contaminant exceeds naturally occurring background values and regulatory guidelines thus posing a risk to human health or the environment (Treasury Board of Canada Secretariat 2015). The number of contaminated sites occurring in various jurisdictions and countries varies considerably with information source and criteria. Many believe the numbers of listed sites and their associated remediation costs are highly underestimated (Naeth 2015). There are over 22,000 contaminated or suspected contaminated sites in Canada, either on federal land or under at least partial federal financial responsibility (Treasury Board of Canada Secretariat 2015). The number of contaminated sites under the responsibility of private companies or individuals, provincial or municipal governments, or enterprise Crown corporations is uncertain and not listed in any comprehensive national database. Environmental Careers Organization Canada (2007) estimates the number of non federal contaminated sites in Canada to be approximately 28,000; however, the National Round Table on the Environment and the Economy (2003) estimates the number in urban Canadian locations alone to be approximately 30,000.

The reported liability for remediating federal contaminated sites in Canada is approximately 4.9 billion dollars, although an assessment by the Parliamentary Budget Officer has estimated that the actual remediation cost will be at least 7 billion dollars (Story and Yalkin 2014). Singh et al (2009) estimate the cost would be approximately 250 to 500 million dollars for soil remediation in the United States of America (US) alone, where there are at least 250,000 sites that are listed as contaminated. Remediation for these contaminated sites is estimated to have a market value

of 12 billion US dollars, equivalent to approximately 30 % of the international remediation market value of 30 to 35 billion US dollars.

2. CONTAMINANTS

2.1. Salts

Salt affected soils contain significant quantities of inorganic soluble elements or compounds in the aqueous phase (Corwin 2003). The most common salts in soils include sodium (Na^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), chloride (Cl^-), and sulfate (SO_4^{2-}) (Allison et al 1954, Miller and Curtin 2008). Other salts, usually present in soil in smaller quantities, include potassium (K^+), bicarbonate (HCO_3^-), carbonate (CO_3^{2-}), and nitrate (NO_3^-) (Allison et al 1954). Salts are relatively mobile and can move easily within the soil profile; positive ions are typically less mobile than negative ions due to their participation in cation exchange reactions on soil surfaces (Alberta Environment 2001, Environment Canada 2001).

A saline soil has electrical conductivity $> 4 \text{ dS m}^{-1}$ and exchangeable sodium percentage < 15 (sodium adsorption ratio < 13) (Jordan et al 2004). These soils typically have $\text{pH} < 8.5$ and may be referred to as white alkali soils due to the visible salts at the soil surface (Allison et al 1954, Brady and Weil 2008). In the past, soils with electrical conductivity $> 4 \text{ dS m}^{-1}$ and exchangeable sodium percentage > 15 were called saline-alkali soils (Jordan et al 2004). The term alkali was replaced with sodic by 1979. Soils with exchangeable sodium percentage > 15 , $\text{pH} > 8.3$, and soluble bicarbonate and carbonate are now considered alkali soils (Gupta and Abrol 1990, Jordan et al 2004). Over time, sodic soils develop characteristic features. Downward movement of dispersed clay particles causes the soil surface to be coarse and underlain by an area of low permeability with a columnar or prismatic structure (Allison et al 1954). Sodic soils generally have pH between 8.5 and 10 due to increased hydrolysis and formation of sodium hydroxide (NaOH). Sodic soils may be referred to as black alkali soils because of the dispersion and visible accumulation of organic matter at the soil surface (Allison et al 1954, Brady and Weil 2008). Although sodic soils are defined as having exchangeable sodium percentage > 15 , soil properties change gradually with increasing sodium, rather than changing at abrupt specific thresholds (Allison et al 1954).

2.1.1. Sources and occurrence

Salts can be naturally occurring in the soil profile. Salts are present in the structure of primary minerals of soil parent material (Allison et al 1954, Jordan et al 2004). Weathering of these

minerals causes the salts to be solubilized, resulting in primary or fossil salinity (Allison et al 1954, Qadir and Oster 2004). Salts can be from ancient marine deposits, drainage basins, or inland seas (Allison et al 1954, Jordan et al 2004). Salts may come from ocean water or spray in coastal regions. Surface water can be a source of salts during flooding and ground water can be a source of salts when it rises through the soil profile to the soil surface. The amount of salt brought by surface and ground water depends on the salt content of the material which the water has contacted.

Soil salinity is typically a concern in areas with arid or semiarid climates (Jordan et al 2004). Dry areas have insufficient precipitation for complete leaching and high rates of evaporation (Allison et al 1954). Saline soils do not usually occur in humid regions because any salts in the soil are washed down the profile, where they will ultimately enter the ground water and are transported to streams, lakes, or oceans. Areas subject to restricted drainage can often become saline (Jordan et al 2004). Drainage can be restricted due to a high water table or low soil permeability (Allison et al 1954). Hydrologically isolated basins with no outlet are common in dry regions. These basins receive salt containing water from higher areas which collects and raises the water table or even ponds on the soil surface. Evapotranspiration of this salt containing water leaves behind salts in the soil. Low soil permeability can be caused by a fine soil texture, poor soil structure, or soil compaction. Soils in low landscape positions have a greater risk of salinization because it is more likely that water will reach the soil surface and leave behind salts upon water evaporation (Jordan et al 2004). Anthropogenic activities associated with salt contamination include irrigation, oil and gas production, salt processing and storage for road maintenance, rendering, and use of saline material for various industrial purposes (Alberta Environment 2001, Jordan et al 2004).

2.1.2. Effects

Salt contamination impacts several environmental components. Soil is highly affected by the presence of sodium. Although calcium and magnesium are preferentially adsorbed on soil surfaces over sodium, when sodium dominates the soil solution in sodic soils, it replaces calcium and magnesium on adsorption sites (Allison et al 1954). Sodium is a monovalent cation which causes dispersion of soil particles (Jordan et al 2004) because its low valency increases the thickness of the diffuse double layer and forces soil particles away from each other (Quirk 2001). Sodium induced dispersion can cause soil structure issues such as swelling, surface crusting, and hard setting (Qadir and Oster 2004). This can decrease water and air movement into and within the soil profile and increase runoff and erosion (Jordan et al 2004, Qadir and

Oster 2004). Saline sodic soils remain flocculated as multivalent cations in the soil solution prevent particle dispersion caused by sodium (Allison et al 1954). The high ionic concentration of the soil solution and the high charge of the multivalent cations compress the diffuse double layer and allow the soil particles to remain close together (Quirk 2001).

Soil salinity can affect plants in multiple ways. High concentrations of salt in the soil increase the osmotic pressure with which water is held in the soil, making it more difficult for plants to access (Corwin 2003, Qadir and Oster 2004). Salts can harm plants through specific ion toxicities and nutrient imbalances (Qadir and Schubert 2002, Qadir and Oster 2004). The poor structure of sodic soils can impede seedling emergence and plant root penetration. The effects of salinity on plants can be seen as reduced growth and yield (Corwin 2003).

Salinity reduces the quality of ground water and may cause it to be unsuitable for uses such as irrigation and consumption by humans or other organisms. Salinity is not typically a concern for drinking water safety but may cause an undesirable taste (World Health Organization 2011). Ground water may transport saline water to fresh water systems where it can potentially impact aquatic organisms (Environment Canada 2001).

2.1.3. Regulations

The Environmental Protection and Enhancement Act guides the remediation of salt impacted sites in Alberta (Alberta Environment 2001). According to the Act, remediation must reduce contaminant concentrations to or below generic guidelines, site specific objectives, or background levels. The remaining contamination must not negatively affect any receptors under the current land use and the land capability must not be lower than it was prior to contamination.

According to Alberta Tier 1 soil remediation guidelines, topsoil with electrical conductivity $< 2 \text{ dS m}^{-1}$ is rated good, 2 to 4 dS m^{-1} is rated fair, 4 to 8 dS m^{-1} is rated poor, and $> 8 \text{ dS m}^{-1}$ is rated unsuitable (Alberta Environment and Parks 2016). Subsoil with electrical conductivity $< 3 \text{ dS m}^{-1}$ is rated good, 3 to 5 dS m^{-1} is rated fair, 5 to 10 dS m^{-1} is rated poor, and $> 10 \text{ dS m}^{-1}$ is rated unsuitable. Topsoil or subsoil with sodium adsorption ratio < 4 is rated good, 4 to 8 is rated fair, 8 to 12 is rated poor, and > 12 is rated unsuitable. For commercial or industrial land, remediation guidelines of 4 dS m^{-1} for electrical conductivity and 12 for sodium adsorption ratio are adopted from the Interim Canadian Environmental Quality Criteria for Contaminated Sites (Alberta Environment 2001).

The Alberta Tier 1 guideline for ground water remediation of electrical conductivity is 1 dS m^{-1} for agricultural land (Alberta Environment and Parks 2016). The Tier 1 guideline for ground

water remediation of chloride is 120 mg L^{-1} for natural areas, residential areas, parkland, commercial, and industrial land and 100 mg L^{-1} for agricultural land. The Tier 1 guideline for ground water remediation of sodium is 200 mg L^{-1} .

Environmental regulations for other countries are difficult to find and typically not readily available on government web sites. In the United States of America, the aquatic health guidelines for chloride in fresh water are 230 and 860 mg L^{-1} for chronic and acute exposure, respectively (United States Environmental Protection Agency 2015).

2.2. Chlorinated Organic Compounds

Chlorinated organic compounds are carbon based molecules which contain chlorine. A wide variety of chlorinated organic compounds exist, including chlorinated methanes, chlorinated ethanes, chlorinated ethenes, and chlorinated aromatics. These types of chlorinated organic compounds are typically volatile and recalcitrant (Huang et al 2014). This research focuses on the chlorinated methanes chloroform (CHCl_3) and dichloromethane (CH_2Cl_2).

Chloroform is also known as trichloromethane, methane trichloride, trichloroform, methyl trichloride, and formyl trichloride (World Health Organization 2004). Chloroform is a volatile, clear, colourless liquid at room temperature (Canadian Council of Ministers of the Environment 1999a, World Health Organization 2004). It dissolves slightly in water with a solubility of 7.4 g L^{-1} at $25 \text{ }^\circ\text{C}$ (Cappelletti et al 2012). Due to its volatility and low sorption, chloroform is typically lost from surface soils via volatilization (World Health Organization 2004). At equilibrium, over 99 % of chloroform in a given system is expected to partition into the atmosphere (McCulloch 2003).

Dichloromethane is also known as methylene chloride, methylene dichloride, and methylene bichloride (Canadian Council of Ministers of the Environment 1999b, Health Canada 2011). At room temperature, dichloromethane is a volatile, clear, colourless liquid with a sweet odour. Dichloromethane in surface soils readily evaporates to the atmosphere due to its high volatility and low sorption (United States Environmental Protection Agency 1994). Once in the atmosphere, the compound undergoes photochemical degradation with a half life of 53 to 127 days (Wilson et al 2000).

2.2.1. Sources and occurrence

Chlorinated organic compounds are used extensively in industry around the world. They have high solvent capacity, high stability, and low flammability (Armstrong and Green 2004, Martin-Martinez et al 2013). These properties make chlorinated organic compounds suitable for use as

solvents, cleaners, and degreasing agents. Pesticides, wood preservatives, and adhesives also contain chlorinated organic compounds. Chlorinated organic compounds contaminate the environment through their release during production, use, storage, processing, and disposal (Canadian Council of Ministers of the Environment 1999a, Huang et al 2014).

According to McCulloch (2003), the global chloroform flux through the environment is relatively steady at approximately 660 Gg yr⁻¹. Approximately 90 % of chloroform is of natural origin, mainly occurring from off shore sea water and soil processes. Smaller amounts are released into the environment due to volcanic and geologic activity. Anthropogenic activities result in the release of approximately 66 Gg yr⁻¹. Chloroform is miscible with most organic solvents and relatively unreactive; therefore, it is used widely as a solvent (Huang et al 2014). Chloroform is also used as a degreasing agent and for refrigerant, plastic, pharmaceutical, pesticide, and aerosol propellant production (Canadian Council of Ministers of the Environment 1999a, World Health Organization 2004). Chloroform was used as a surgical anaesthetic and an ingredient in cough syrups and toothpastes from 1847 until 1976 when it was banned from consumer products due to health concerns (Rosenthal 1987). Chloroform can be released during paper bleaching and water treatment due to the reaction between chlorine and naturally occurring organic compounds such as humic and fulvic acids (Canadian Council of Ministers of the Environment 1999a, World Health Organization 2004, Huang et al 2014).

Dichloromethane is not naturally occurring in the environment (United States Environmental Protection Agency 1994, Health Canada 2011). Contamination is the result of the industrial and household use of products containing the chemical dichloromethane (Wilson et al 2000). Dichloromethane containing products include paint and furniture strippers, solvents, pesticides, metal and automotive cleaners, and aerosol sprays (United States Environmental Protection Agency 1994, Wilson et al 2000). Dichloromethane is also used to produce foams and photographic film.

2.2.2. Effects

Most chlorinated organic compound impacts are related to human health concerns. Many chlorinated organic compounds are considered toxic and carcinogenic (Martin-Martinez et al 2013). Humans can be exposed to chlorinated organic compounds in various ways, such as through inhalation, ingestion of food and water, and direct contact (Huang et al 2014). Chloroform was used historically as an anaesthetic due to its effects on the central nervous system (Chou and Spoo 1997, World Health Organization 2004). Inhalation and ingestion of chloroform cause cardiovascular, liver, and kidney damage in humans (World Health

Organization 2004). Miscarriages, birth defects, and liver and kidney cancer in laboratory mice and rats have been reported (Chou and Spoo 1997). Chloroform is a probable carcinogen for humans and may be linked to cases of colon and bladder cancer. Inhalation of dichloromethane has acute negative impacts on the human nervous and cardiovascular systems (United States Environmental Protection Agency 1994, Armstrong and Green 2004). Direct contact with dichloromethane can result in skin and eye irritation or burns. Dichloromethane does not appear to be carcinogenic to humans (Armstrong and Green 2004).

Chlorinated organic compounds have minor effects on the abiotic environment. Chloroform has only very slight contributions to low altitude ozone formation, stratospheric ozone depletion, and the greenhouse effect (McCulloch 2003).

2.2.3. Regulations

The Environmental Protection and Enhancement Act guides remediation of sites contaminated with chlorinated organic compounds in Alberta. The Alberta Tier 1 guidelines for chloroform are 0.0029 mg kg⁻¹ in soil and 0.0018 mg L⁻¹ in ground water (Alberta Environment and Parks 2016); guidelines for dichloromethane are 0.10 mg kg⁻¹ for soil in natural, residential, parkland, commercial, and industrial areas, 0.052 mg kg⁻¹ for soil in agricultural land, and 0.05 mg L⁻¹ for ground water. The Health Canada drinking water guideline for dichloromethane is 0.05 mg L⁻¹ (Health Canada 2011).

The international drinking water guideline for chloroform is 0.3 mg L⁻¹ for the World Health Organization (World Health Organization 2011). International drinking water guidelines for dichloromethane are 0.02 mg L⁻¹ for the World Health Organization (World Health Organization 2011) and 0.004 mg L⁻¹ for Australia (National Health and Medical Research Council 2011). The drinking water guideline for total trihalomethanes is 0.25 mg L⁻¹ for Australia (National Health and Medical Research Council 2011). Regulations and specific standards for other countries are not readily available.

3. REMEDIATION

Public concern for land contamination began around the 1970s and continues today (Philp et al 2005). Contaminant behaviour varies greatly between and within contaminant types due to inherent differences in physical, chemical, and biological properties and differences in the environments where the contaminants are found. A multitude of remediation methods have been developed to treat contaminated media. These strategies all have the common goal of

preventing, minimizing, or mitigating adverse effects of contamination on human or environmental health (Zvomuya and Murata 2012).

Remediation methods can be broadly categorized as in situ or ex situ based on their application. Ex situ strategies require excavation or pumping of contaminated material prior to its treatment whereas in situ treatments do not (Singh et al 2009). Remediation methods can be more narrowly classified as physical, chemical, biological, or thermal according to their operation. Physical remediation methods include mechanical removal of contaminants by washing and scrubbing soil with water or extracting vapours with vacuum pumps (Sims and Sims 1991, Khan et al 2004, Singh et al 2009). Chemical techniques include addition of chemicals to react with contaminants so that they are more easily removed, less toxic, or less mobile. Biological strategies use microorganisms or plants to degrade, immobilize, or remove contaminants (Sims and Sims 1991, Knox et al 1999, Khan et al 2004, Singh et al 2009). Thermal methods use heat to degrade, vaporize, or entrap contaminants (Khan et al 2004, Singh et al 2009).

The most straightforward remediation technique currently available is commonly known as dig and dump. This method entails excavation of impacted material and disposal at a suitable landfill (Philp et al 2005). This method could be applied to both salt and chlorinated hydrocarbon contaminated sites. It has low risk of harming potential receptors (Alberta Environment 2001). However, it is expensive due to material removal, transportation, and disposal costs. It is a popular method among industries as it removes their likely liability in a short period of time. The treatment does not result in contamination removal from, or stabilization in, the excavated soil material. It is less popular with many environmental practitioners as it removes soil from the contaminated area (Naeth 2015). Additional expenses are usually associated with replacing or building soils to fill the void created by soil removal.

3.1. Salts

The typical treatment for saline soils is leaching. Leaching requires net transport of water and salts in a downwards direction through the soil profile. In some cases, precipitation may be sufficient to wash salts from the soil. In other cases, irrigation may be required. Improvement of soil drainage may be needed (Alberta Environment 2001, Lee et al 2013). In many jurisdictions, if leachate release has potential to cause further damage to living organisms or previously uncontaminated media, it must be collected and properly discarded (Alberta Environment 2001). Soil can be excavated and washed in ex situ salt removal treatment (Sastre-Conde et al 2015). If soils are also sodic, sodium must be replaced by calcium on the soil surface prior to leaching

(Alberta Environment 2001). Calcium can be added in various forms. Commonly used solid calcium amendments are gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and calcium nitrate ($\text{Ca}(\text{NO}_3)_2$). Calcium amendments may be dissolved and added to the soil in liquid form to increase the depth of addition and reduce treatment time.

Vegetation can be used to promote leaching through the entire root zone rather than just the soil volume with calcium added (Qadir and Oster 2004). Oxidation of root exudates increases the partial pressure of carbon dioxide (CO_2) in the root zone. The carbon dioxide dissolves to form carbonic acid (H_2CO_3) which dissociates and releases protons which then react with the calcite (CaCO_3) naturally present in soil to produce calcium ions. Calcium replaces sodium on soil surfaces and allows it to be leached through the soil profile. Organic acids and protons released by plants can also facilitate dissolution of calcite. Plants increase hydraulic conductivity of soil by creating macro pores, increasing aggregate stability, and removing air trapped in the conducting pores. Plants contribute nutrients to the soil which can be lost during the leaching process. Water used by plants is unavailable to percolate through the soil and transport salts which accumulate in areas of discharge and evaporation (Mankin and Koelliker 2000). In a field study by Kushiev et al (2005), land in the Aral Sea basin which had been abandoned due to salinization was planted with *Glycyrrhiza glabra* Linnaeus (licorice) for four years. Following treatment, the land supported increased seed germination and biomass yield for *Triticum* L (wheat) and *Gossypium* L (cotton) crops relative to a control of bare fallow land. Treatment also decreased extractable anion and cation concentrations in the soil and maintained or increased the depth to the water table.

Vegetation can remediate salt affected soils through the process of phytoextraction. Halophytes are plants that exclude salts from entering their tissues, accumulate salts in their tissues, or conduct and excrete salts into the atmosphere through salt glands (Jesus 2015). Salt accumulators with high above ground biomass can remove the most salt from the soil (Qadir and Oster 2004). Estimated salt uptake ranges from $91 \text{ kg ha}^{-1} \text{ y}^{-1}$ for *Lotus corniculatus* L (birdsfoot trefoil) to $5,376 \text{ kg ha}^{-1} \text{ y}^{-1}$ for *Sesuvium portulacastrum* L (shoreline purslane) (Jesus 2015). Perennial plants are desirable as they have longer growing seasons for active salt uptake to take place. Phytoextraction is dependent on soil salt concentration with greater reduction in electrical conductivity with high initial values. Greater reduction in sodium adsorption ratio occurs at lower initial sodium adsorption ratio, likely due to greater hydraulic conductivity. Phytoextraction and leaching can be combined using salt accumulators that can withstand temporary saturated conditions (Qadir and Oster 2004). Providing more water than

the amount required for plant use means that surplus water is available for transporting salts downwards out of the soil profile.

Soil salinity may be addressed by electro kinetic treatment. In this in situ technique, a direct current is applied to an area of soil spanned by electrodes (Cho et al 2009, Lee et al 2013). Electro kinetic treatment moves salts through the soil by two primary mechanisms: electro migration, movement of ions within pore water; and electro osmosis, transport of water from the anode to the cathode. The electrodes induce hydrolysis reactions which produce hydrogen (H^+) and hydroxyl (OH^-) ions and decrease soil pH to < 3 at the anode and increase soil pH to 8 to 12 near the cathode (Cho et al 2009). Electro kinetic remediation is more effective than most other techniques in soils with low permeability and fine texture (Essa et al 2013). In a laboratory experiment using a voltage of $1 V cm^{-1}$, Cho et al (2009) reported significant transport of anions towards the anode but no transport of cations. Nitrate transport and removal was greatest due to its high solubility and conversion to nitrogen (N_2) gas at low pH. Following electro kinetic treatment, electrical conductivity was lower near the cathode due to transport and accumulation of salts. Using pulses of energy instead of a constant current can substantially decrease energy consumption while achieving the same amount of salt removal (Lee et al 2013).

3.2. Chlorinated Organic Compounds

Chlorinated organic compounds can be biodegraded under aerobic and anaerobic conditions (Cappelletti et al 2012). Aerobic chlorinated methane degradation is typically part of a cometabolic process whereby species of microorganisms require a separate carbon source. The microorganisms produce non specific oxygenase enzymes that catalyze degradation of the carbon source as a primary growth substrate and degradation of chlorinated organic compounds (Arp et al 2001). Aerobic chlorinated methane degradation results in the complete conversion of organic chlorine to chloride ions (Cappelletti et al 2012). Carbon sources for aerobic cometabolic chloroform and dichloromethane degradation include methane, propane, butane, hexane, toluene, ammonia, and acetone. Degradation rate depends on enzyme competition among substrates, toxicity of degradation products, and energy consumption. Dey and Roy (2011) reported that *Bacillus* sp 2479 at least partially degraded $40 \mu M$ chloroform over 48 hours as a primary growth substrate under aerobic conditions. The detected decrease in chloroform was accompanied by increased chloride ions and dry cell mass.

Under anaerobic conditions, chlorinated methane degradation is typically a cometabolic reductive dechlorination process (Cappelletti et al 2012). Carbon sources include volatile fatty

acids, methanol, acetate, lactate, fructose, and glucose. Chloroform cometabolic reductive dechlorination is often incomplete, resulting in accumulation of degradation products such as dichloromethane. Anaerobic chlorinated methane degradation can occur through dehalorespiration. Lee et al (2012) reported the complete degradation of chloroform via dehalorespiration by *Dehalobacter* bacteria.

Abiotic chemical reduction may be carried out using micro and nano scale zero valent metals. Zero valent metal provides electrons for degradation (Lien 2005). Common metals used in this technique include iron, aluminium, and zinc (Feng and Lim 2005, Lien 2005). Since chlorine atoms are removed from organic molecules and replaced by hydrogen, the process is pH dependent (Lien 2005). Reactivity or reducibility of the zero valent metal also depends on pH since precipitates often form under basic conditions. Noble or catalytic metals such as palladium, nickel, platinum, and silver combined with zero valent iron can increase reaction rate (Xiaoqin et al 2006).

Chemical oxidation can be used to treat various chlorinated organic compounds through redox reactions with chlorinated contaminants as electron donors and various chemical amendments as electron acceptors (Huling and Pivetz 2006). Dichloromethane has good reactivity with Fenton's reagent, activated persulfate, ozone, and perozone. Chloroform only has good reactivity with persulfate. Chloroform and dichloromethane are both poorly reactive with permanganate. Chemical hydrolysis treatment removes chloride from chlorinated organic compounds through reactions with water or hydroxide ions (Torrento et al 2014). Chemical hydrolysis under alkaline conditions is markedly faster than under neutral conditions.

Chlorinated organic compounds can be physically remediated by air stripping. In this method, ground water is pumped out and particulates are filtered out (Flathman 1992). In a holding tank, free phase chlorinated organic compounds sink. Dissolved chlorinated organic compounds are removed by heating the ground water and passing it through a column in the opposite direction to a flow of air. Treatment cost increases if vapour phase treatment is required to reduce air emissions. In the gas phase, chlorinated organic compounds can be degraded using photolysis. Alapi and Dombi (2007) carried out photo oxidation of carbon tetrachloride, chloroform, and dichloromethane using 184.9 nm radiation to break the carbon chlorine bonds.

Activated carbon can be used to sorb chlorinated organic compounds in ground water. Feed stock used in the production of activated carbon includes wood, coal, peat, lignite, and agricultural by products such as coconut, pecan, and almond shells (Bansode et al 2003). Methods of activation include injecting steam or carbon dioxide at high temperature and soaking

in phosphoric acid. Sorption is greater when materials with greater surface areas, such as activated carbon fibres, are used (Tsai et al 2008). The sorption capacity of the activated carbon and the cost of replacement and disposal must be considered (Flathman et al 1992).

Electro kinetic treatment may be used to remediate soils contaminated with chlorinated hydrocarbons. Chlorinated methanes are not well studied relative to chlorinated phenols because phenols are more polar (Gomes et al 2012). Enhanced electro kinetic treatments use amendments such as surfactants and cyclodextrins or pH control to increase mobility of chlorinated organic compounds.

Ultrasound energy has been used for remediation of chlorinated organic compounds. Ultrasound causes the formation of micro scale bubbles which create high pressure and temperature when they collapse (Adewuyi 2001). Thangavadivel et al (2010) used low frequency ultrasound at 20 kHz to mineralize approximately 8 mg L⁻¹ chloroform within one hour from ground water in batch and flow cell treatments.

4. RESEARCH SITE

Research was conducted at the University of Alberta's former Ellerslie Waste Management Facility at the Ellerslie Research Station in Edmonton, Alberta (Appendix). The approximately 0.25 ha site is located within the Province of Alberta's Edmonton Transportation Utility Corridor (Alberta Infrastructure 2003). Surroundings are zoned for agricultural purposes (AMEC Earth and Environmental 2010). A major road is located immediately north and residential areas are north and east. Whitemud Creek is located approximately 450 m north. Surficial geology has been classified mainly as fine textured clay and clay till (Stantec Consulting Limited 2013). Some sand has been identified in samples 11.5 to 12.5 below ground (AMEC Earth and Environmental 2010). The extent and connectivity of this potential sand unit is unknown. The site is located in the Central Parkland natural subregion (Natural Regions Committee 2006).

The Ellerslie facility operated from 1972 to 2007 to process laboratory waste from the University of Alberta and from outside sources (Stantec Consulting Limited 2013). Activities included temporary waste storage, acid dilution and neutralization, and solvent incineration. A waste water pond existed on site until 1986. A University of Alberta report (1984) indicated that the polyvinyl chloride membrane in the pond cracked sometime prior to 1983 and the pond leaked. The pond was decommissioned; however, soil and ground water contamination with organic and inorganic substances persisted.

The Ellerslie Waste Management Facility was decommissioned in 2007 and all buildings were removed by 2012 (Stantec Consulting Limited 2010). The site is currently used for land reclamation research by the University of Alberta in partnership with the Government of Alberta. The land, originally leased from the Province of Alberta in 1961, is to be returned in a condition similar to that present before the land was leased.

5. RESEARCH APPROACH

Soil remediation research has mainly focussed on soil that is not fine textured. Conventional in situ physical and chemical oxidation techniques can be employed in fine textured soils; however, transport of contaminants and amendments may be significantly restricted in such low permeability substrates. This would strongly impact remediation effectiveness. Conventional bioremediation often requires an aerated environment which is difficult, if not impossible, to consistently achieve in fine textured soils. Understanding of remediation in fine textured soils with multiple contaminants is particularly limited. Contaminant mixtures complicate remediation because each contaminant has unique properties which affects its toxicity, movement, and degradation. Any remediation technique selected must be effective in treating its target contaminant(s) in the presence of others. Remediation plans including multiple techniques which can be combined concurrently or sequentially may be required. The former Ellerslie Waste Management Facility site provides an ideal opportunity to conduct research which addresses the issues of fine textured soils and contaminant mixtures. The Ellerslie site will serve as a case study for this research, which can then be extrapolated and expanded to other similar sites.

The research will address multiple steps of remediation, from characterizing the site to assessing strategies for its remediation, which will contribute to the long term management and reclamation plan for the site. Objectives are to understand contaminant dynamics in fine textured soils through assessment of temporal and spatial trends, characterize the microbial communities on site, and assess the potential for physical, chemical, and biological remediation treatments. Detailed objectives are provided for each of the studies in their respective chapters. Although a variety of soil and ground water contaminants are present, the research focuses on chlorinated organic compounds and salts. These contaminants were selected because they were found to have high concentrations and frequency of detection in previous Ellerslie site reports. They are also common contaminants at other sites.

Figure 1.1 illustrates the context of the research and relationships among studies in a flow chart. The Ellerslie site has naturally occurring slightly saline soil due to its parent material. Anthropogenic activity has drastically changed site conditions. The waste water pond leak during site operation caused a marked increase in salt concentrations and introduction of chlorinated organic compounds into the soil and ground water. The resulting contaminated site needs to be addressed for the protection of human health and the environment, regulatory compliance, and the return of the land to the province.

The first step in the research program is a thorough site investigation to obtain detailed knowledge of site conditions to be used for the development of effective remediation and management strategies. Chapter II will characterize the Ellerslie soil and ground water to provide information on contaminant concentrations and their spatial and temporal trends, contributing to our understanding of basic processes on site.

The second step in the research program is to research active and passive methods of remediation. Chapter III will investigate active biostimulation and bioaugmentation. The use of microorganisms to degrade chlorinated organic compounds has been demonstrated in the literature and is a promising low cost option. Chapter IV will characterize the microbial communities in background and contaminated locations. It will provide information on passive natural attenuation of chlorinated organic compounds, and will contribute to our understanding of native microbial population dynamics under contaminated conditions. Chapter V will investigate active chemical reduction using zero valent iron. This remediation technique is promising in the literature and will be tested using soil from the research site. Chapter VI will investigate active salt leaching treatment. It will provide information on the applicability of leaching in a fine textured soil setting.

Chapter VII will provide an overview of results including applications and limitations. Potential future research and remediation plans for the Ellerslie site will be discussed. Overall, this research will provide critical information for development of an effective plan for the site which will result in minimization of risk, regulatory compliance, and return of the site to the province.

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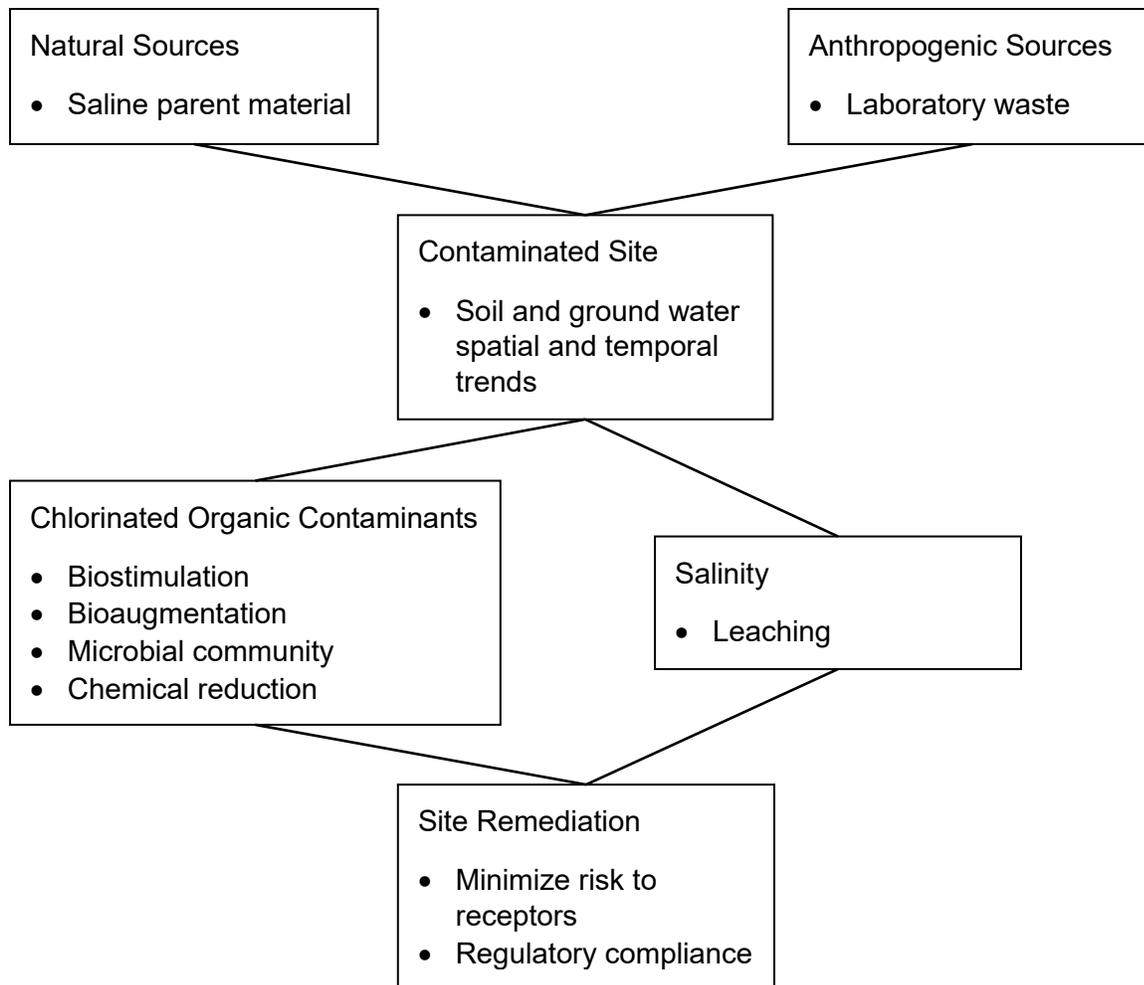


Figure 1.1. Research components including context, experiments, and implications.

II. SPATIAL AND TEMPORAL DYNAMICS OF INORGANIC AND ORGANIC CONTAMINANTS IN FINE TEXTURED SOIL AND GROUND WATER OVER 30 YEARS

1. INTRODUCTION

Environmental contamination is a global issue affecting land, water, and air. Numerous substances can be considered contaminants including solvents, petroleum products, volatile organic compounds, and trace elements (Singh et al 2009). Contaminants can be naturally occurring or of anthropogenic origin. Soil salinity can arise from weathering of parent material containing salts, irrigation with salt bearing water, and oil and gas production (Allison et al 1954, Alberta Environment 2001, Jordan et al 2004, Qadir and Oster 2004). Chloroform sources include natural soil and oceanic processes, chemical production and use, paper bleaching, and water treatment (Canadian Council of Ministers of the Environment 1999, McCulloch 2003, World Health Organization 2004, Huang et al 2014). Contaminants can cause a wide variety of negative effects to human and environmental health. High soil salinity increases the osmotic pressure with which water is held in soil and hinders plant access to water (Corwin 2003, Qadir and Oster 2004). The chlorinated solvents chloroform and dichloromethane cause human health effects such as cardiovascular, liver, and kidney damage (World Health Organization 2004).

Knowledge of contaminants and the environment in which they are found is critical to development of management and remediation plans for contaminated sites. Important contaminant information includes type and concentration. Sites are often impacted by multiple contaminants. Of 157 federal sites in Canada impacted by halogenated hydrocarbons, many contain contaminant mixtures (Treasury Board of Canada Secretariat 2019). This complicates remediation as each contaminant has unique properties which affect its toxicity, movement, and degradation. Any remediation technique selected must be effective in treating its target contaminant(s) in the presence of others. Contaminant concentration can impact the effectiveness of remediation techniques. Chlorinated organic compounds may be biodegraded cometabolically at lower concentrations but often have toxic effects on soil microorganisms at higher concentrations (Cappelletti et al 2012). Important site information includes soil type, geology, hydrology, topography, climate, land use, and location. Fine textured soils generally have low hydraulic conductivities and greater specific surface areas on which contaminants may be adsorbed (Duraes et al 2018). Therefore, contaminant movement is typically restricted in clay soils relative to sands which is beneficial in preventing contaminant spread. Clay soils can also restrict movement and incorporation of amendments introduced for the purpose of

remediation. Detailed contaminated site information can be determined through site characterization and monitoring. Long term monitoring in particular allows for the identification of spatial and temporal contaminant trends. Understanding trends is useful in making management and remediation decisions. For example, if contaminants are spreading towards a surface water body, rapid remediation may be necessary. If contaminant concentrations are decreasing across the site, natural attenuation may be sufficient for remediation.

The overall objective of this research was to identify soil and ground water contaminants at a site with fine textured soil and describe their spatial and temporal trends. Long term temporal trends in organic and inorganic contamination of ground water were determined. Spatial trends in organic and inorganic contamination of soil and ground water were assessed and compared. This study will be used to guide remediation of the research site and can be applied to numerous similar sites with fine textured soils and multiple contaminants.

2. MATERIALS AND METHODS

2.1. Research Site

Research was conducted at the University of Alberta's former Ellerslie Waste Management Facility in Edmonton, Alberta (Appendix). The approximately 0.25 ha site is located within the Province of Alberta's Edmonton Transportation Utility Corridor with surroundings zoned for agricultural purposes (Alberta Infrastructure 2003, AMEC Earth and Environmental 2010). A major road is located immediately north and residential areas are north and east. Whitemud Creek is located approximately 450 m north. Surficial geology has been classified mainly as fine textured clay and clay till (Stantec Consulting Limited 2013). Some sand has been identified in samples approximately 11.5 to 12.5 below ground (AMEC Earth and Environmental 2010). The site is located in the Central Parkland natural subregion (Natural Regions Committee 2006).

The Ellerslie facility operated from 1972 to 2007 to process laboratory waste from the university and outside sources (Stantec Consulting Limited 2013). Activities included temporary waste storage, acid dilution and neutralization, and solvent incineration. Site layout changed over time; at the end of operation, infrastructure included buildings for acid neutralization (Figure 2.1), vial crushing and drum storage, a storage tank, radioactive waste storage, chemical waste storage, and two 10,000 L above ground solvent storage tanks. A waste water pond existed until 1986. A University of Alberta report (1984) indicated that its polyvinyl chloride membrane cracked prior

to 1983 and the pond leaked. The pond was decommissioned although soil and ground water contamination with organic and inorganic substances persisted.

The facility was decommissioned in 2007 and all buildings were removed by 2012 (Stantec Consulting Limited 2010). The site is currently used for land reclamation research by the University of Alberta in partnership with the Government of Alberta. The land, originally leased from the Province of Alberta in 1961, will need to be returned in a condition similar to that present before the land was leased.

2.2. Soil And Ground Water Monitoring

Soil data were collected from direct push drilling programs at the Ellerslie site in 2009/2010 and 2014. All analyses were conducted at commercial laboratories. In late 2009 and early 2010, 42 bore holes were drilled evenly across the site to a depth of 10 m (Figure 2.2). Representative samples were collected every metre and analyzed for chloride, electrical conductivity, pH, total organic carbon, and volatile organic compounds (Table 2.1). In 2014, 50 bore holes were drilled, focusing on the former waste water pond and site boundaries (Figure 2.3). Two bore holes, 14-05 and 14-06, were drilled to a depth of 14 m, the rest to 10 m. Representative samples were collected every metre. Samples from all depths of a subset of bore holes across the site were analyzed for salinity parameters and particle size distribution for a detailed site description. Samples from all depths of a subset of bore holes most likely to have been impacted by site operations were analyzed for trace elements. Bore holes SST09-1 and STG09-1 from the 2009/2010 drilling program were also analyzed for trace elements. For each bore hole, samples from the depths with the highest field organic vapour concentrations, measured using an RKI Eagle portable gas detector, were analyzed for volatile organic compounds.

Ground water data were collected from annual monitoring programs since 1988. At the end of 2017, there were 42 slotted monitoring wells on site (Figure 2.4). The final two digits in each well name indicate its approximate depth. The oldest wells were installed in the 1980s during investigation of the pond leak. Additional wells were installed in 1991, 2001, 2003, 2006, 2008, 2009, 2010, 2014, 2016, and 2017.

Early ground water monitoring analyzed for salinity parameters, hydrocarbons, and radioisotopes (AMEC Earth and Environmental 2008). Volatile organic compounds, including chlorinated organic compounds, were added in 2004. Beginning in 2013, ground water monitoring was conducted according to a rationalized program (Stantec Consulting Limited 2012). In spring, water level was measured using a Rice water level meter for each well. Well

purging was completed using dedicated bailers or Waterra tubing for a subset of wells critical for site representation installed prior to 2014 and all wells installed in 2014 or later. Approximately one week after purging, wells were sampled and temperature, electrical conductivity, and pH were measured in the field with a portable meter. Wells installed prior to 2014 were analyzed for chloride, electrical conductivity, pH, uranium, volatile organic compounds, and radioisotopes (Table 2.2). Wells installed 2014 or later were analyzed for major ions, total dissolved solids, electrical conductivity, pH, dissolved trace elements, chemical oxygen demand, total organic carbon, volatile organic compounds, and radioisotopes. In fall, only wells installed in 2014 or later were monitored unless spring anomalies were identified for other wells. Water level and field measurements were conducted as in spring. Samples were analyzed the same as in spring except for dissolved trace elements, total organic carbon, and radioisotopes.

Duplicate samples were collected for approximately 10 % of samples for quality control. Samples for dissolved trace elements and mercury analysis were filtered and preserved with nitric acid and hydrochloric acid, respectively. Samples were preserved for chemical oxygen demand and total organic carbon analysis with sulfuric acid, for volatile organic compound analysis (except methanol) with sodium bisulfate. Samples were kept in coolers with ice until delivered to a commercial laboratory for analyses.

2.3. Data Analyses

Organic and inorganic soil and ground water contaminants were identified by comparing analytical results to Alberta Tier 1 soil and ground water remediation guidelines for fine textured soil and agricultural land use (Alberta Environment and Parks 2016) as clay and clay till surficial material dominated and the site is to be returned to the Province in pre-lease condition. Contaminant identification was completed for all soil data and spring ground water data for every even year of ground water data plus 2015 and 2017. Where a guideline was dependent on the value of a secondary parameter, the most stringent guideline value was used. Where an analytical detection limit exceeded the Alberta Tier 1 guideline, the value was not counted.

Temporal trends were assessed using the Mann Kendall test in R for ground water chloride, sodium, sulfate, total dissolved solids, electrical conductivity, pH, aluminum, arsenic, boron, cadmium, chromium, copper, iron, lead, manganese, mercury, nickel, selenium, silver, uranium, zinc, carbon-14, 1,1-dichloroethene, 1,2-dichloroethane, benzene, carbon tetrachloride, chloroform, dichloromethane, ethylbenzene, tetrachloroethene, and toluene. Each well was analyzed separately for each contaminant for which it had at least four data points using its

lifetime data. Only well 92-09-07 was analyzed for change in carbon tetrachloride concentration over time as it was the only well with values above detection limit. When a concentration was below laboratory detection limit, a value equal to the detection limit was used. Autocorrelation and partial autocorrelation were computed; block bootstrapping was used when serial correlation was significant (Ghement Statistical Consulting Company Limited 2014). A trend was considered statistically significant with $p < 0.05$ or when the 95 % confidence interval from bootstrapping did not contain zero. Statistically significant trends suspected to be a result of laboratory detection limits were identified where at least half the data were originally below detection limit and the detection limit changed over time and/or the detection limit values were distributed predominantly at one end of the time series so as to guide the trend.

Spatial trends for soil and ground water contaminants were assessed by mapping in two and three dimensions using ArcGIS. Ten contaminants, chloride, sodium, electrical conductivity, cadmium, nickel, uranium, benzene, carbon tetrachloride, chloroform, and dichloromethane, were selected for assessment to include a variety of salinity parameters, trace elements, and volatile organic compounds with high concentrations and/or widespread contamination. Where an analytical detection limit exceeded the Alberta Tier 1 guideline, the value was not used as there was no certainty of its actual value. No Alberta Tier 1 soil guideline exists for chloride or sodium so threshold values of 100 mg kg^{-1} and 160 mg kg^{-1} , respectively, were used (AGVISE Laboratories 2012, Millennium EMS Solutions Limited 2016).

Three dimensional soil maps were prepared in ArcScene. For each selected contaminant, colour was used to indicate contaminant concentration and spatial distribution was visually assessed. For electrical conductivity, a ribbon was drawn at approximately 2 m depth intervals showing the approximate extent of soil with electrical conductivity $> 5 \text{ dS m}^{-1}$, rated poor to unsuitable (Alberta Environment 2001). Polygons were stacked and a grid fit to estimate a volume of soil to be excavated for ex situ remediation. Electrical conductivity was selected for volume estimation since it had a large number of data points and widespread exceedances.

Two dimensional ground water maps were prepared in ArcMap. Maps were prepared using data from 1992, 2004, 2014, and 2017 to visually examine changes in contaminant concentrations and distributions over time. Water level was mapped using spline interpolation of depth values adjusted according to site topography. For groups of two or three wells located less than 2 m apart, mean water level and location were used. Contour lines were drawn and flow direction determined perpendicular to contour lines. For each contaminant, concentration was mapped using spline interpolation. For groups of two or three wells located less than 2 m apart, the well

with the highest contaminant concentration was used. Mann Kendall temporal trend results were indicated on the 2017 map for assessment of spatial distribution of temporal trends.

3. RESULTS

3.1. Contaminants

Alberta Tier 1 guidelines were exceeded for 18 soil parameters assessed (Table 2.3). Greatest number of exceedances were for electrical conductivity (335), dichloromethane (205), and chloroform (116). Greatest proportion of exceedances were for 1,2-dichloroethane, carbon tetrachloride, chloroform, and hexachlorobutadiene, with all values above guideline. Greatest maximum magnitude of exceedances relative to guideline were for dichloromethane (103,846 times guideline), chloroform (16,552 times guideline), and benzene (435 times guideline).

Alberta Tier 1 guidelines were exceeded for 37 ground water parameters (Figures 2.5 to 2.40). Greatest mean annual number of wells with exceedances from 2014 to 2017 were for electrical conductivity (27.8), uranium (27.5), and manganese (13.8) (Table 2.4). Greatest mean annual proportion of wells with exceedances from 2014 to 2017 were for sulfate (1.000), electrical conductivity (0.991), and uranium (0.983). Greatest maximum magnitude of exceedances relative to guideline from 2014 to 2017 were for dichloromethane (159,600 times guideline), chloroform (7,278 times guideline), and cadmium (4,750 times guideline). Nitrate nitrogen, nitrite nitrogen, sodium adsorption ratio, tritium, methanol, and total trihalomethanes exceedances were only identified prior to 2014.

3.2. Temporal Trends

Significant temporal trends in ground water chloride, sodium, sulfate, total dissolved solids, electrical conductivity, pH, arsenic, boron, cadmium, copper, iron, lead, manganese, mercury, nickel, selenium, uranium, zinc, carbon-14, benzene, chloroform, dichloromethane, and tetrachloroethene were identified with Mann Kendall analysis (Tables 2.5 to 2.7). Significant temporal trends for ground water chromium, 1,1-dichloroethene, and 1,2-dichloroethane were likely driven by values below detection limit. No significant temporal trends were identified for ground water aluminum, silver, carbon tetrachloride, ethylbenzene, or toluene. At least one significant temporal trend was found for all wells except 08-07-10, 14-01-08, 14-02-08, 14-05-12, 14-06-13, and 14-08-08.

The directions of significant temporal trends were generally not consistent within either contaminant or well. However, all significant trends for copper, lead, and dichloromethane were

decreasing, and both of the significant trends for carbon-14 were increasing. All significant trends not likely driven by values below detection limit were decreasing for wells 88-03-07, 03-08-06, and 09-10-08, and increasing for wells 01-02-05, 01-03-09, 01-04-05, 09-06-08, and 09-09-08 over the evaluated time period.

3.3. Spatial Trends

Soil contamination was generally spatially associated with the former waste water pond (Figures 2.41 to 2.51). Salinity parameters chloride, sodium, and electrical conductivity, were highest in the pond footprint and to the north and east. Moderately high sodium and electrical conductivity presented at depth across the site. Trace elements cadmium and nickel only had one Alberta Tier 1 exceedance each, while uranium had none. Cadmium and nickel exceedances were in the pond footprint. The organic contaminants dichloromethane and benzene were highest in the pond footprint and to the east and west. Highest chloroform values were spread across the site and carbon tetrachloride was only found above detection limit on the west side. In the pond area, soil contamination reached at least 10 m below surface. An estimated 12,580 m³ of soil had electrical conductivity > 5 dS m⁻¹. This includes soil with lower electrical conductivity closer to the surface, but not the additional volume that would need to be excavated to create sloped pit walls for worker safety.

Since 1988, ground water levels for wells containing water were 0.82 to 8.71 m below surface. In 1992, water level was 2.08 to 2.52 m below surface with a mean of 2.35 m below surface. In 2004, water level was 2.25 to 5.89 m below surface with a mean of 3.47 m below surface. In 2014, water level was 0.82 to 4.09 m below surface with a mean of 2.33 m below surface. In 2017, water level was 1.75 to 7.32 m below surface with a mean of 3.18 m below surface. In 1992 and 2014, ground water flow direction in the pond area was approximately north east (Figure 2.52). In 2004 and 2017 that flow direction was approximately north west. The general spatial pattern of ground water contamination is associated with the pond (Figures 2.53 to 2.62). Salinity parameters chloride, sodium, and electrical conductivity were highest in the pond footprint, extending approximately north east. Trace elements cadmium, nickel, and uranium had no clear spatial trends. The organic contaminants benzene, chloroform, and dichloromethane were highest in the pond area. Carbon tetrachloride was focused on the west side of the site. Visual assessment of the spatial distribution of temporal trends in ground water contamination did not yield clear results. Both significant increasing and decreasing trends were observed in the pond footprint and north in the direction of ground water flow. A cluster of wells with significantly increasing chloride concentrations were located north of the pond.

4. DISCUSSION

Of soil and ground water contaminants identified at the site, those of greatest concern may be selected based on frequency and magnitude of detection. Rankings based on frequency of detection must be considered in context of how frequency was determined. Total and mean exceedance counts and proportions of exceedances are dependent on number of samples analyzed and number of values removed with high detection limits. Not all samples were analyzed for all parameters in all years. For some parameters, a large number of values were removed because the detection limit was higher than the Alberta Tier 1 guideline. Therefore, contaminants with greatest total or mean exceedances are not the same as those with greatest proportions of exceedances. For example, soil carbon tetrachloride did not have a high enough total exceedance count to rank in the top three. However, all concentrations below detection limit had to be removed, meaning 100 % of remaining values exceeded the Alberta Tier 1 guideline. Dominant soil and ground water contaminants based on frequency and magnitude of detection include electrical conductivity, chloroform, and dichloromethane, which rank highly in more than one category. This is a good indication the parameters are of high concern.

Analysis of temporal trends in ground water contamination by Mann Kendall analysis yielded variable results depending on the contaminant and the well. The Mann Kendall test detects monotonic trends; therefore, if the concentration of a contaminant were to increase and then decrease over time, no significant trend would be detected. This was likely the case for chloride and electrical conductivity at well 88-03-07. Mann Kendall analysis was also impacted by the number of data points available and whether or not they were above analytical detection limit. Wells with few data and many values below detection limit were unlikely to have significant temporal trends. This was evident in the wells installed in 2014, which were monitored over a relatively short time and only had one significant temporal trend. The variable trends identified could be the result of heterogeneous contaminant distribution. The pond leak could have occurred from multiple points in the liner and pond decommissioning could have left discrete zones of contaminated soil. These would both result in pockets of contamination which could reach monitoring wells at various times or miss them altogether. Differing contaminant solubilities, and preferential flow through biopores or along infrastructure such as utility lines would also contribute to heterogeneous contaminant distribution and inconsistent trends. Mapping ground water temporal trends did not reveal any stark spatial patterns. Thus it is likely simpler and potentially more useful to evaluate contaminant changes based on maps of contaminant concentrations.

Analysis of spatial trends in soil and ground water showed an expected association between contamination and the pond which leaked. However, the source of carbon tetrachloride contamination in the west area of the site is unknown. The unexpectedly widespread chloroform contamination may be related to the decommissioning of the pond. Very little record of the decommissioning process exists, specifically the steps taken. It is likely that material excavated from the pond may have been sorted into contaminated and uncontaminated piles (Jobson 2019). If there was an error in sorting, the piles were mixed up, or the soil was not analyzed for all contaminants, soil containing chloroform could have been spread across the site in small patches. Chloroform may also have entered the soil from additional sources such as the solvent storage tanks.

Maximum depth of contamination is uncertain as the majority of bore holes only extended to 10 m below surface and some contamination was still detected at that depth. An insufficient amount of data is available to determine size and continuity of the sand unit found approximately 11.5 to 12.5 m below surface. If the sand forms a large, continuous unit and contaminants were to reach it, they would be expected to spread more rapidly through the highly permeable material. The spatial pattern of high soil sodium and electrical conductivity at depth across the site may indicate natural salinity, as the Malmo silty clay loam of the area is formed on slightly saline lacustrine parent material (Bowser et al 1962).

Changes in spatial distribution of ground water contamination over time are difficult to identify because interpolation used in mapping is dependent on number and location of sampling points which change over time. Ground water chloride and sodium may show some evidence of movement to the north with ground water flow. In the 2017 map for chloride, a group of wells with significantly increasing trends is found at the north end of the mapped chloride plume. In the maps for sodium, the centre of the plume appears to move north over time. Salts are highly soluble and mobile so they are expected to have spread further and more easily from the pond with ground water flow than the other contaminant present on the site. Transport of organic contaminants may be too restricted in the fine textured material to detect any of their potential movement through the soil.

Results of this research are important for guiding management and remediation of the site. Knowledge of contaminant types is critical in remediation technique selection. Salinity and chlorinated organic compounds identified together on site in soil and ground water make remediation difficult since most methods will not be simultaneously effective on both these types of contaminants. A remediation plan including multiple techniques which can be combined

concurrently or sequentially may be required. Analysis of contaminant temporal trends did not identify strong, consistent trends which could be indicative of rapid contaminant movement or their biodegradation. Therefore, natural attenuation is not likely to be adequate in a reasonable time frame and active remediation is warranted.

Analysis of spatial trends confirmed that most contaminants are associated with the pond area. Soil contaminant maps, especially the three dimensional electrical conductivity map, could be used to guide excavation for ex situ soil remediation. They could also be used to determine the area to which a remediation amendment such as zero valent iron should be added for in situ remediation of chlorinated organic compounds. Ground water contaminant maps could be used to inform installation of an interceptor trench or permeable reactive barrier, both of which should be located down gradient of the contamination with a large enough span to capture all contaminated ground water flowing towards them. Knowledge of spatial trends is important when evaluating risk to current or future land uses. Contamination does not appear to have reached the major road immediately north of the Ellerslie site. Contamination is located in areas where one pipeline has been installed and another is planned along with a light rail transit line. When the first pipeline was installed, soil was excavated along its route, clean soil was used as fill, a protective sheet was installed, and extra worker safety protocols were implemented.

Methods developed or explored from this research could be applied to numerous other contaminated sites. Although temporal trend analysis did not yield clear results for the Ellerslie site, it has great potential, when combined with spatial trend analysis to provide information essential for characterizing site contamination. Changes in contaminant concentrations and their distribution could be identified either in natural attenuation or active remediation scenarios. This would provide a good understanding of bioremediation potential and success.

5. CONCLUSIONS

Soil and ground water at the former Ellerslie Waste Management Facility were impacted by a variety of contaminants including salinity, trace elements, and organic compounds. Temporal trends in ground water contaminant concentrations were not consistent within parameter or well. Spatial trends in soil and ground water contaminant concentrations showed an association with the former waste water pond area. These results are important in developing a site management and remediation strategy and in understanding contaminant movement in similar types of disturbed sites.

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Table 2.1. Ellerslie soil analytical methods.

Parameter	Analytical Method	
	2009/2010	2014
Chloride	APHA 4110 B ion chromatography (APHA 2000)	APHA 4500 Cl ⁻ E automated ferricyanide colorimetry (APHA 2011a)
Calcium, Magnesium, Potassium, Sodium, Sulfur		CSSS 15 saturated paste (Miller and Curtin 2008)
Electrical Conductivity	CSSS 18.3 saturated paste (Janzen 1993)	CSSS 15.2 and 15.3 saturated paste (Miller and Curtin 2008)
Sodium Adsorption Ratio		CSSS 15.4.4 calculation (Miller and Curtin 2008)
PH	CSSS 16.3 calcium chloride (Hendershot et al 1993)	CSSS 15.2 and 15.3 saturated paste (Miller and Curtin 2008)
Mercury		EPA 200.2 nitric and hydrochloric acid extraction (USEPA 1994a), EPA 245.1 cold vapour atomic absorption spectrometry (USEPA 1994b)
Antimony, Arsenic, Barium, Beryllium, Cadmium, Chromium, Cobalt, Copper, Lead, Molybdenum, Nickel, Selenium, Silver, Thallium, Tin, Uranium, Vanadium, Zinc		EPA 200.2 nitric and hydrochloric acid extraction (USEPA 1994a), EPA 6020 A inductively coupled plasma mass spectrometry (USEPA 2007a)
Total Organic Carbon	LECO 203-821-170 combustion (LECO Corporation 2010)	
Volatile Organic Compounds	EPA 8260 C gas chromatography mass spectrometry (USEPA 2006)	EPA 8260 C gas chromatography mass spectrometry (USEPA 2006), CCME CWS PHC 1310 gas chromatography flame ionization detection (CCME 2001)

Table 2.2. Ellerslie ground water analytical methods.

Parameter	Analytical Method
Chloride, Nitrate (as nitrogen), Nitrite (as nitrogen), Sulfate	APHA 4110 B ion chromatography (APHA 2000)
Total Alkalinity (as calcium carbonate), Bicarbonate, Carbonate	APHA 2320 titration (APHA 2011b)
Hardness (as calcium carbonate), Ion Balance	APHA 1030 E calculation (APHA 2011c)
Total Dissolved Solids	APHA 2540 C dry at 180 °C (APHA 2011d)
Electrical Conductivity	APHA 2510 electrode (APHA 2011e)
PH	APHA 4500 H ⁺ electrometric (APHA 2011f)
Aluminum, Antimony, Arsenic, Barium, Beryllium, Bismuth, Boron, Cadmium, Calcium, Chromium, Cobalt, Copper, Iron, Lead, Lithium, Magnesium, Manganese, Molybdenum, Nickel, Phosphorus, Potassium, Selenium, Silicon, Silver, Sodium, Strontium, Thallium, Tin, Titanium, Uranium, Vanadium, Zinc	APHA 3030 B and E filtration and nitric acid digestion (APHA 2004), EPA 6020 A inductively coupled plasma mass spectrometry (USEPA 2007a)
Mercury	EPA 245.7 cold water atomic fluorescence spectrometry (USEPA 2005), EPA 245.1 cold vapour atomic absorption spectrometry (USEPA 1994b)
Sulfur	APHA 3120 B inductively coupled plasma atomic emission spectrometry (APHA 2011g)
Chemical Oxygen Demand	APHA 5220 D closed reflux colorimetry (APHA 2011h)
Total Organic Carbon	APHA 5310 B high temperature combustion (APHA 2014)
1,1,1-Trichloroethane, 1,1,2,2-Tetrachloroethane, 1,1,2-Trichloroethane, 1,1-Dichloroethane, 1,1-Dichloroethene, 1,2,3-Trichloropropane, 1,2-Dibromoethane, 1,2-Dichlorobenzene, 1,2-Dichloroethane, 1,2-Dichloropropane, 1,3-Dichlorobenzene, 1,4-Dichlorobenzene, 2-Butanone, 2-Hexanone, 4-Methyl-2-Pentanone, Acetone, Acrolein, Acrylonitrile, Benzene, Bromodichloromethane, Bromoform, Bromomethane, Carbon Disulfide, Carbon Tetrachloride, Chlorobenzene, Chloroethane, Chloroform, Chloromethane, cis-1,2-Dichloroethene, cis-1,3-Dichloropropene, cis-1,4-Dichloro-2-Butene, Dibromochloromethane, Dibromomethane, Dichlorodifluoromethane, Dichloromethane, Ethanol, Ethyl Methacrylate, Ethylbenzene, Iodomethane, m+p-Xylenes, o-Xylene, Styrene, Tetrachloroethene, Toluene, trans-1,2-Dichloroethene, trans-1,3-Dichloropropene, trans-1,4-Dichloro-2-Butene, Trichloroethene, Trichlorofluoromethane, Vinyl Acetate, Vinyl Chloride	EPA 8260 C gas chromatography mass spectrometry (USEPA 2006)
2-Nitropropane, Cyclohexanone, Ethyl Acetate, Ethyl Ether, Methyl Ethyl Ketone, n-Butyl Alcohol, Pyridine, Xylenes	EPA 5021 equilibrium headspace (USEPA 2014), EPA 8260 C gas chromatography mass spectrometry (USEPA 2006)
Methanol	EPA 3550 C ultra sonic extraction (USEPA 2007b), EPA 8015 D gas chromatography flame ionization detection (USEPA 2003)

Table 2.3. Soil parameters in exceedance of Alberta Tier 1 guidelines.

Parameter	Guideline (mg kg ⁻¹)	Maximum (mg kg ⁻¹)	Exceedances	
			Number	Proportion Of Samples
Electrical Conductivity	3 dS m ⁻¹	23.1 dS m ⁻¹	335	0.580
PH	6.0 to 8.5	8.84	4	0.007
Cadmium	1.4	1.93	1	0.026
Copper	63	168	1	0.026
Nickel	45	207	1	0.026
Selenium	1.0	1.55	7	0.179
1,1-Dichloroethene	0.15	0.190	1	0.002
1,2-Dichloroethane	0.0062	0.219	3	1.000
Benzene	0.046	20.0	38	0.073
Carbon Tetrachloride	0.013	3.30	17	1.000
Chloroform	0.0029	48.0	116	1.000
Dichloromethane	0.052	5,400	205	0.470
Ethylbenzene	0.073	6.70	17	0.033
Hexachlorobutadiene	0.026	0.080	3	1.000
Tetrachloroethene	0.26	19.0	29	0.059
Toluene	0.52	41.0	7	0.013
Total Xylenes	0.99	14.0	8	0.019
Trichloroethene	0.054	0.220	8	0.016

Table 2.4. Ground water parameters in exceedance of Alberta Tier 1 guidelines 2014 to 2017.

Parameter	Guideline (mg L ⁻¹)	Maximum (mg L ⁻¹)	Mean Exceedances (year ⁻¹)	
			Number	Proportion Of Samples
Chloride	100	14,450	12.5	0.445
Sodium	200	7,840	8.5	0.599
Sulfate	128	3,240	9.5	1.000
Total Dissolved Solids	500	16,700	9.2	0.977
Electrical Conductivity	1.0 dS m ⁻¹	42.0 dS m ⁻¹	27.8	0.991
PH	6.5 to 8.5	6.380	0.5	0.193
Aluminum	0.007	0.256	5.3	0.503
Arsenic	0.005	0.190	1.3	0.099
Boron	1.0	1.30	0.3	0.008
Cadmium	0.00004	0.190	12.0	0.840
Chromium*	0.001	0.178	1.3	0.114
Copper	0.007	0.181	2.0	0.162
Iron	0.3	237	6.3	0.496
Lead	0.0009	0.141	1.0	0.091
Manganese	0.05	75.4	13.8	0.944
Mercury	0.000005	0.000112	2.3	0.273
Nickel	0.004	0.527	13.5	0.936
Selenium	0.001	0.182	5.8	0.338
Silver	0.0001	0.0034	1.0	0.077
Uranium	0.01	0.4198	27.5	0.983
Zinc	0.03	0.391	3.8	0.329
Carbon-14	200 Bq L ⁻¹	508 Bq L ⁻¹	1.8	0.061
1,1-Dichloroethene	0.014	0.089	1.3	0.045
1,2-Dichloroethane	0.005	0.128	3.8	0.135
Benzene	0.005	1.69	5.8	0.205
Carbon Tetrachloride	0.002	0.204	1.0	0.036
Chloroform	0.0018	13.1	7.8	0.278
Dichloromethane	0.05	7,980	7.3	0.258
Ethylbenzene	0.0016	0.0137	1.3	0.045
Tetrachloroethene	0.01	0.0416	1.5	0.054
Toluene	0.024	0.851	3.0	0.106

* Total chromium was measured but the guideline is for hexavalent chromium

Table 2.5. Ground water contaminant temporal trends for wells 88-02-07 to 03-08-06.

Parameter	Well													
	88-02-07	88-03-07	88-06-07	92-01-07	92-07-08	92-08-08	92-09-07	01-01-10	01-02-05	01-03-09	01-04-05	03-06-06	03-07-06	03-08-06
Chloride	SD	NS	SD	NS	SI	SI	NS	NS	NS	SI	NS	SI	SI	SD
Sodium	SD	NS	SI	SI	SD	NS	NS	NS	NS	NS	SI	SD	SD	SD
Sulfate	NS	NS	NS	SI	SD	SD	NS	NS	SI	NS	NS	NS	NS	NS
Total Dissolved Solids	SD	SD	SI	SI	SD	SD	NS	NS	NS	NS	SI	NS	SD	SD
Electrical Conductivity	SD	NS	SI	SI	SD	SD	NS	NS	NS	SI	NS	SD	SD	SD
PH	SI	NS	NS	SI	SI	SI	SI	SD	NS	NS	NS	NS	SD	SD
Aluminum	NS		NS	NS										
Arsenic	NS	NS	NS	NS	NS	SD	NS	NS	NS	NS	NS		NS	NS
Boron	NS		NS	NS										
Cadmium	SI	NS	NS	UD	NS		SD	NS						
Chromium	NS	UD	UD	UD	UD	UD	UD	NS	NS	NS	NS		NS	NS
Copper	SD	NS	NS	NS	NS		NS	NS						
Iron	NS													
Lead	NS	SD	SD	UD	UD	UD	UD	NS	NS	NS	NS		NS	NS
Manganese	SD	NS	NS	NS	NS	NS	SD	NS						
Mercury	UD	NS	UD	UD	NS	NS	UD	NS	NS	NS	NS		UD	NS
Nickel	SD	NS	SD	SI	SD	SD	SD	NS	NS	NS	NS		SD	NS
Selenium	NS		NS	NS										
Silver	NS		NS	NS										
Uranium	NS	NS	SI	NS										
Zinc	SD	SD	SD	SD	SD	NS	SD	NS	NS	NS	SI		NS	NS
Carbon-14	SI	UI	UI	UI	UI	UI	NS	NS	SI	NS	NS	UI	UI	NS
1,1-Dichloroethene	NS	UI	UI	NS	NS									
1,2-Dichloroethane	NS	UD	NS											
Benzene	SD	NS	UD	UD	NS									
Carbon Tetrachloride							NS							
Chloroform	NS	UD	NS											
Dichloromethane	NS	NS	NS	NS	NS	NS	UD	NS	NS	NS	UD	UD	NS	NS
Ethylbenzene	NS													
Tetrachloroethene	NS	UD	NS	SD										
Toluene	NS													

SI (orange), SD (blue), UI (light orange), UD (light blue), and NS indicate significant increase, significant decrease, uncertain increase, uncertain decrease, and no significant change, respectively.

Table 2.6. Ground water contaminant temporal trends for wells 03-09-06 to 09-06-08.

Parameter	Well												
	03-09-06	03-09-09	06-01-05	06-02-06	08-05-09	08-06-09	08-07-10	08-08-14	08-09-14	09-03-08	09-04-07	09-05-07	09-06-08
Chloride	SD	NS	SI	SI	NS	NS	NS	SD	NS	SI	NS	NS	NS
Sodium	NS	SD	NS	SI	NS								
Sulfate	NS	SD	NS	SD	NS	NS	SI						
Total Dissolved Solids	SD	NS	SD	SD	NS	SD	NS	NS	NS	NS	NS	SI	NS
Electrical Conductivity	SD	NS	NS	NS	SD	NS	NS	NS	SI	NS	NS	NS	NS
PH	NS	SD	SD	SD	NS	SD	NS						
Aluminum	NS												
Arsenic	NS	NS	SI	NS									
Boron										NS	NS	NS	NS
Cadmium	NS	NS	SD	NS									
Chromium	NS												
Copper	NS												
Iron	NS	SI	SI										
Lead	NS												
Manganese	NS	SD	NS	SI	SI	SI	NS						
Mercury	NS	SD	NS										
Nickel	SI	NS											
Selenium	NS	SI	NS										
Silver	NS												
Uranium	NS												
Zinc	NS												
Carbon-14	NS	NS		NS									
1,1-Dichloroethene	NS	NS				NS							
1,2-Dichloroethane	NS	NS				NS							
Benzene	NS	SI				NS							
Carbon Tetrachloride													
Chloroform		NS				NS	SI						
Dichloromethane	SD	NS				NS	NS	NS	NS	NS	SD	NS	NS
Ethylbenzene	NS	NS				NS							
Tetrachloroethene	NS	NS				NS							
Toluene	NS	NS				NS							

SI (orange), SD (blue), and NS indicate significant increase, significant decrease, and no significant change, respectively.

Table 2.7. Ground water contaminant temporal trends for wells 09-07-08 to 14-08-08.

Parameter	Well												
	09-07-08	09-08-09	09-09-08	09-10-08	09-11-09	10-01-08	10-02-09	14-01-08	14-02-08	14-05-12	14-06-13	14-07-08	14-08-08
Chloride	SD	NS	SI	NS	SD	NS	SI	NS	NS	NS	NS	NS	NS
Sodium	NS	NS	NS	SD	NS	NS	SD	NS	NS	NS	NS	NS	NS
Sulfate	NS	NS	NS	SD	NS								
Total Dissolved Solids	NS	SD	NS	SD	NS								
Electrical Conductivity	NS	NS	SI	NS	NS	NS	SI	NS	NS	NS	NS	SD	NS
PH	NS	NS	NS	NS	NS	NS	SI	NS	NS	NS	NS	NS	NS
Aluminum	NS		NS										
Arsenic	NS		NS										
Boron	NS		NS										
Cadmium	NS		NS										
Chromium	NS		NS										
Copper	NS		NS										
Iron	NS	NS	NS	NS	NS	SD	NS						
Lead	NS		NS										
Manganese	NS												
Mercury								NS	NS	NS	NS	NS	NS
Nickel	NS		NS										
Selenium	NS		NS										
Silver	NS		NS										
Uranium	NS		NS										
Zinc	NS		NS										
Carbon-14	NS												
1,1-Dichloroethene	NS		NS										
1,2-Dichloroethane	NS		NS	NS	NS	NS	UI	NS	NS	NS	NS	NS	NS
Benzene	NS		NS										
Carbon Tetrachloride													
Chloroform	NS		NS										
Dichloromethane	NS		NS										
Ethylbenzene	NS		NS										
Tetrachloroethene	NS		NS										
Toluene	NS		NS										

SI (orange), SD (blue), UI (light orange), and NS indicate significant increase, significant decrease, uncertain increase, uncertain decrease, and no significant change, respectively.

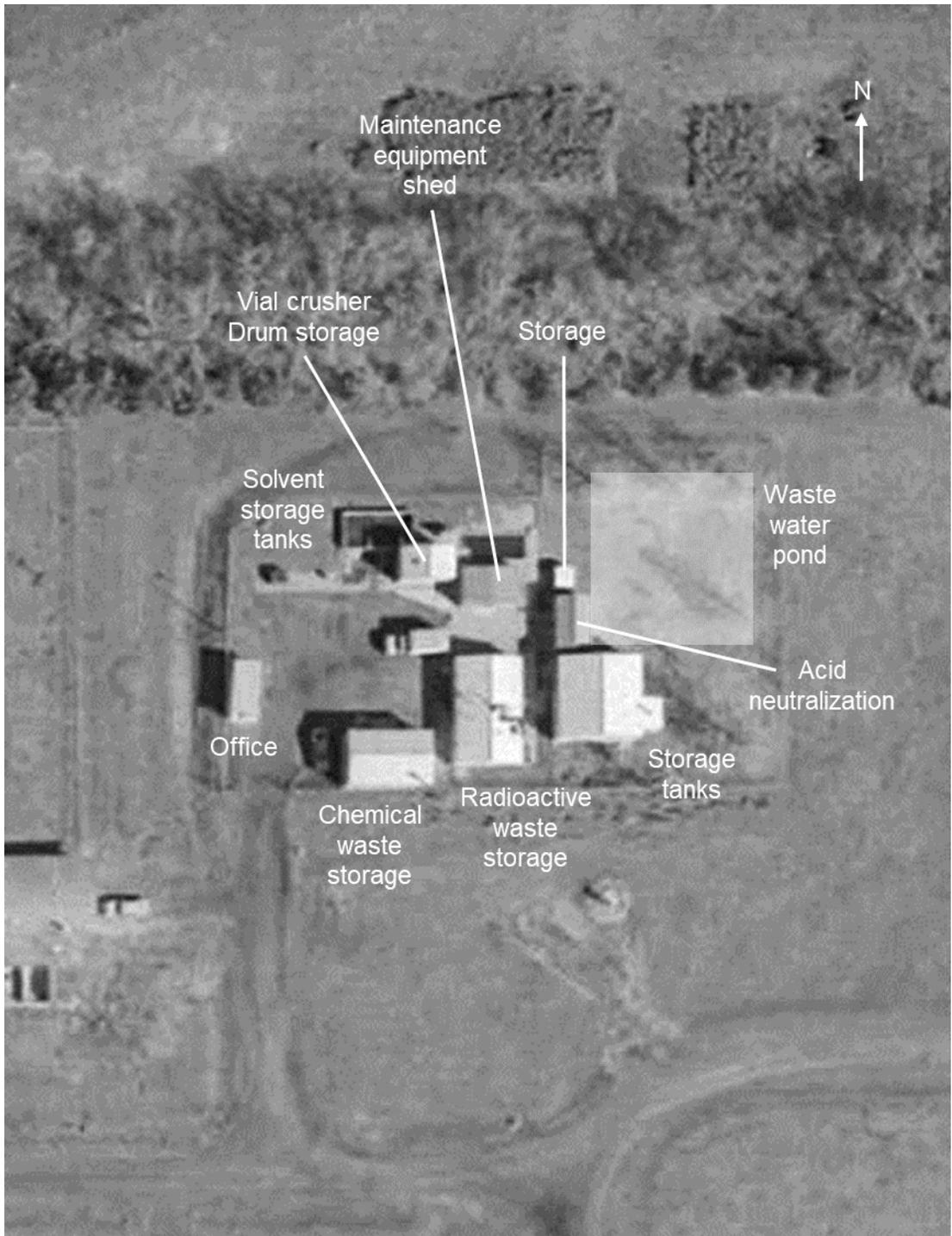


Figure 2.1. Former Ellerslie Waste Management Facility layout. Scale not available.

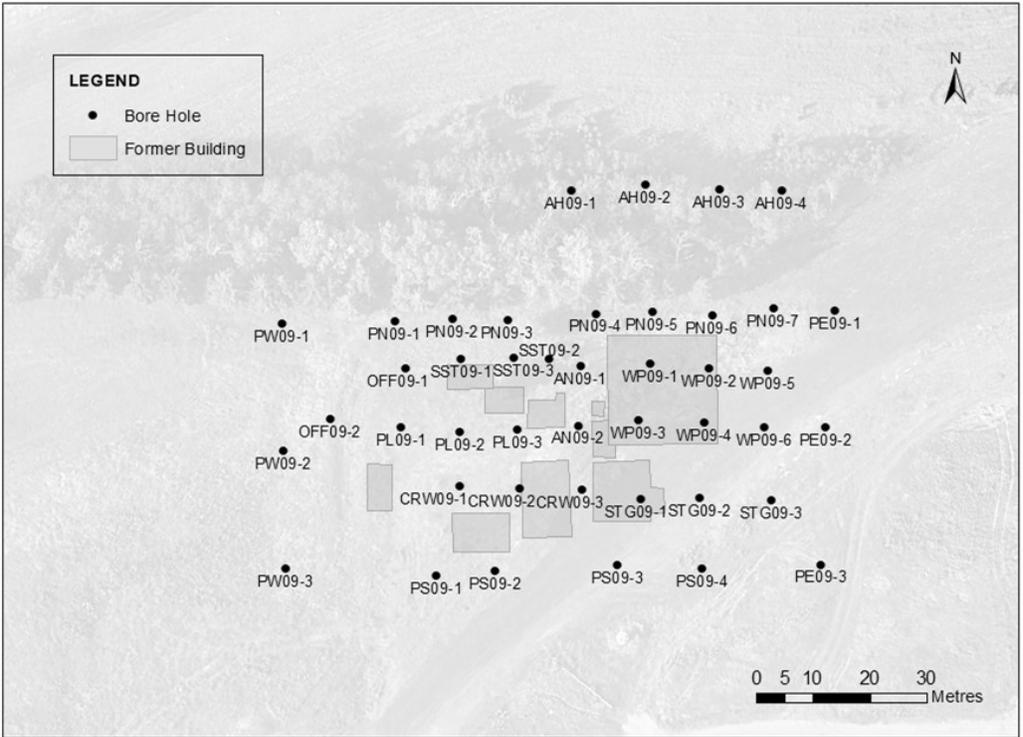


Figure 2.2. Ellerslie 2009/2010 soil sampling locations.

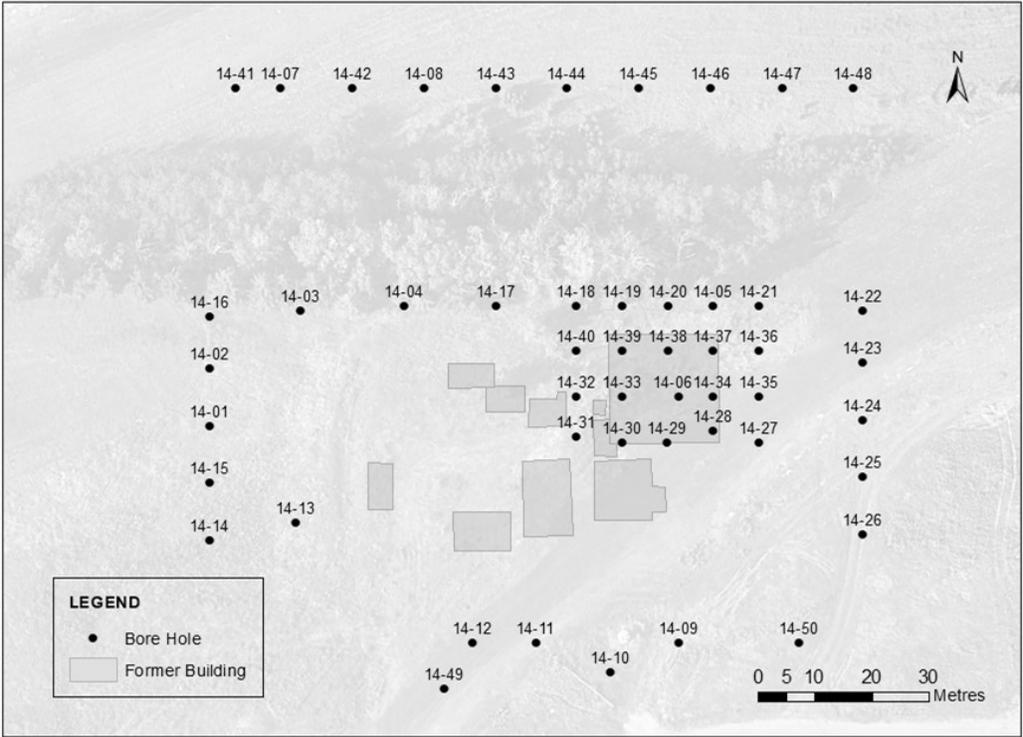


Figure 2.3. Ellerslie 2014 soil sampling locations.



Figure 2.4. Ellerslie monitoring well locations as of October 2017.

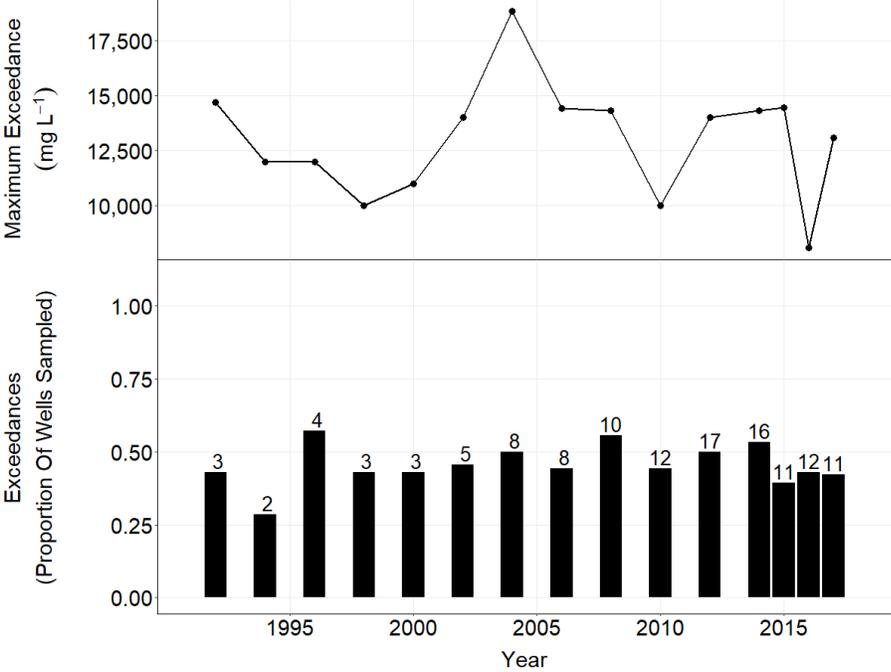


Figure 2.5. Ground water chloride exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

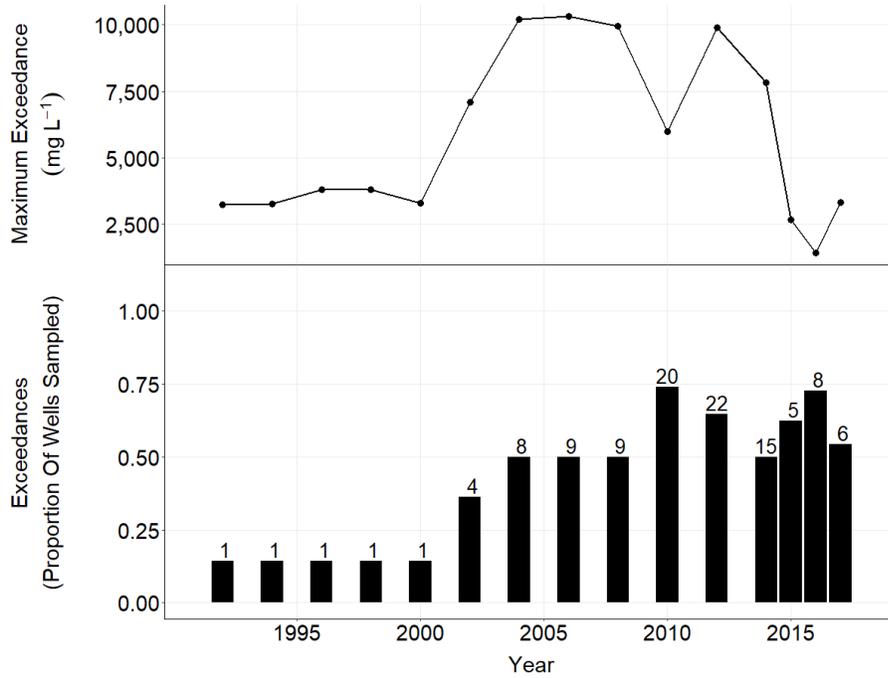


Figure 2.6. Ground water sodium exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

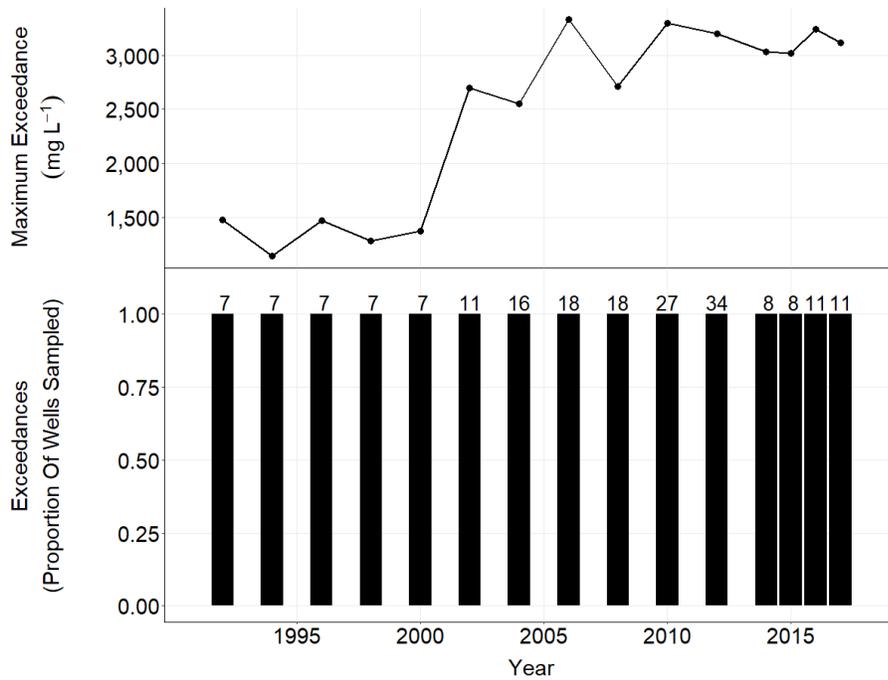


Figure 2.7. Ground water sulfate exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

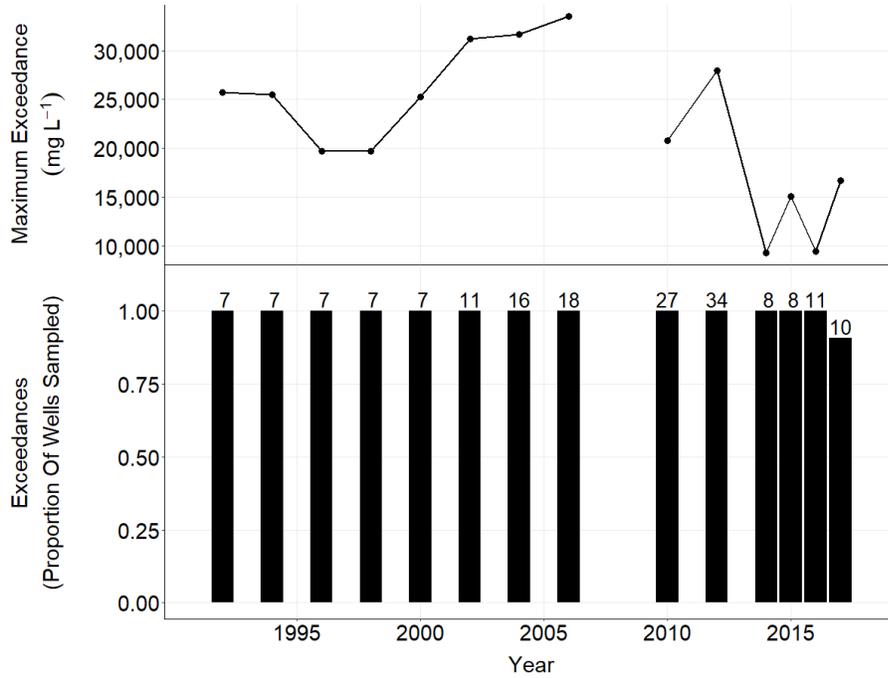


Figure 2.8. Ground water total dissolved solids exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

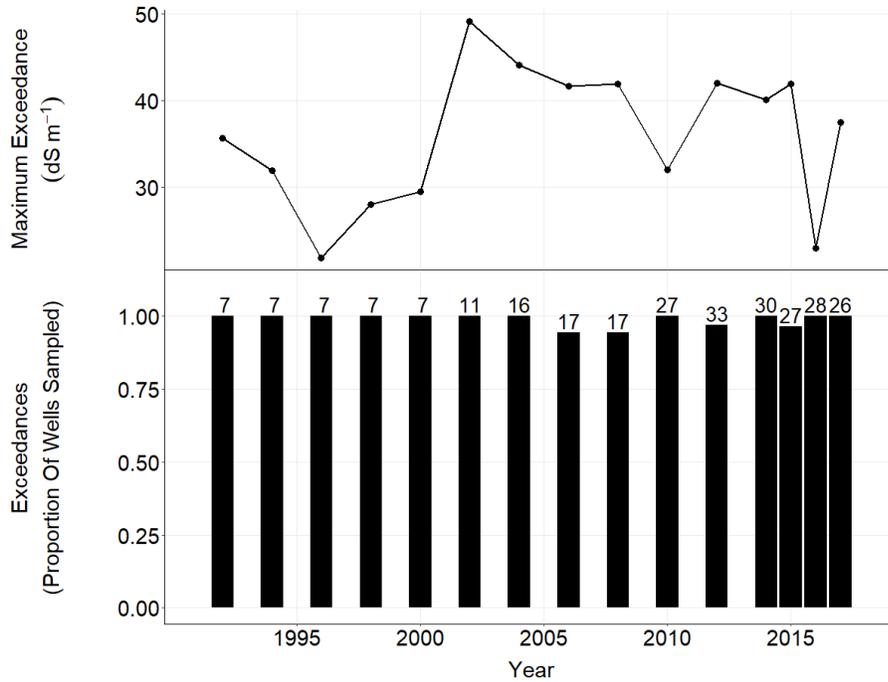


Figure 2.9. Ground water electrical conductivity exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

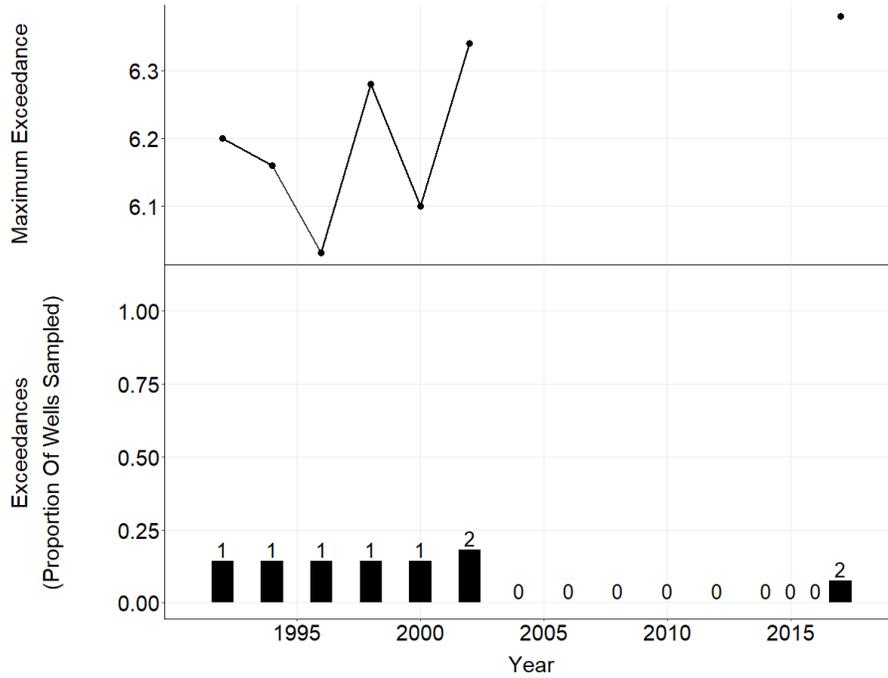


Figure 2.10. Ground water pH exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

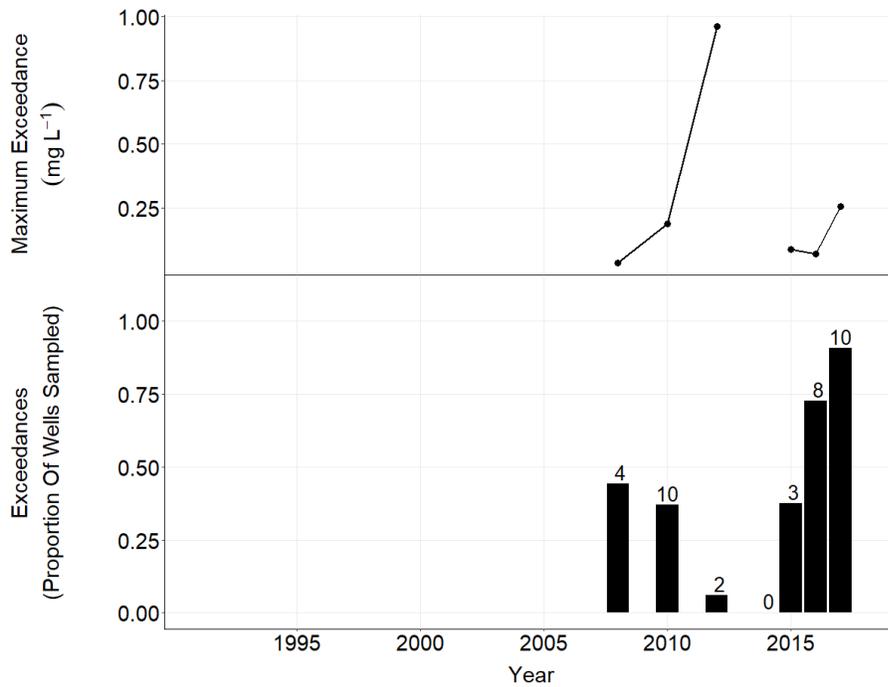


Figure 2.11. Ground water aluminum exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

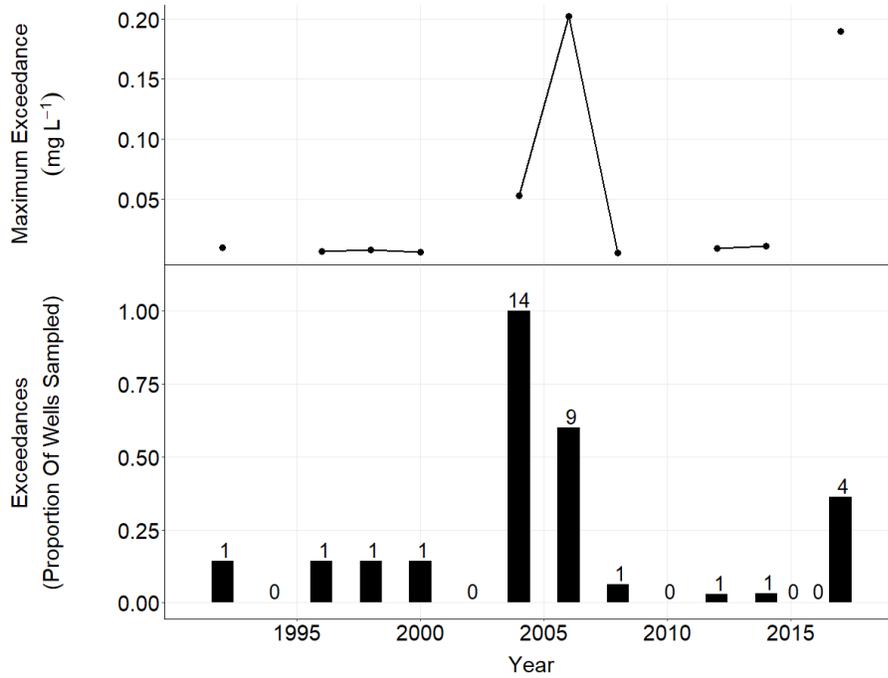


Figure 2.12. Ground water arsenic exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

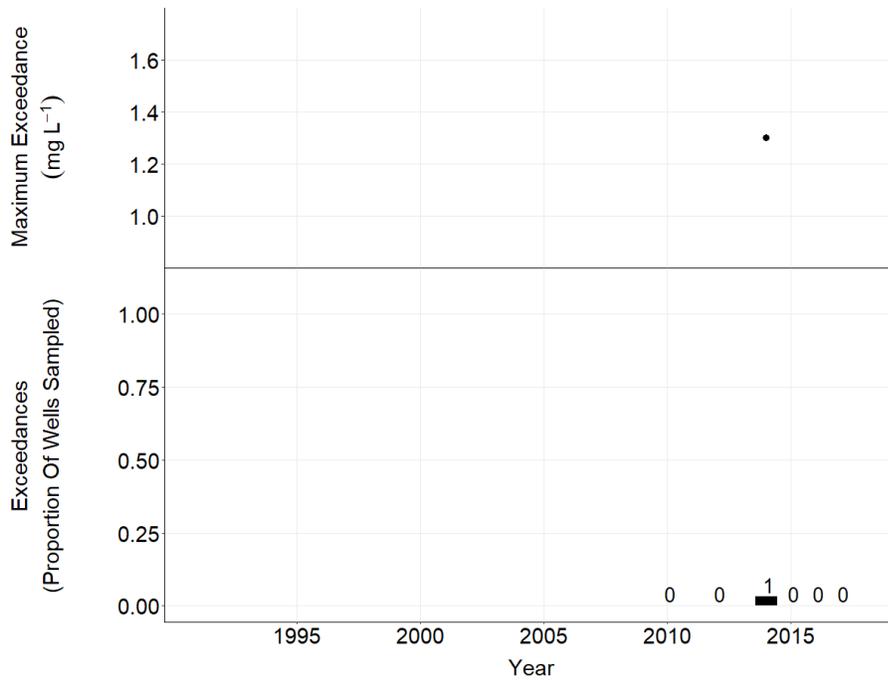


Figure 2.13. Ground water boron exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

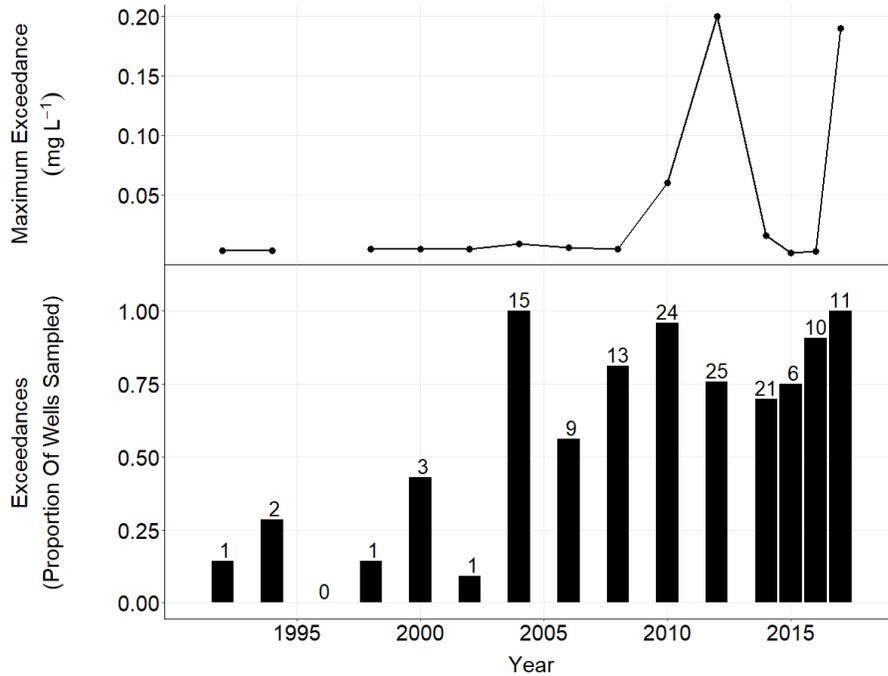


Figure 2.14. Ground water cadmium exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

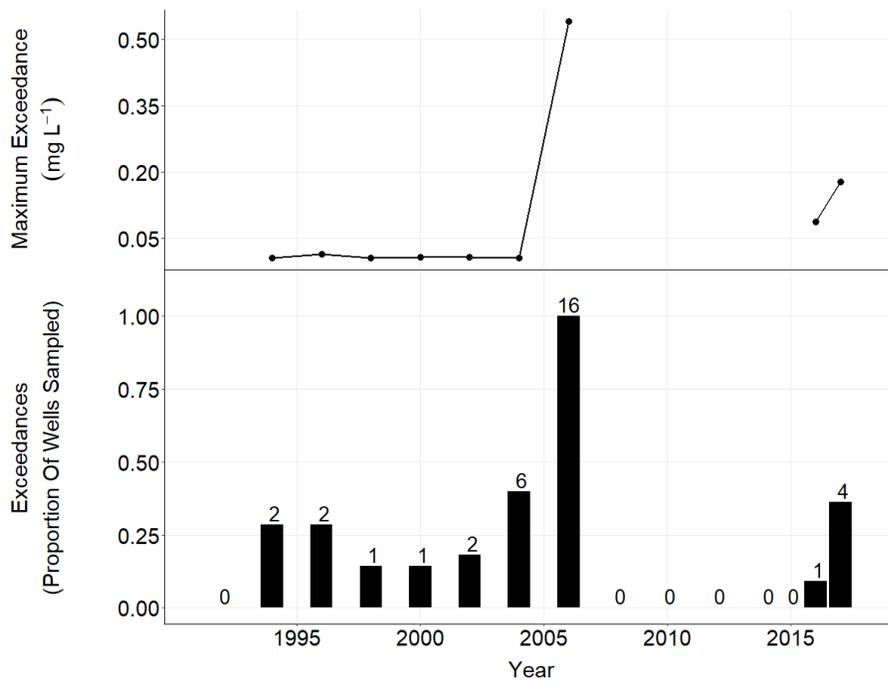


Figure 2.15. Ground water chromium exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

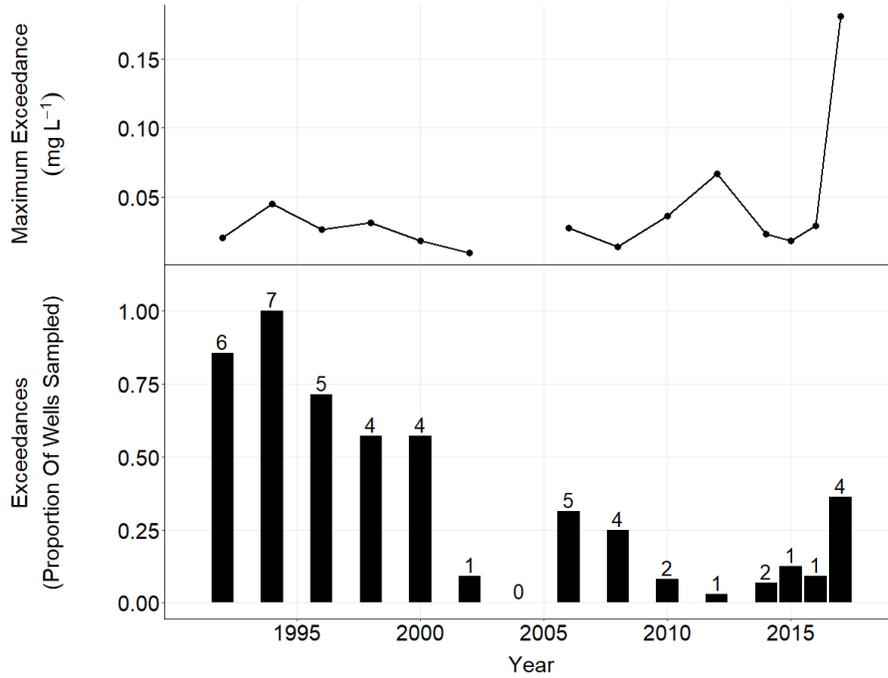


Figure 2.16. Ground water copper exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

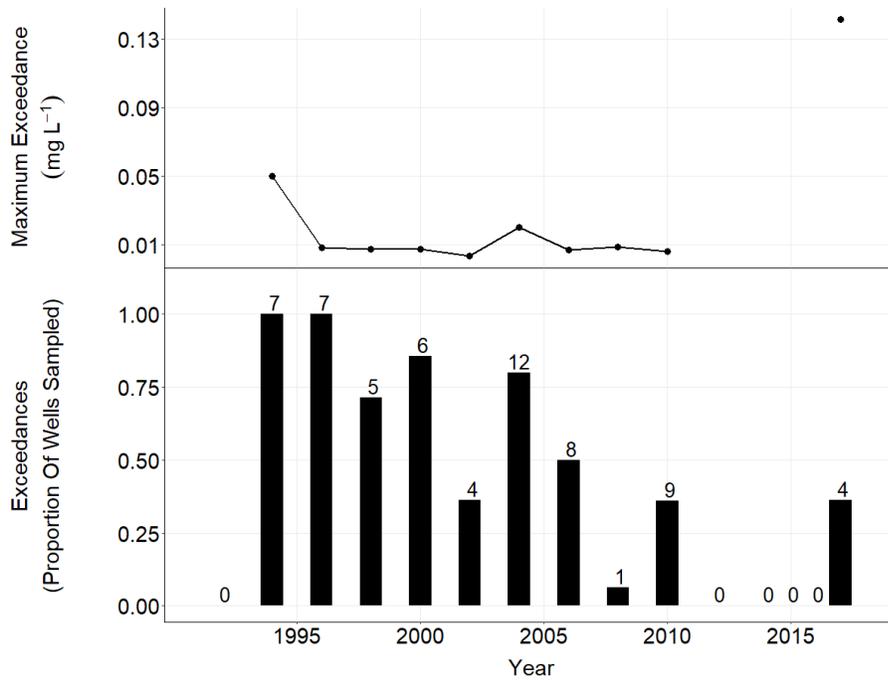


Figure 2.17. Ground water lead exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

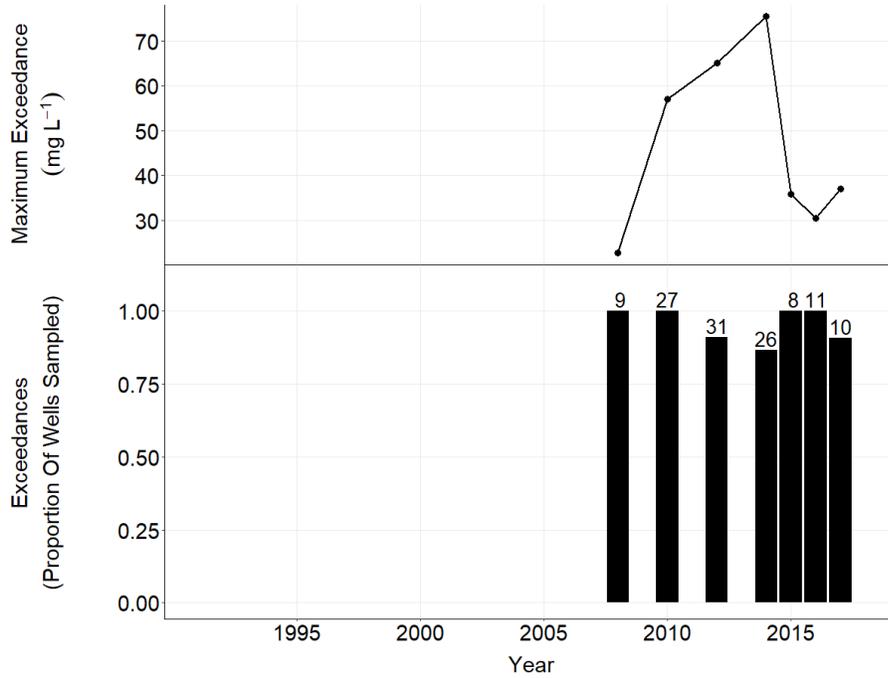


Figure 2.18. Ground water manganese exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

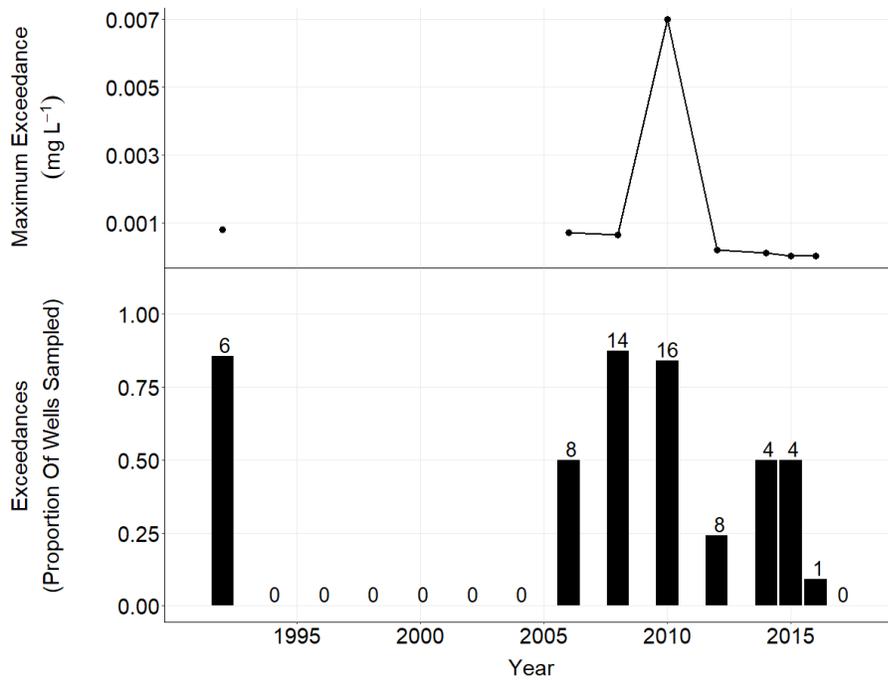


Figure 2.19. Ground water mercury exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

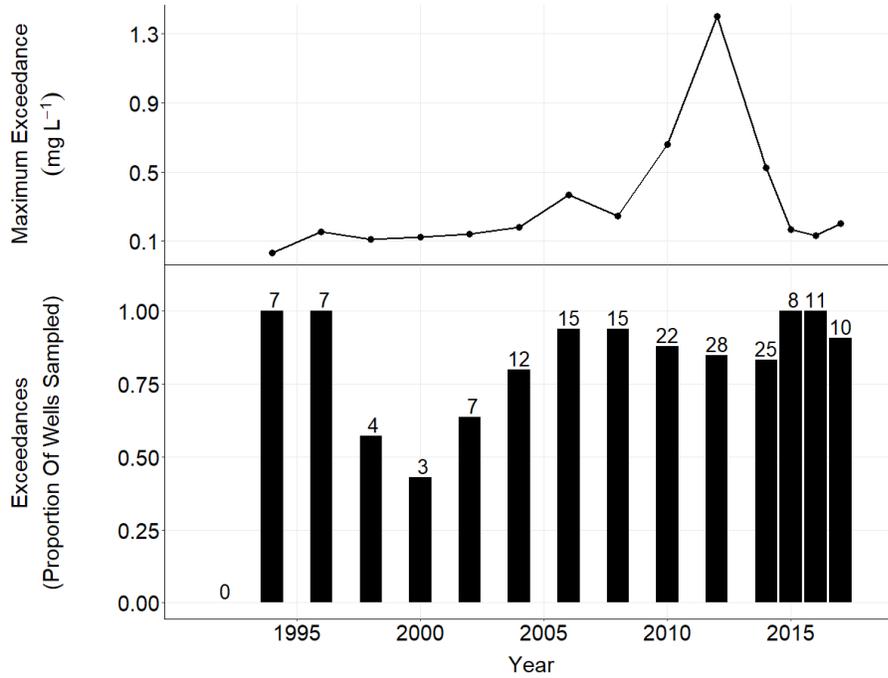


Figure 2.20. Ground water nickel exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

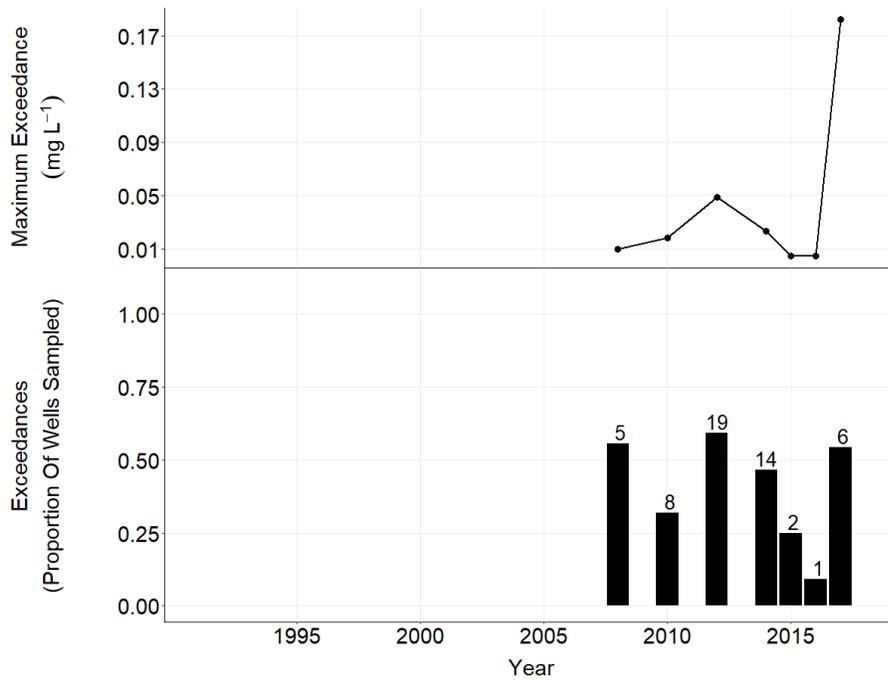


Figure 2.21. Ground water selenium exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

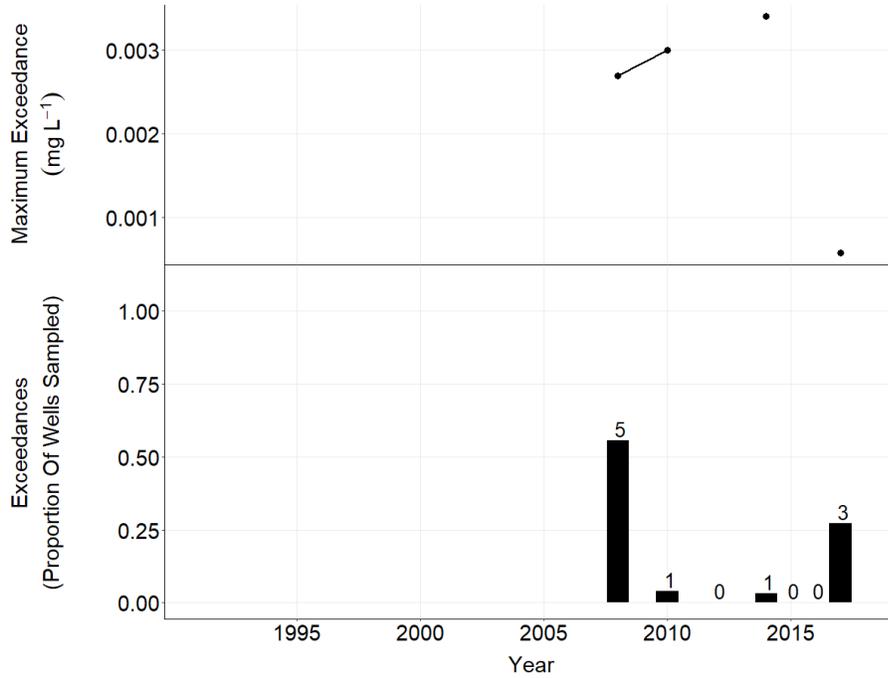


Figure 2.22. Ground water silver exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

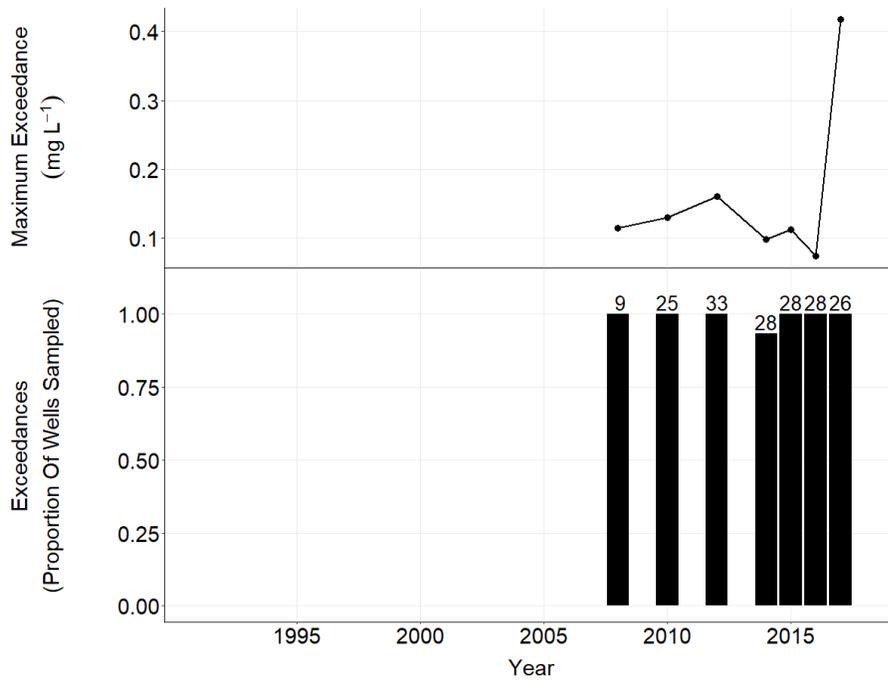


Figure 2.23. Ground water uranium exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

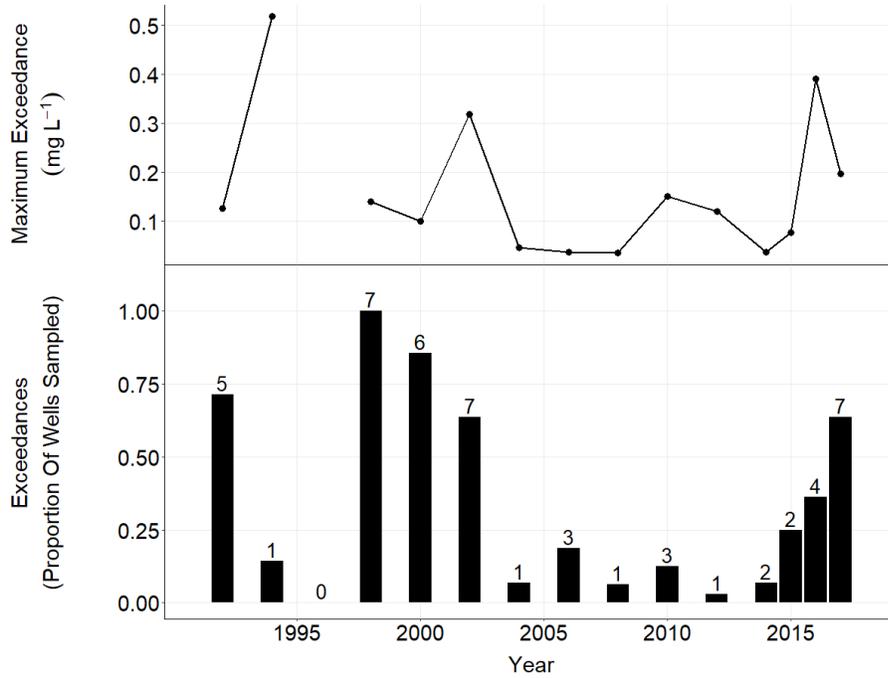


Figure 2.24. Ground water zinc exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

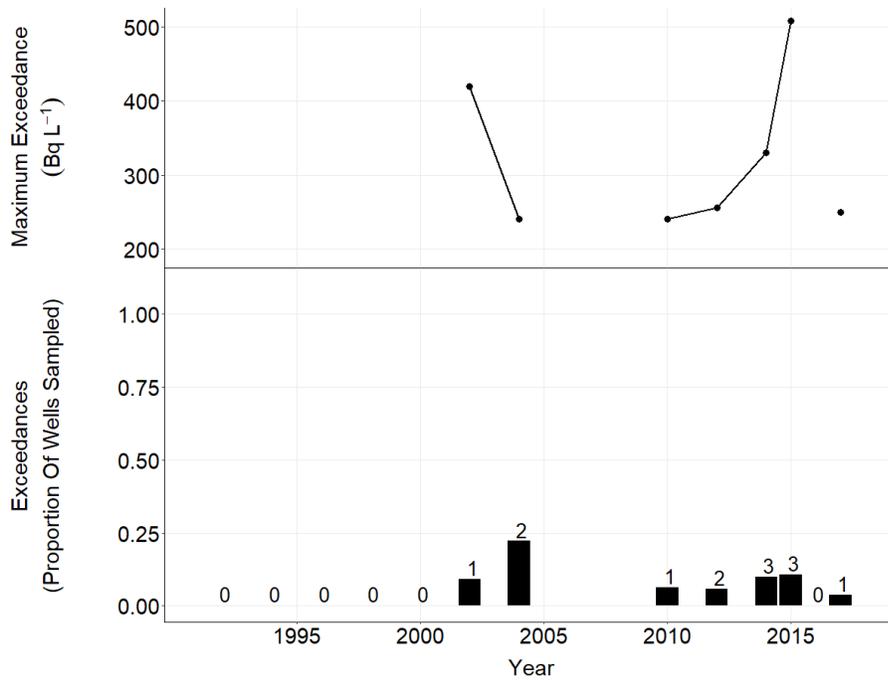


Figure 2.25. Ground water carbon-14 exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

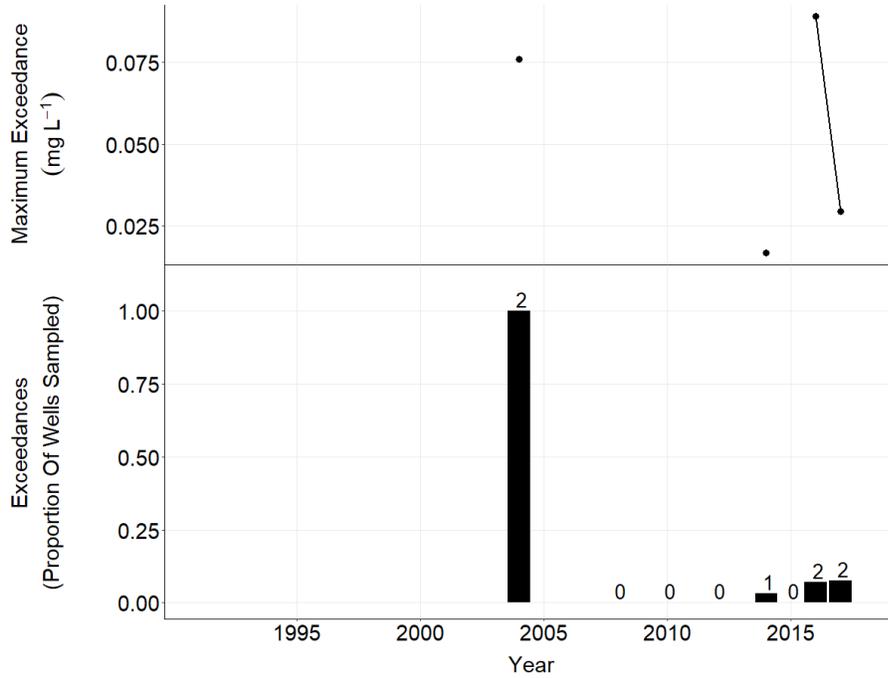


Figure 2.26. Ground water 1,1-dichloroethene exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

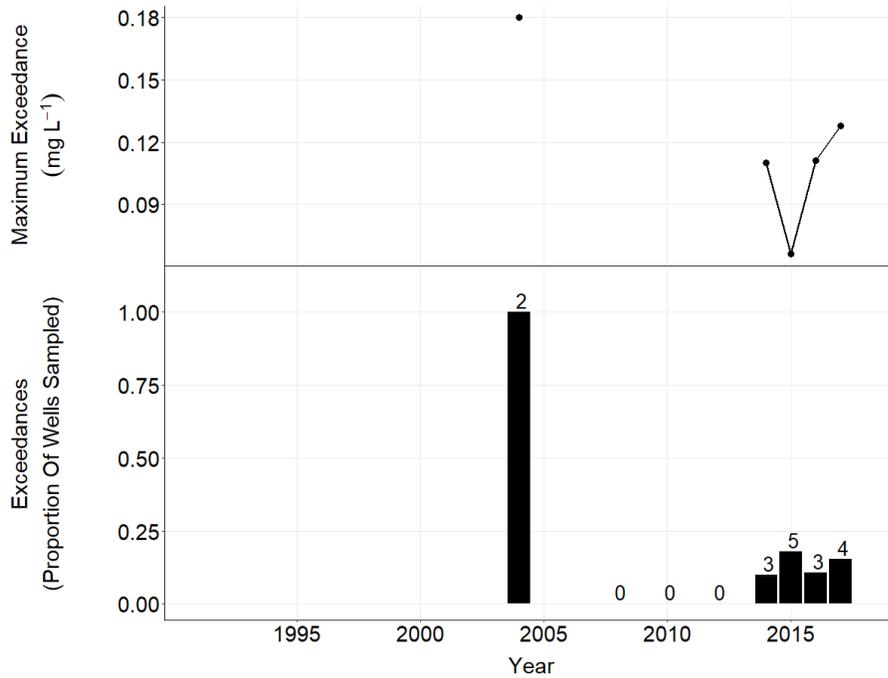


Figure 2.27. Ground water 1,2-dichloroethane exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

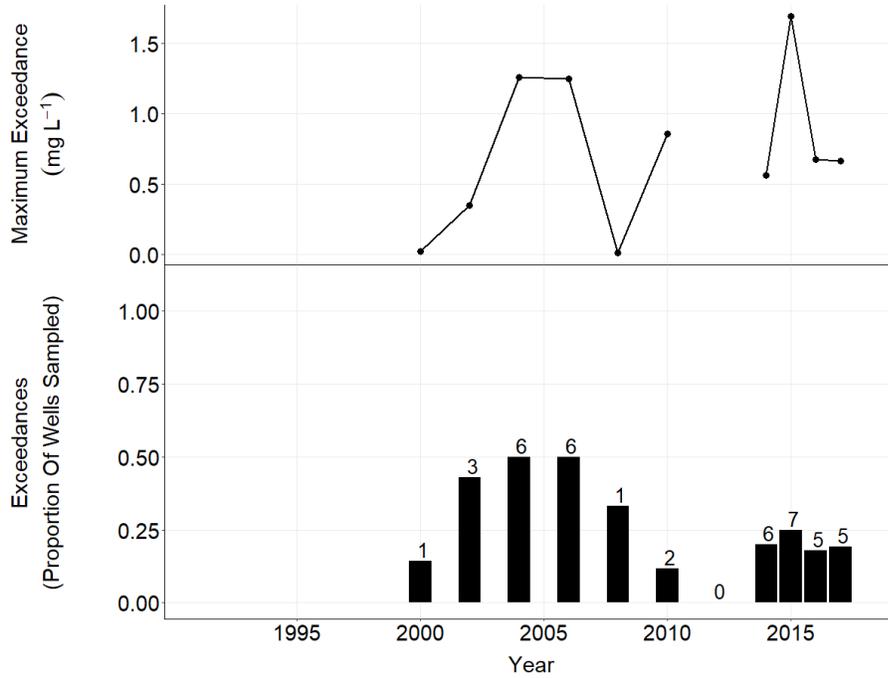


Figure 2.28. Ground water benzene exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

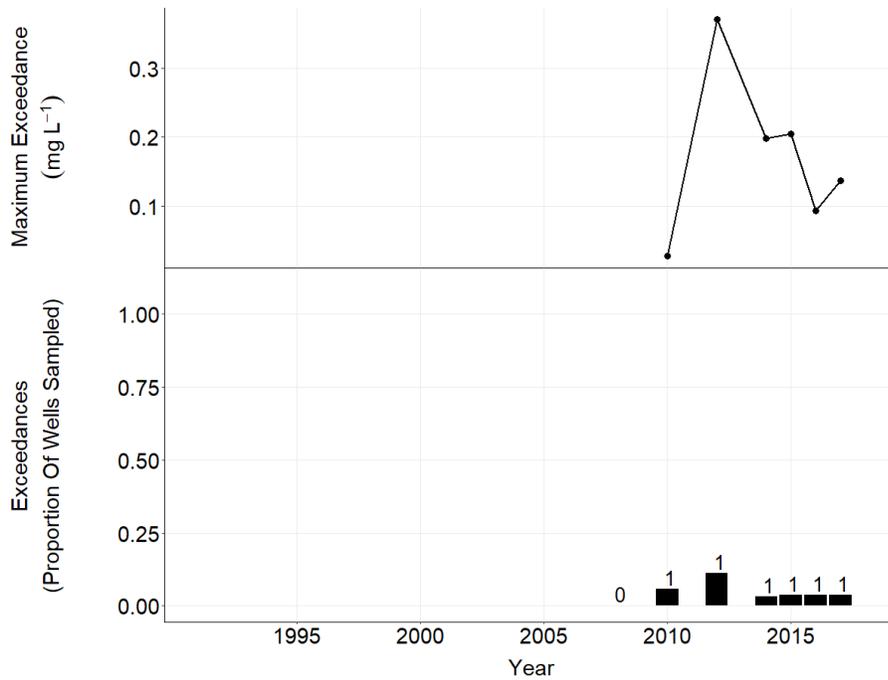


Figure 2.29. Ground water carbon tetrachloride exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

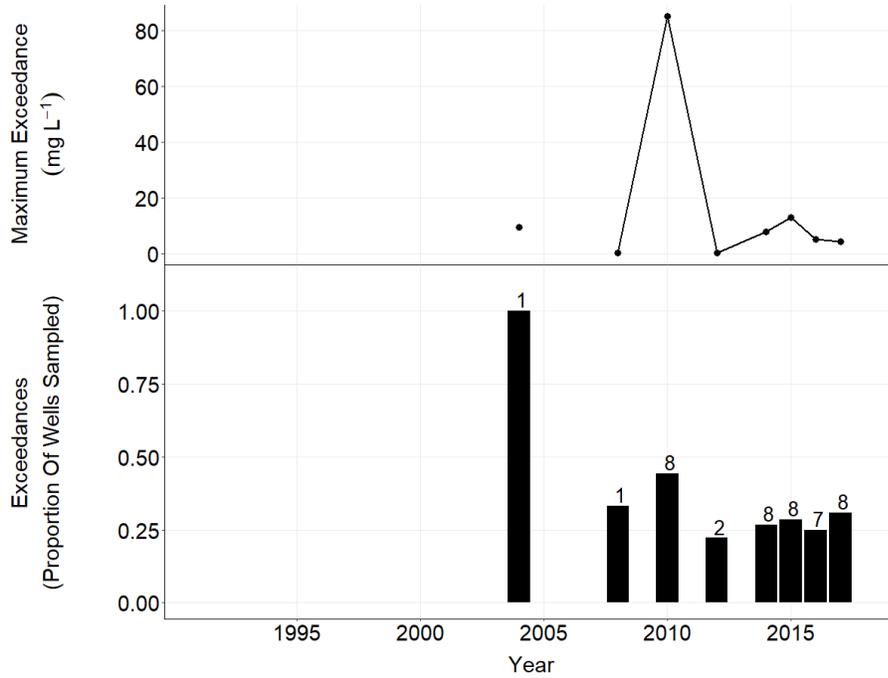


Figure 2.30. Ground water chloroform exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

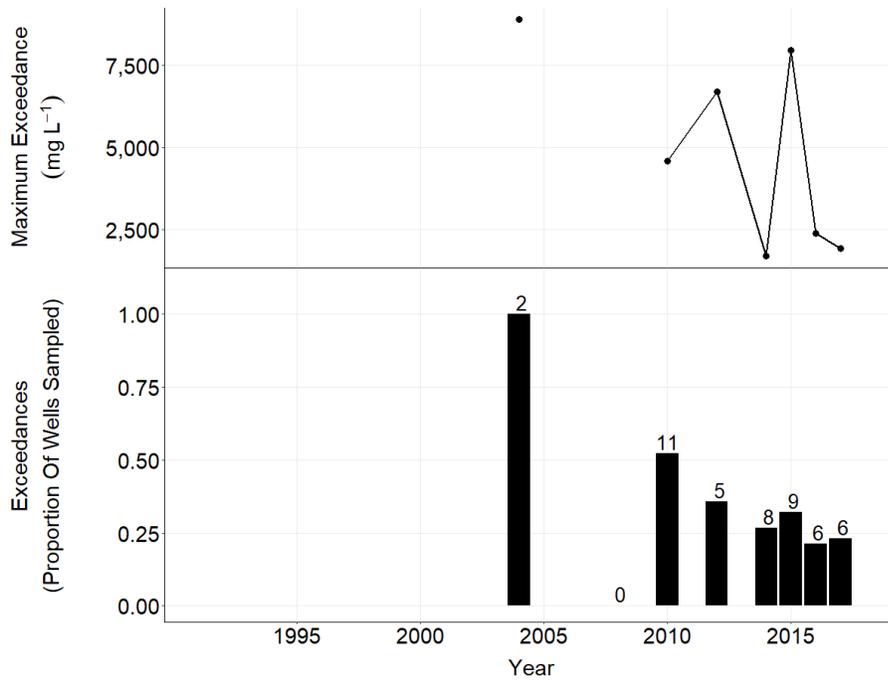


Figure 2.31. Ground water dichloromethane exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

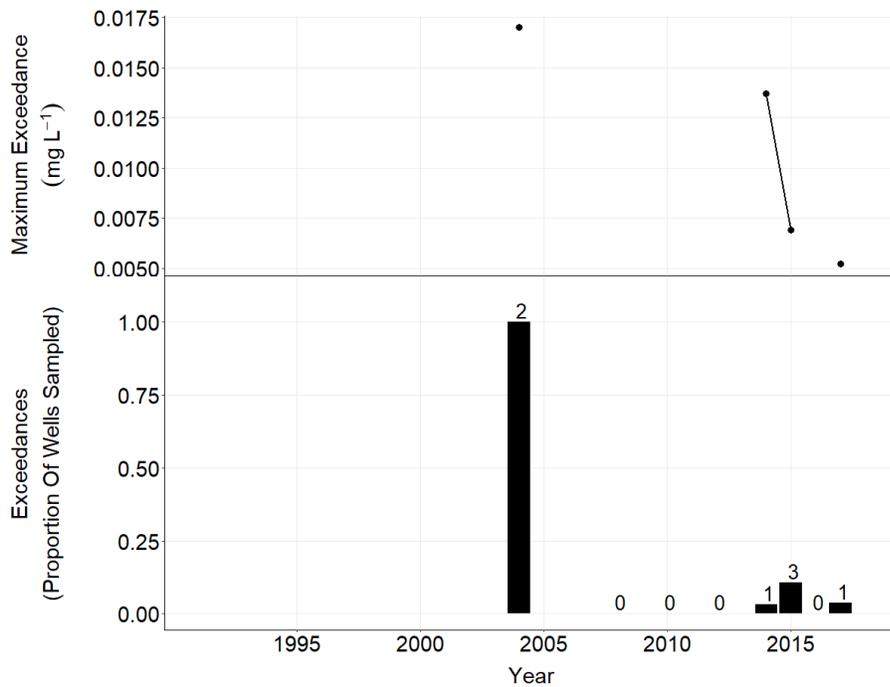


Figure 2.32. Ground water ethylbenzene exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

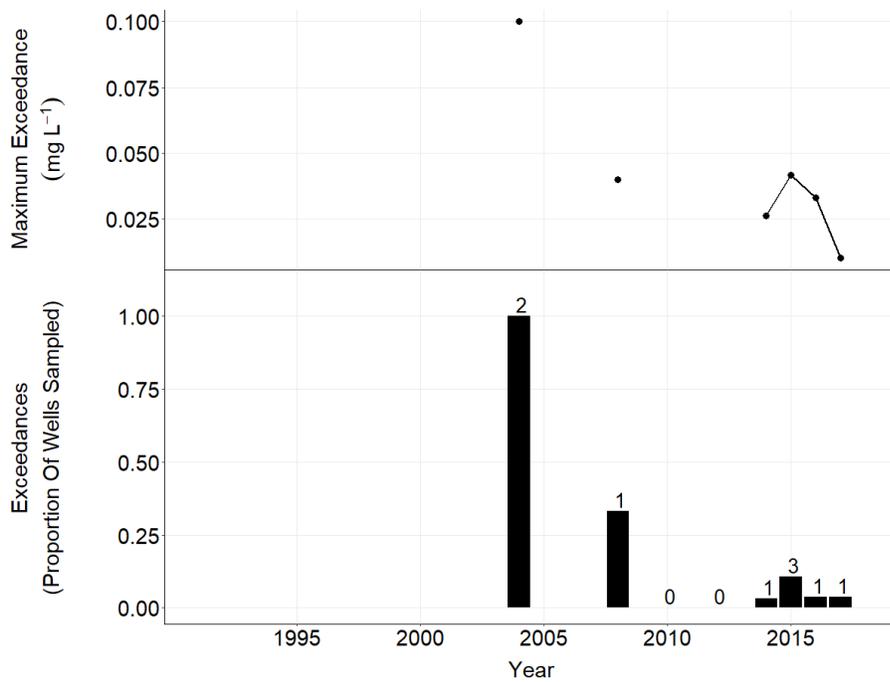


Figure 2.33. Ground water tetrachloroethene exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

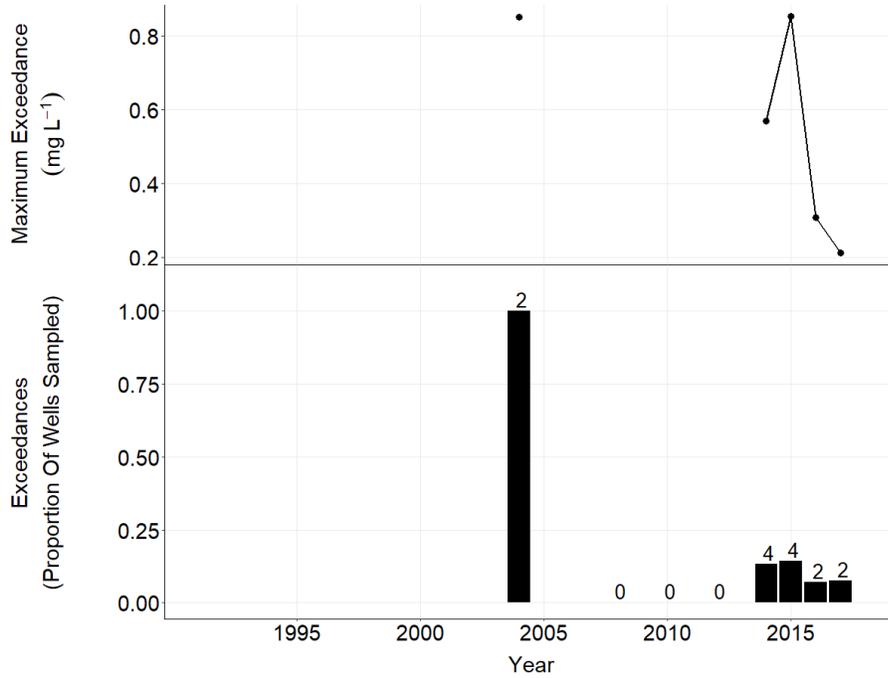


Figure 2.34. Ground water toluene exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

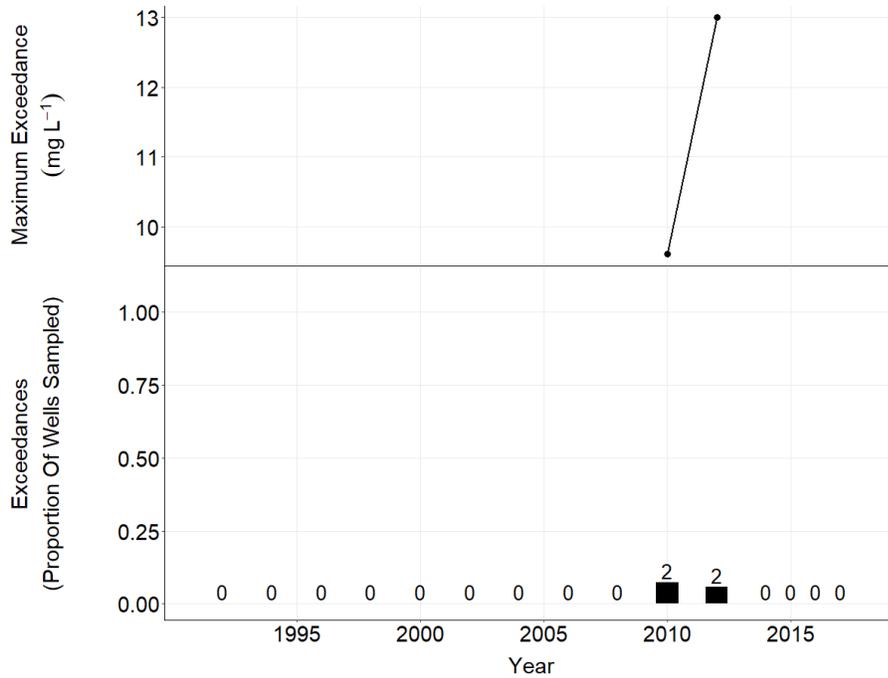


Figure 2.35. Ground water nitrate nitrogen exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

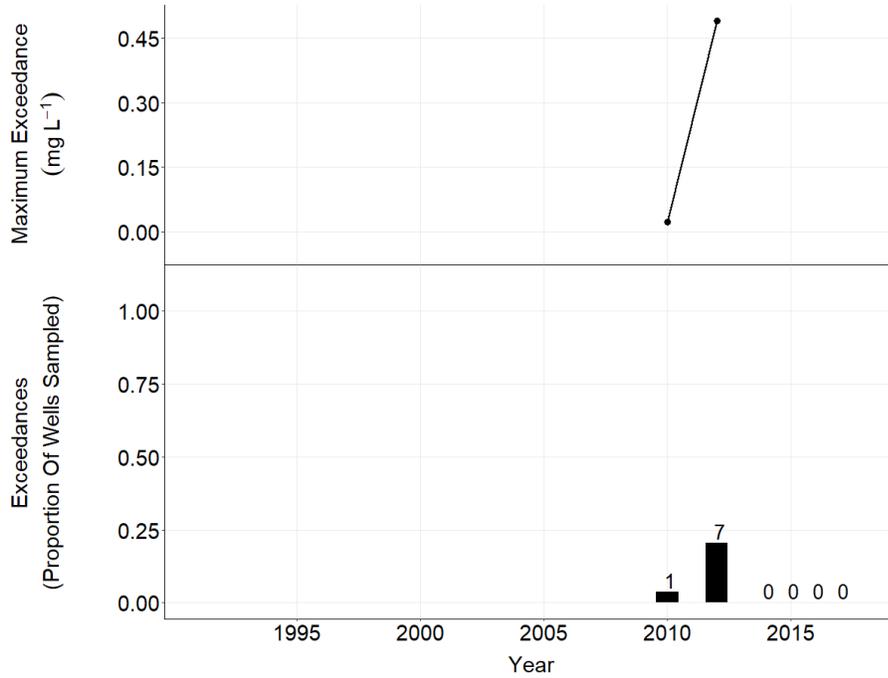


Figure 2.36. Ground water nitrite nitrogen exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

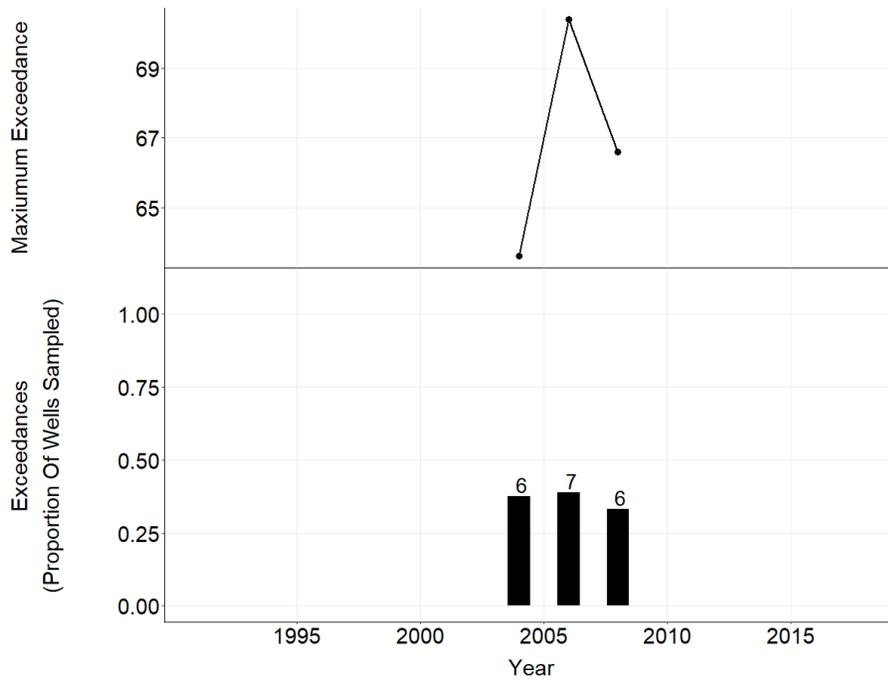


Figure 2.37. Ground water sodium adsorption ratio exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

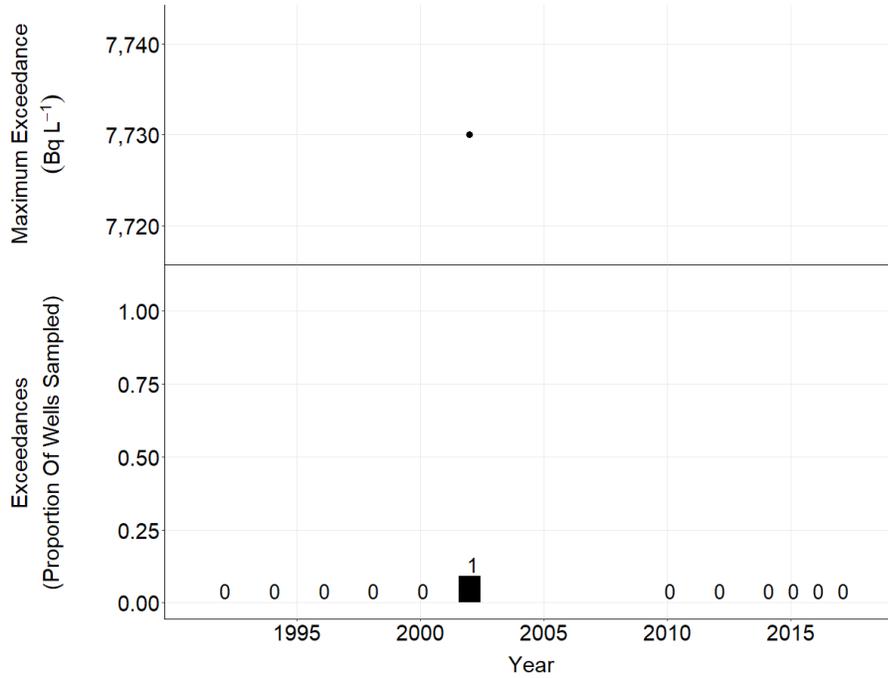


Figure 2.38. Ground water tritium exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

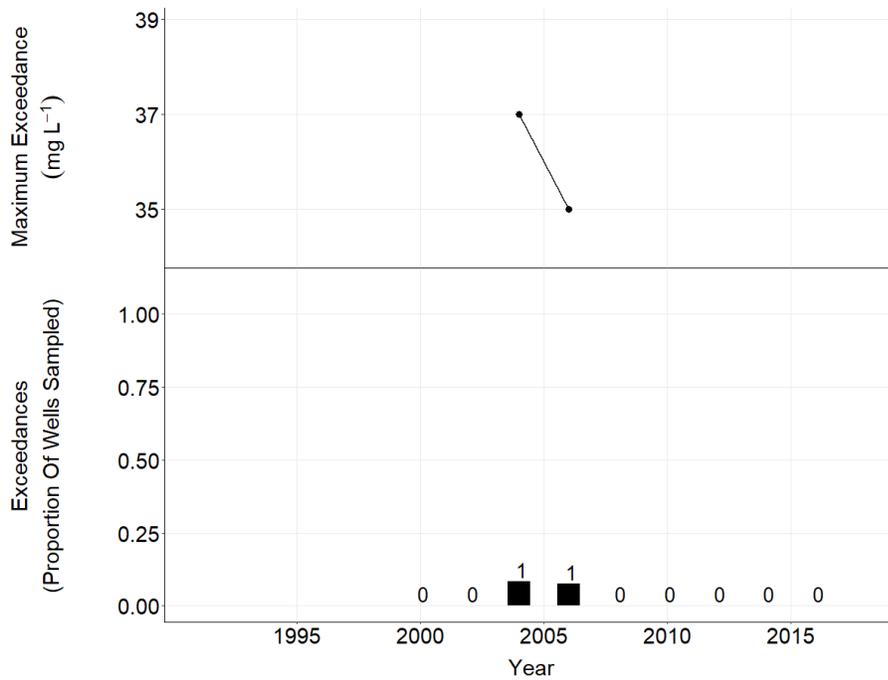


Figure 2.39. Ground water methanol exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

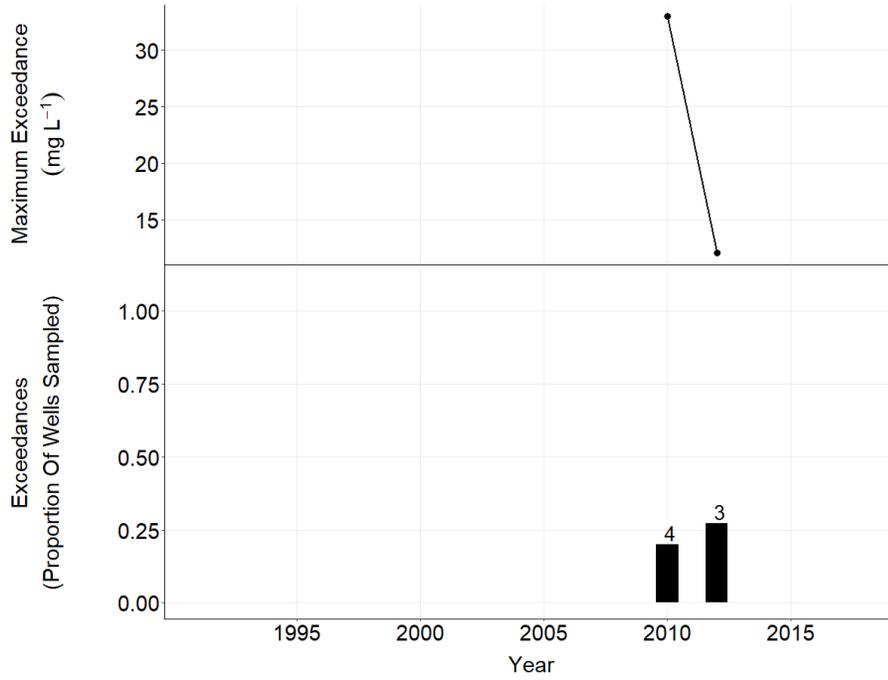


Figure 2.40. Ground water total trihalomethanes exceedances over time. Numbers above bars indicate number of wells with Alberta Tier 1 exceedances.

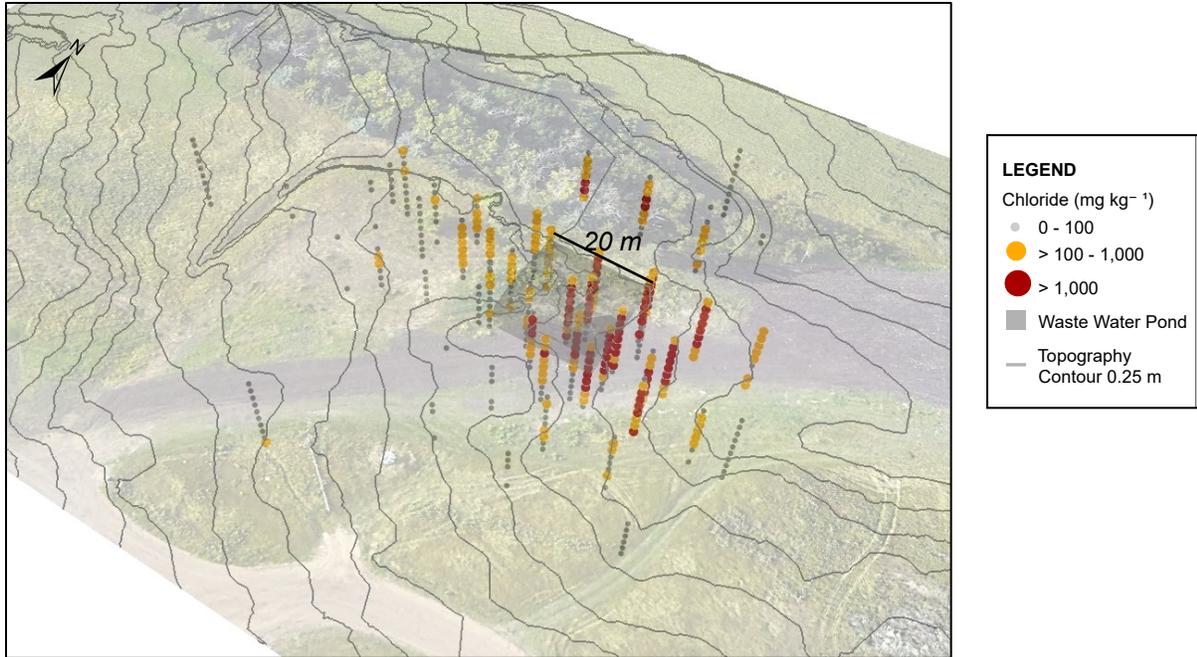


Figure 2.41. Soil chloride spatial distribution. Two times vertical exaggeration.

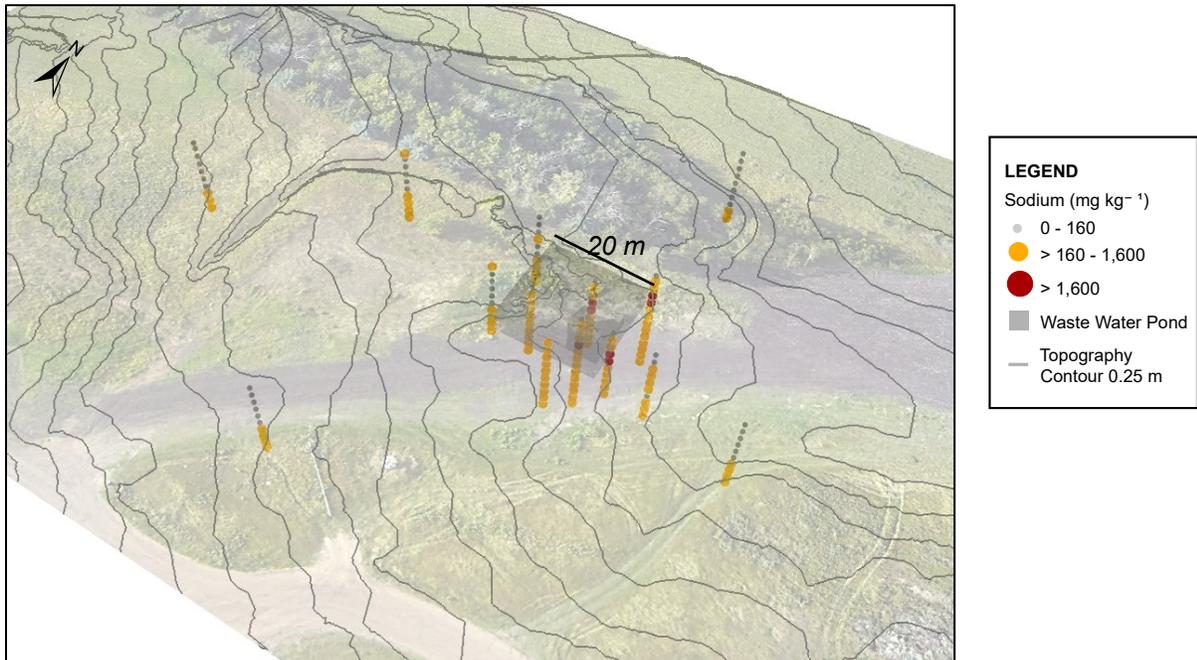


Figure 2.42. Soil sodium spatial distribution. Two times vertical exaggeration.

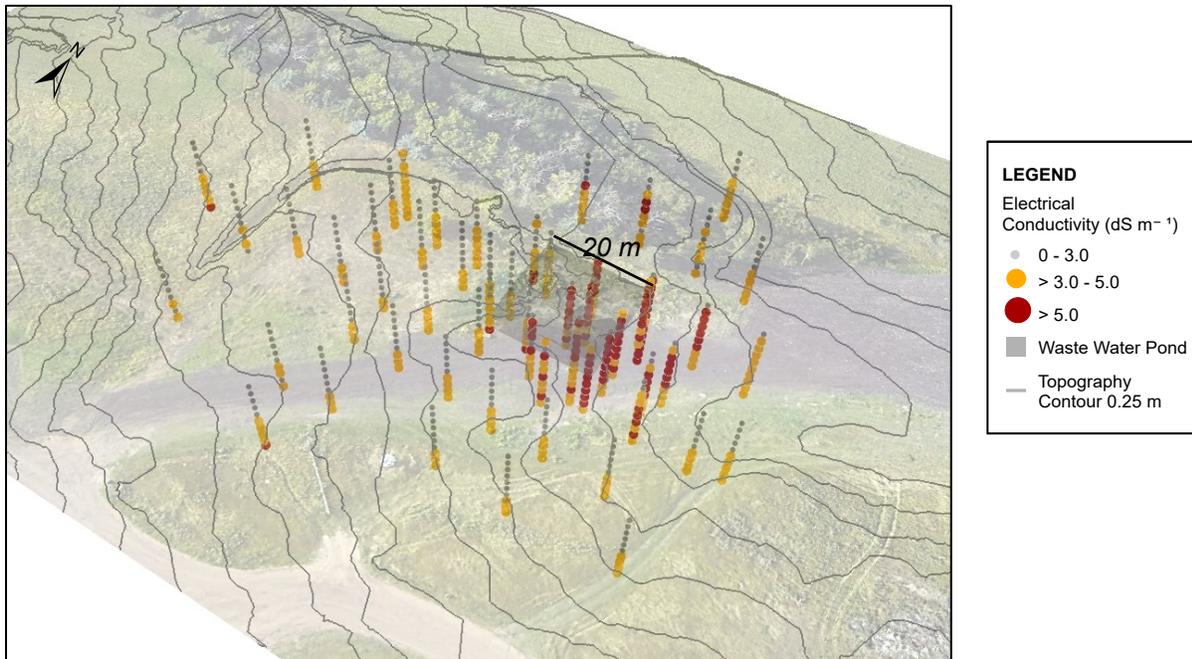


Figure 2.43. Soil electrical conductivity spatial distribution. Two times vertical exaggeration.

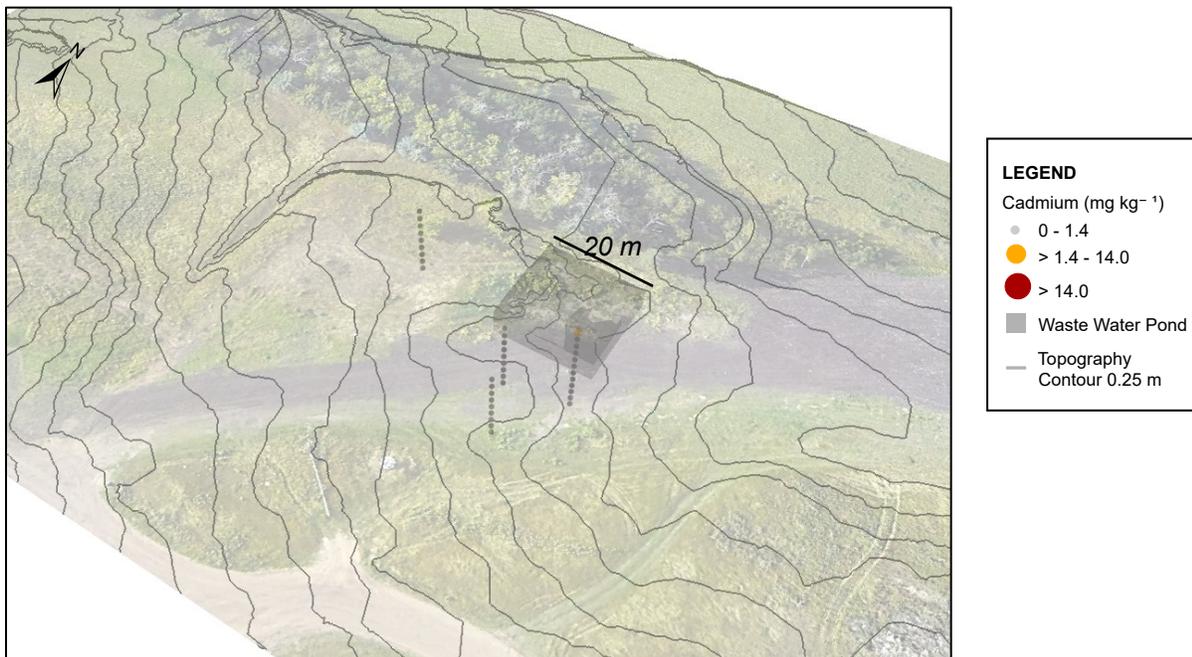


Figure 2.44. Soil cadmium spatial distribution. Two times vertical exaggeration.

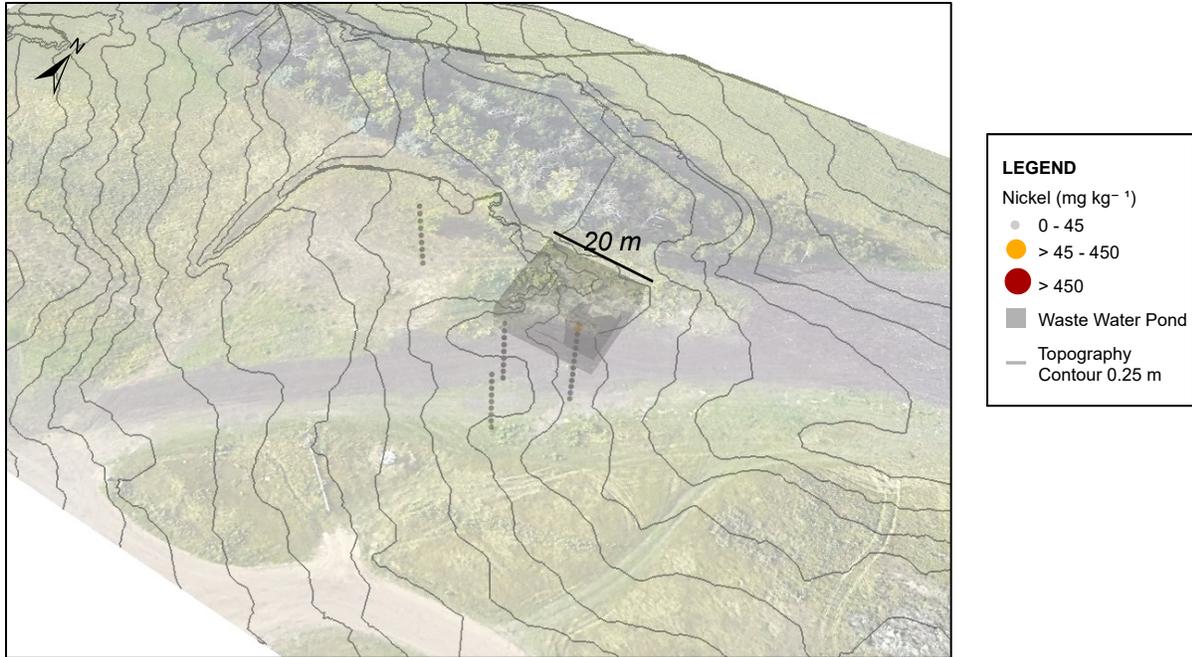


Figure 2.45. Soil nickel spatial distribution. Two times vertical exaggeration.

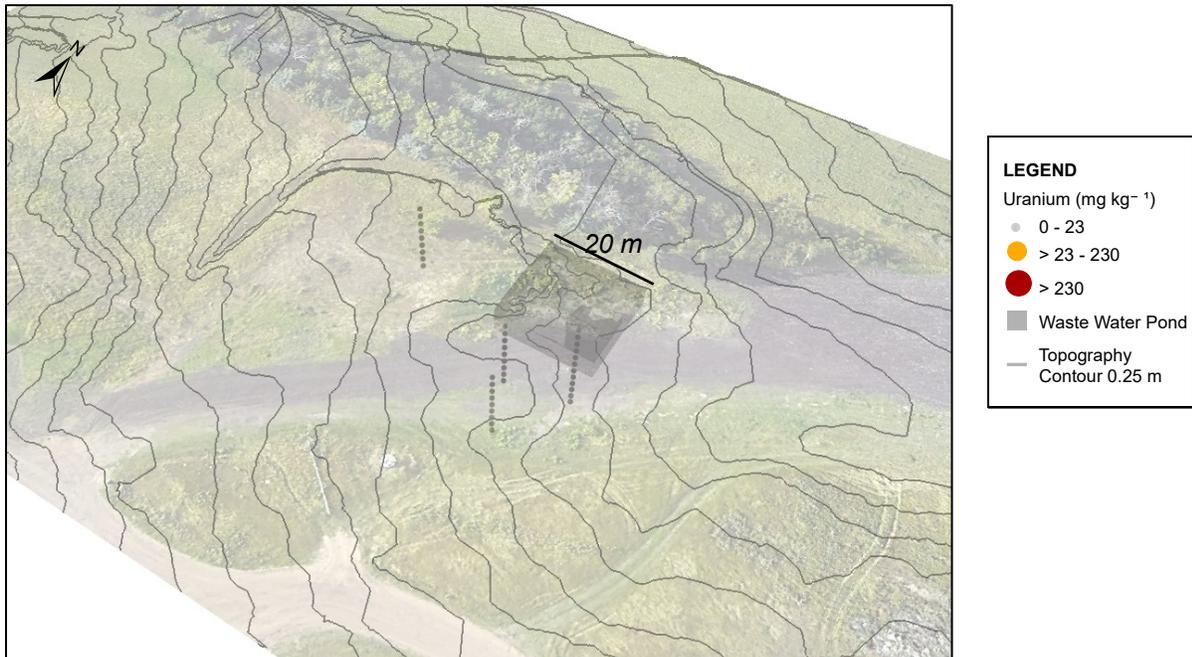


Figure 2.46. Soil uranium spatial distribution. Two times vertical exaggeration.

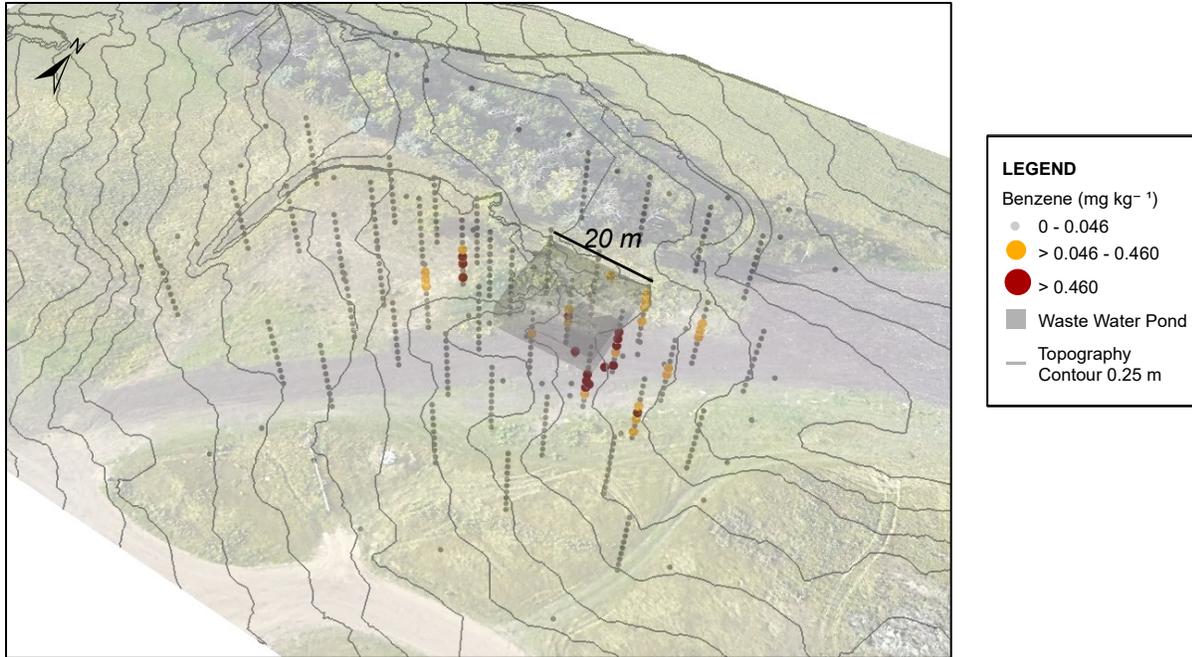


Figure 2.47. Soil benzene spatial distribution. Two times vertical exaggeration.

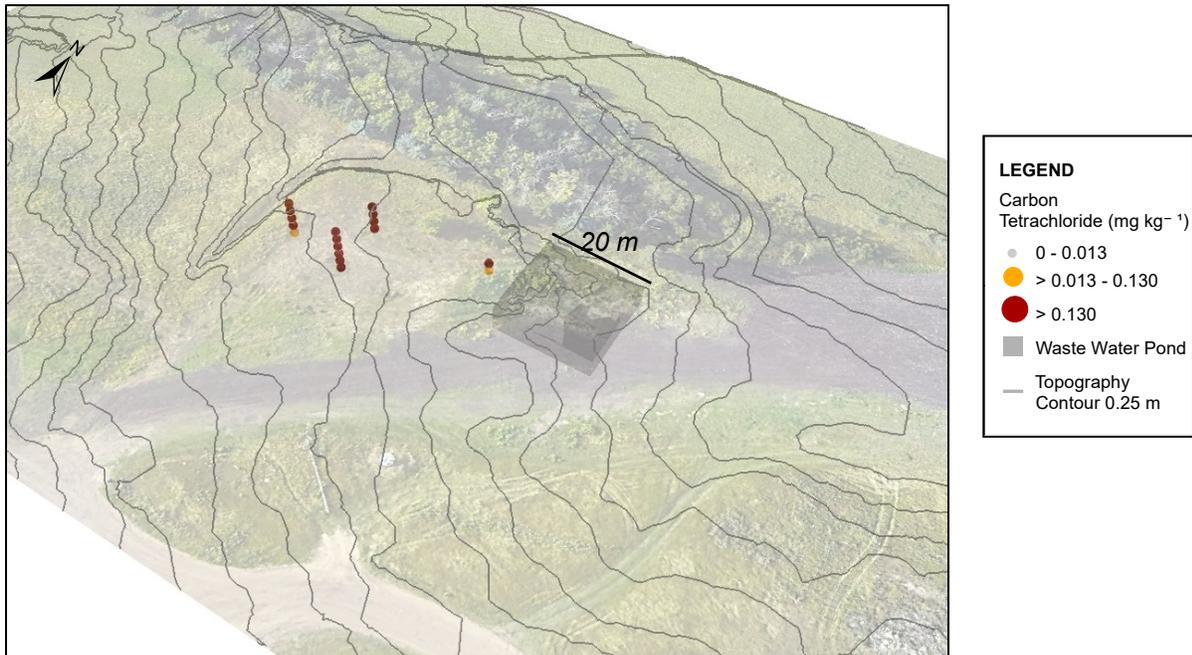


Figure 2.48. Soil carbon tetrachloride spatial distribution. Two times vertical exaggeration. 413 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline.

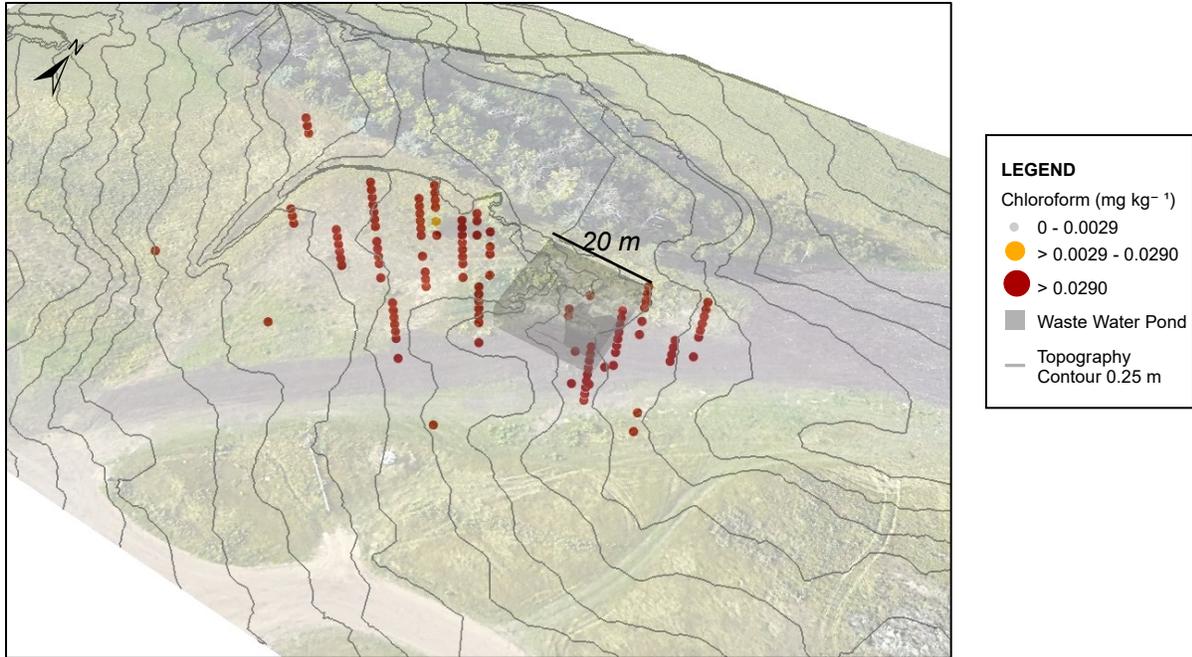


Figure 2.49. Soil chloroform spatial distribution. Two times vertical exaggeration. 374 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline.

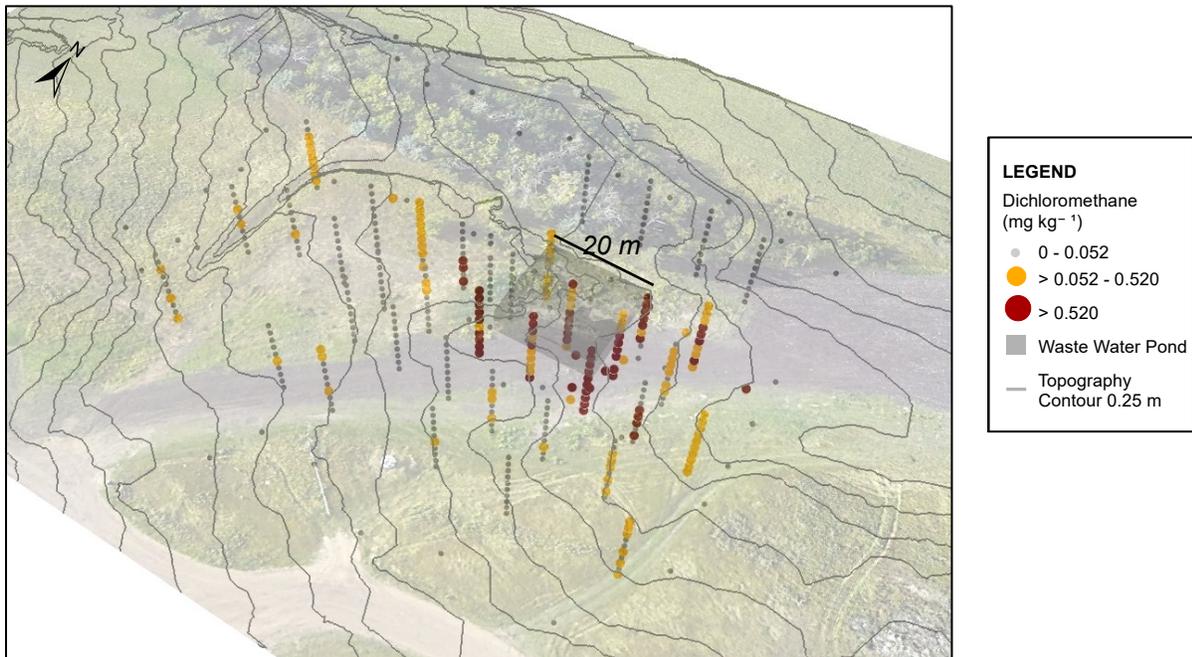


Figure 2.50. Soil dichloromethane spatial distribution. Two times vertical exaggeration. 53 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline.

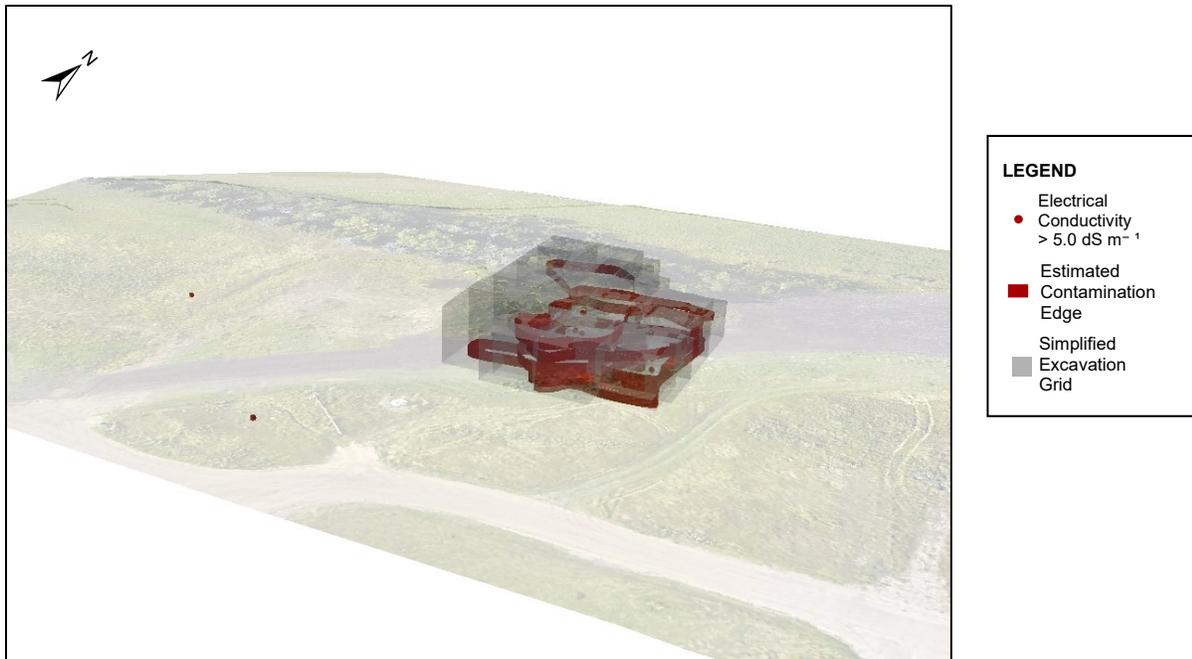


Figure 2.51. Estimated 12,580 m³ excavation volume for soil with electrical conductivity > 5 dS m⁻¹. Maximum east west extent is approximately 44 m, maximum north south extent is approximately 53 m, and maximum depth is approximately 14 m below surface.

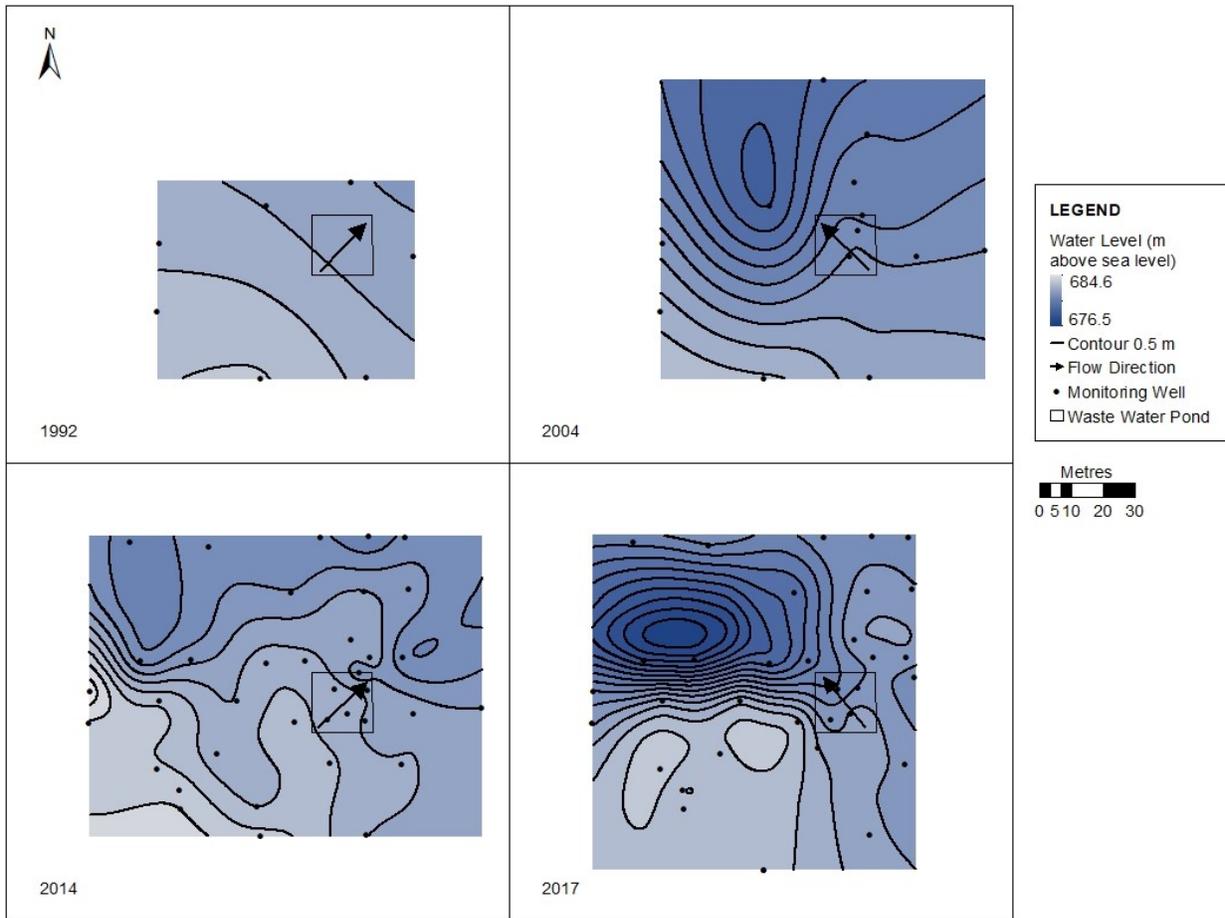


Figure 2.52. Ground water level and flow direction over time.

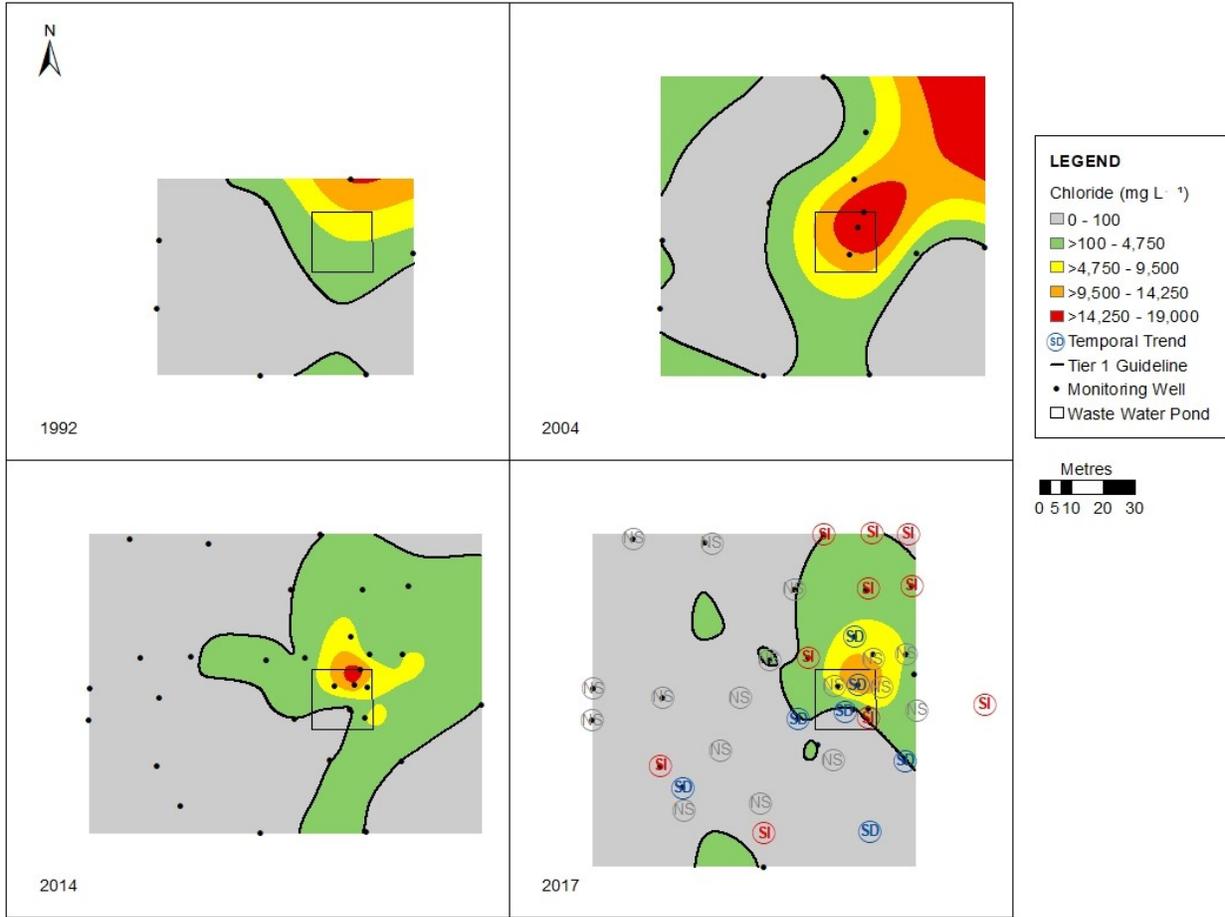


Figure 2.53. Ground water chloride spatial distribution over time. SI, SD, and NS indicate significant increase, significant decrease, and no significant change in concentration over time, respectively.

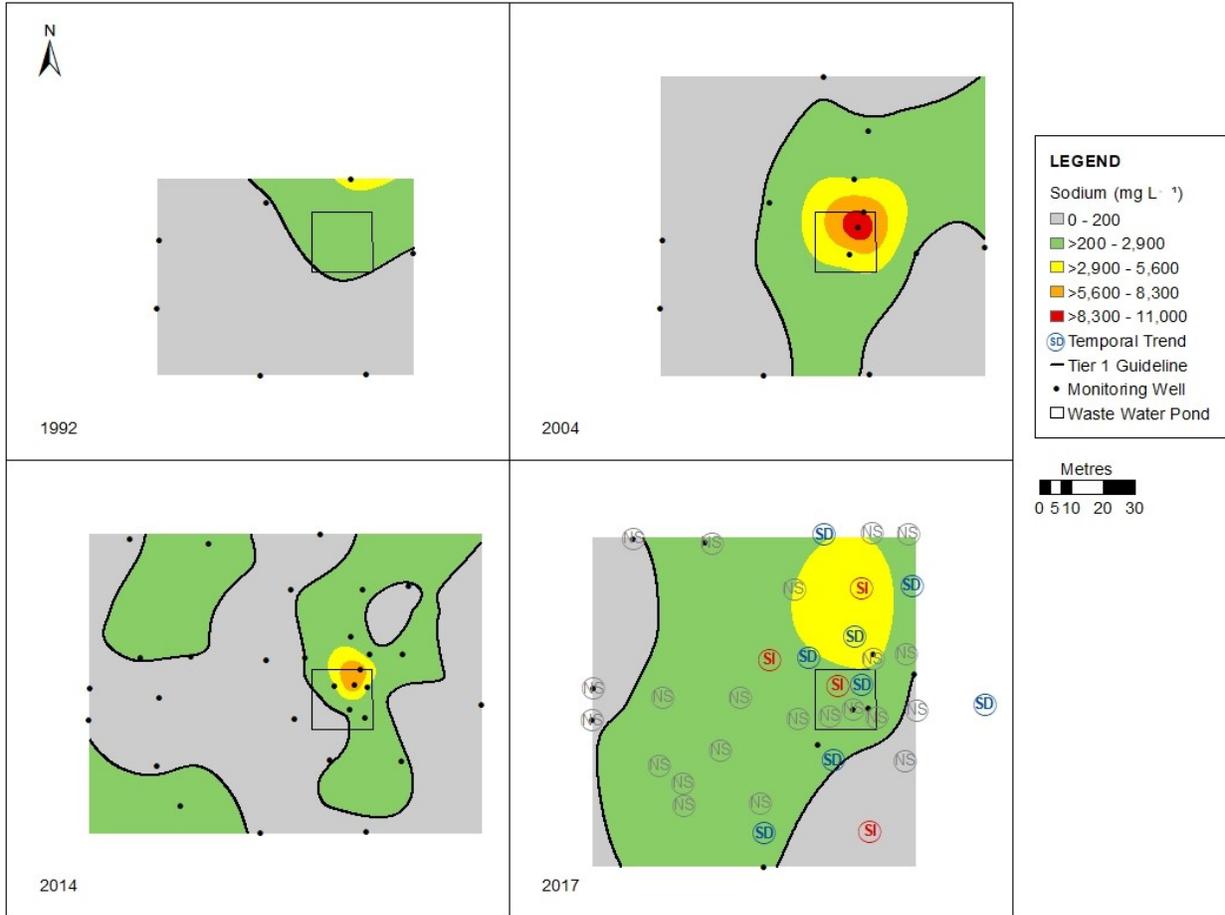


Figure 2.54. Ground water sodium spatial distribution over time. SI, SD, and NS indicate significant increase, significant decrease, and no significant change in concentration over time, respectively.

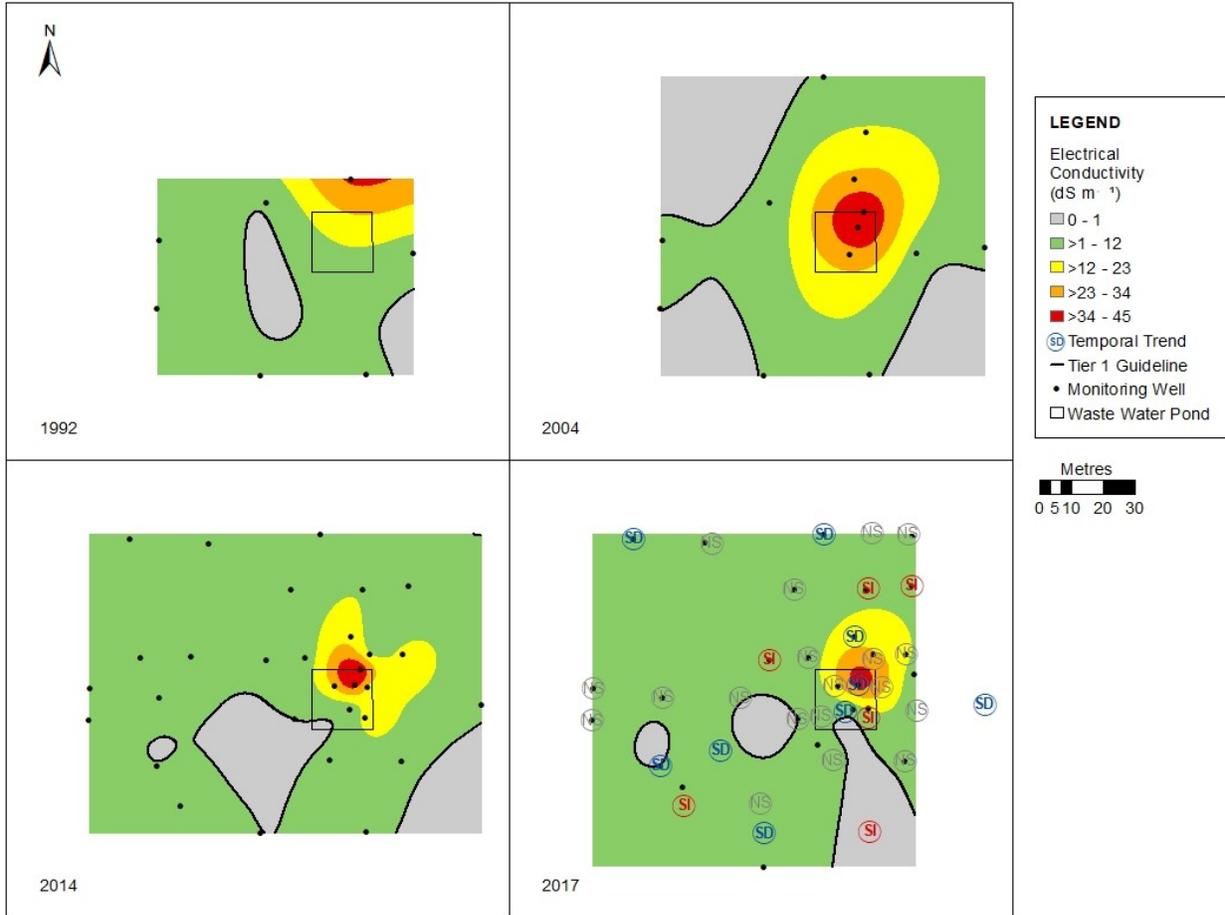


Figure 2.55. Ground water electrical conductivity spatial distribution over time. SI, SD, and NS indicate significant increase, significant decrease, and no significant change in concentration over time, respectively.

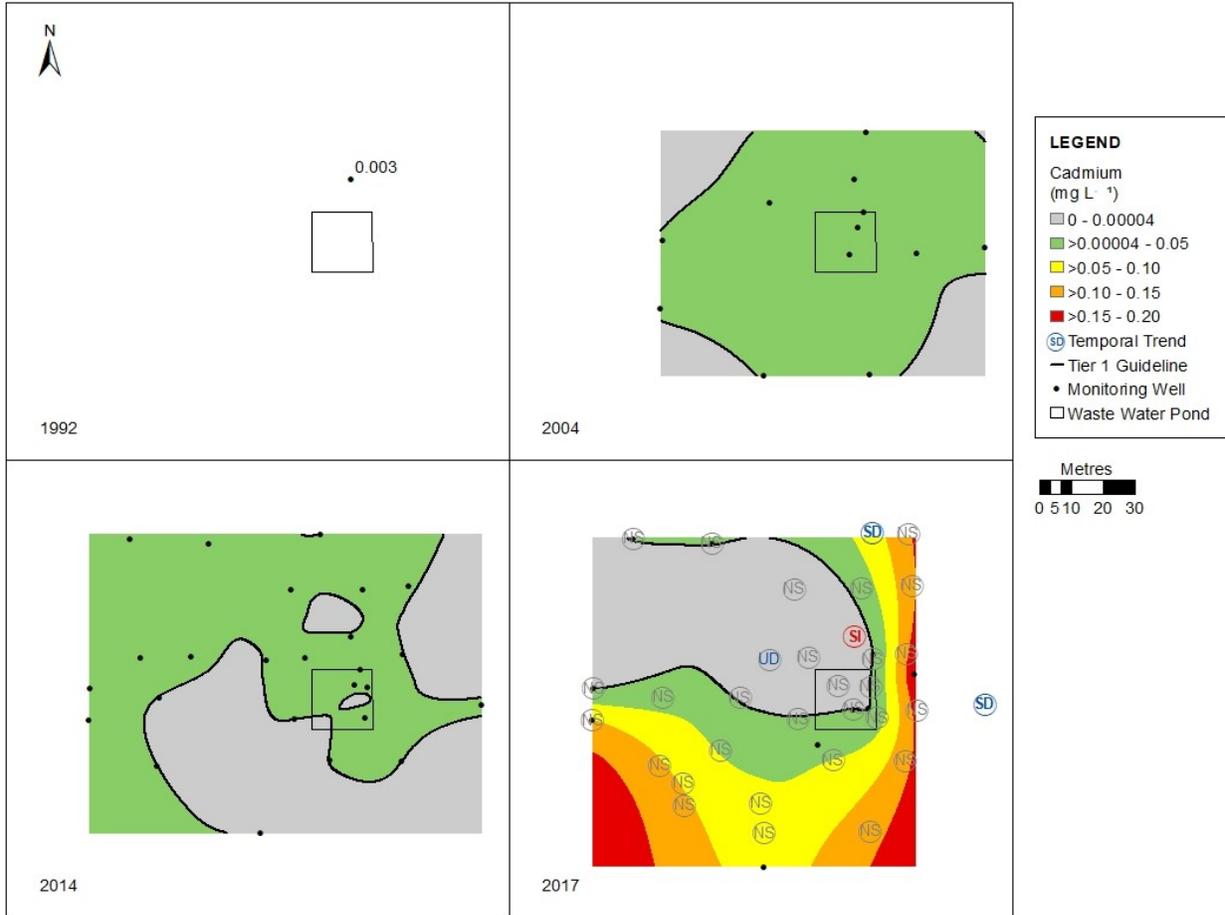


Figure 2.56. Ground water cadmium spatial distribution over time. SI, SD, UD, and NS indicate significant increase, significant decrease, uncertain decrease, and no significant change in concentration over time, respectively. Six 1992 values and five 2014 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline.

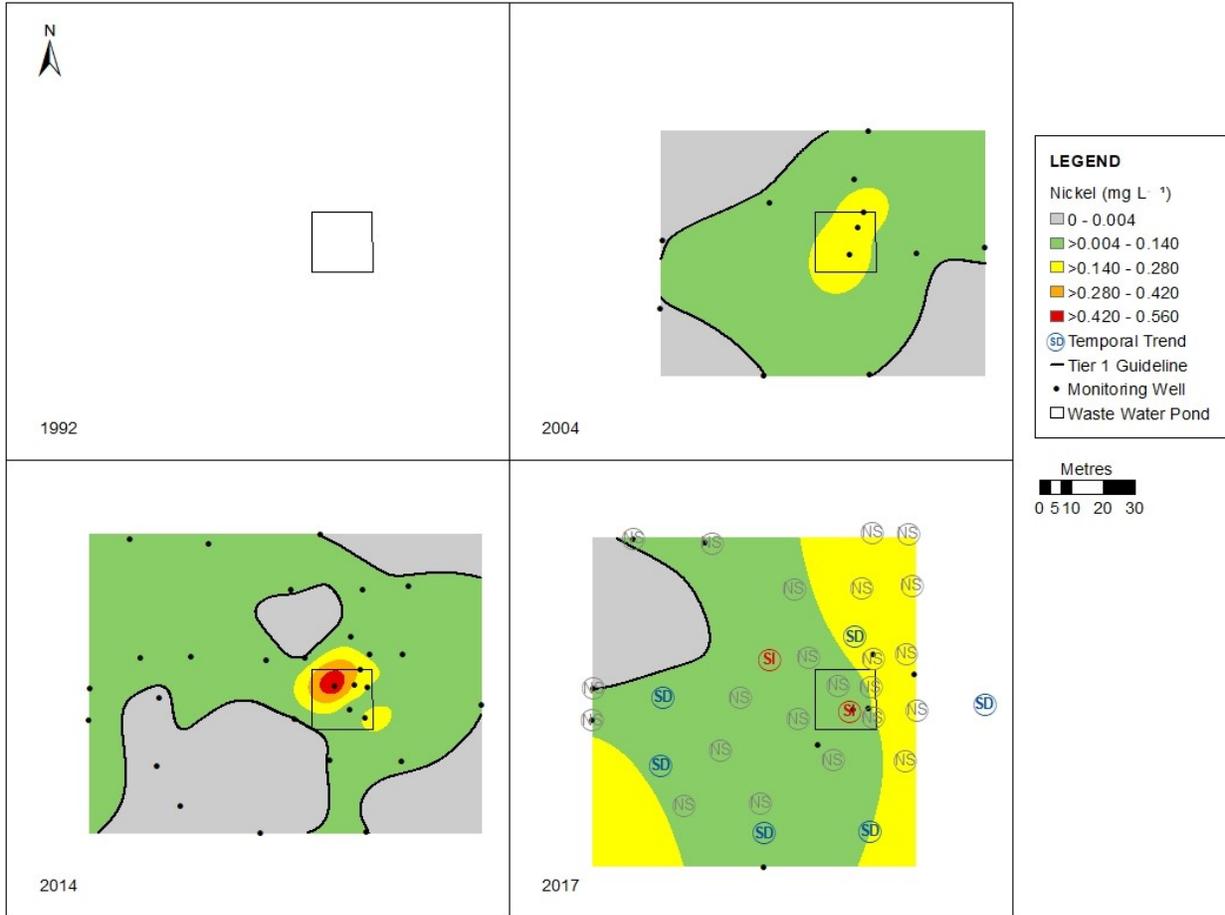


Figure 2.57. Ground water nickel spatial distribution over time. SI, SD, and NS indicate significant increase, significant decrease, and no significant change in concentration over time, respectively. All seven 1992 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline.

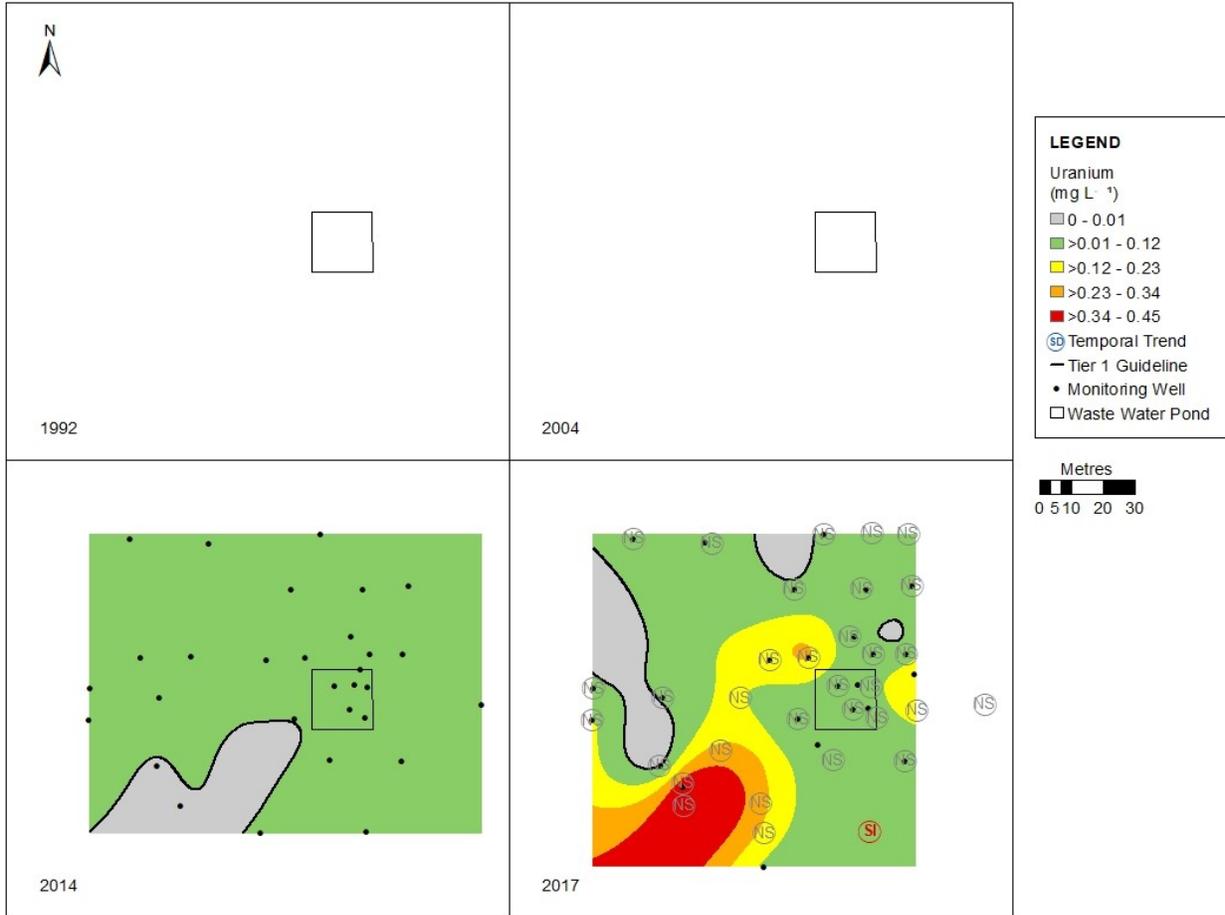


Figure 2.58. Ground water uranium spatial distribution over time. SI and NS indicate significant increase and no significant change in concentration over time, respectively. No data were available for 1992 or 2004.

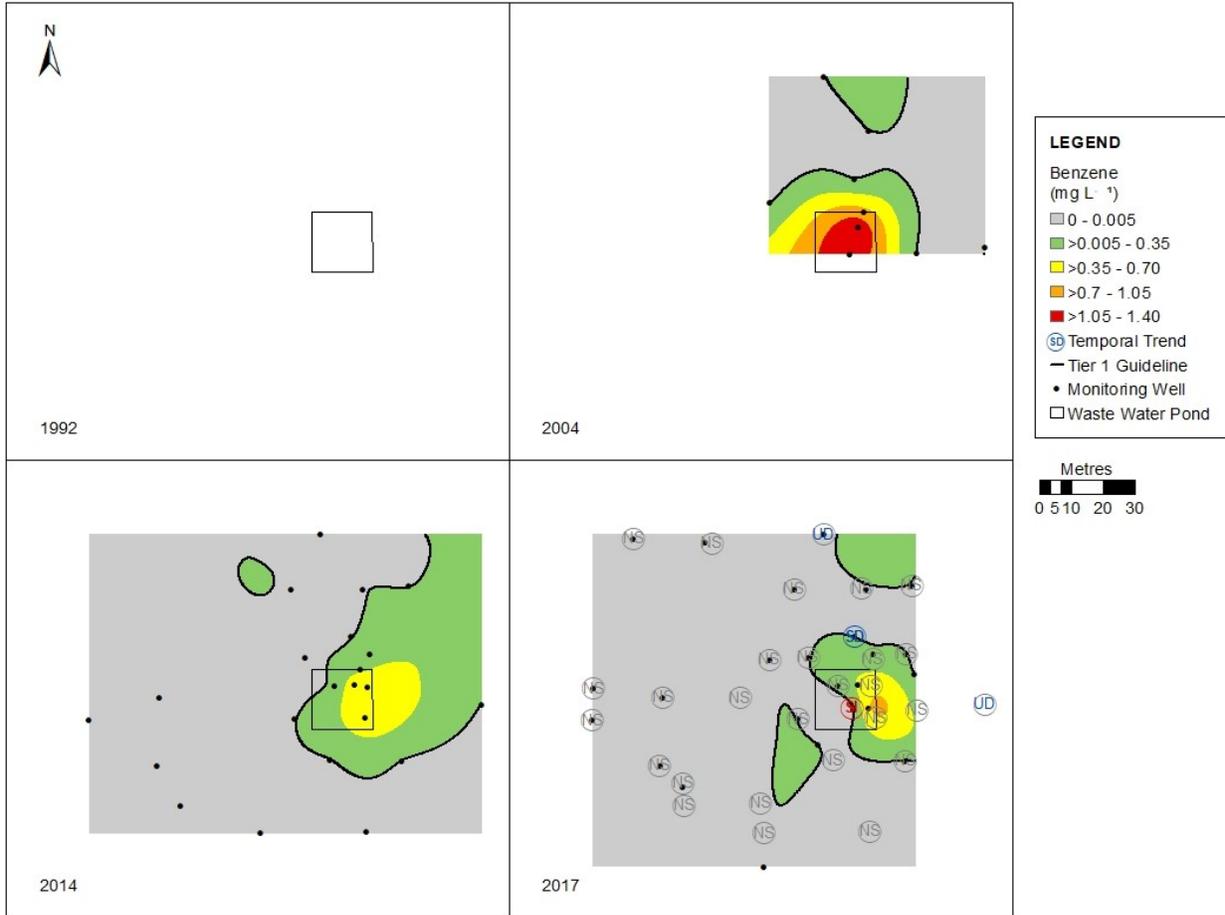


Figure 2.59. Ground water benzene spatial distribution over time. SI, SD, UD, and NS indicate significant increase, significant decrease, uncertain decrease, and no significant change in concentration over time, respectively. Eight 2014 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline. No data were available for 1992.

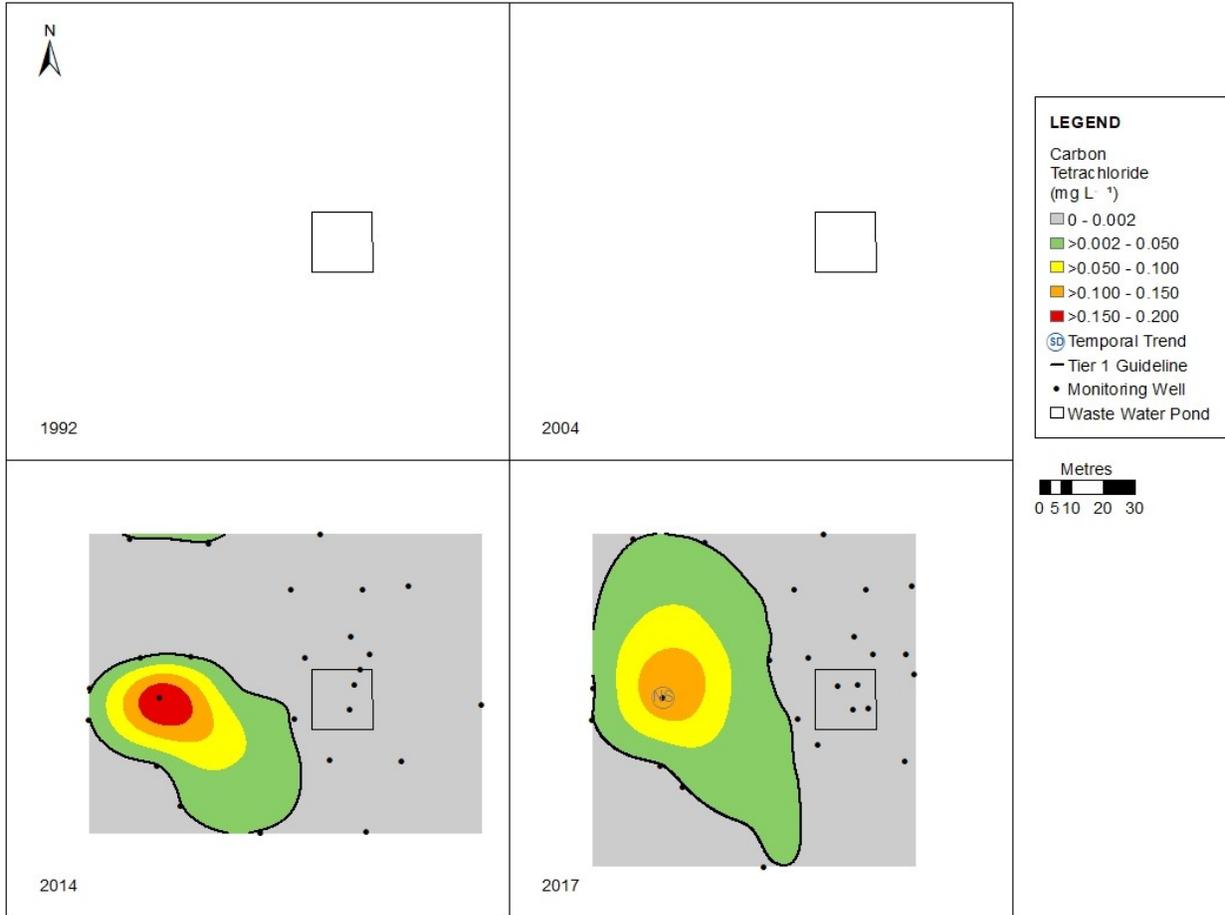


Figure 2.60. Ground water carbon tetrachloride spatial distribution over time. NS indicates no significant change in concentration over time. Five 2014 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline. No data were available for 1992 or 2004.

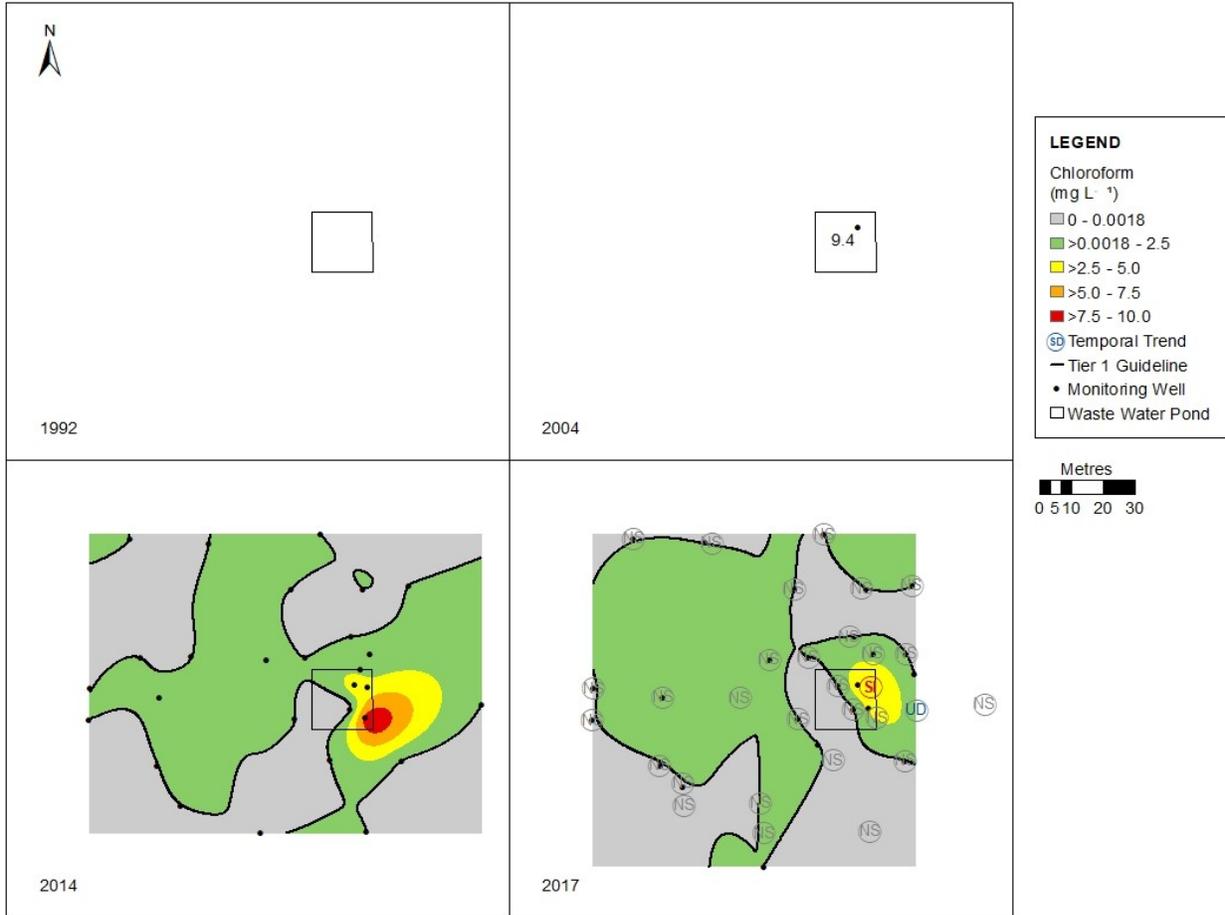


Figure 2.61. Ground water chloroform spatial distribution over time. SI, UD, and NS indicate significant increase, uncertain decrease, and no significant change in concentration over time, respectively. Two 2014 values were removed because they were below a detection limit which was higher than the Alberta Tier 1 guideline. No data were available for 1992.

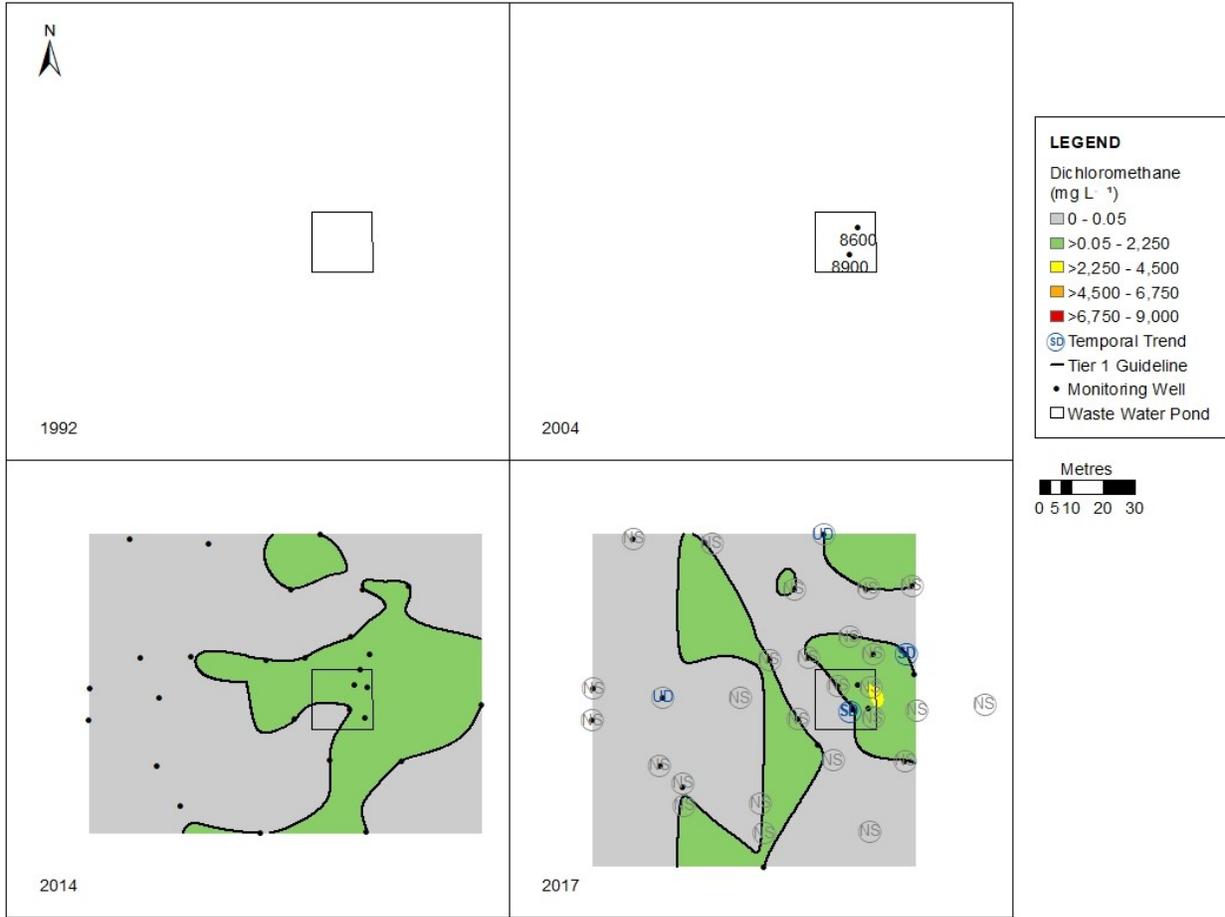


Figure 2.62. Ground water dichloromethane spatial distribution over time. SD, UD, and NS indicate significant decrease, uncertain decrease, and no significant change in concentration over time, respectively. No data were available for 1992.

III. BIOREMEDIATION OF CHLOROFORM CONTAMINATED SOILS EXPOSED TO MULTIPLE CONTAMINANTS IN ANAEROBIC MICROCOSMS

1. INTRODUCTION

Chloroform (CHCl_3) is a volatile solvent produced naturally by soil and oceanic processes (McCulloch 2003) and anthropogenically for industrial purposes such as production of plastics, refrigerants, pharmaceuticals, pesticides, dyes, and aerosol propellants (Chou and Spoo 1997, Canadian Council of Ministers of the Environment 1999). Since chloroform is a probable human carcinogen and causes cardiovascular, liver, and kidney damage (Chou and Spoo 1997, World Health Organization 2004), environmental limits have been set. Alberta Tier 1 remediation guidelines for chloroform in fine textured soils are $0.0029 \text{ mg kg}^{-1}$ soil and 0.0018 mg L^{-1} ground water (Alberta Environment and Parks 2016). The international drinking water guideline is 0.3 mg L^{-1} (World Health Organization 2011).

Chlorinated organic compounds can be biodegraded under aerobic and anaerobic conditions (Cappelletti et al 2012). Since chloroform is more dense than water, it can percolate through soils into anaerobic zones (Lee et al 2012). Under anaerobic conditions, chlorinated methane degradation is typically a cometabolic reductive dechlorination process (Cappelletti et al 2012). Primary carbon sources include volatile fatty acids, methanol, acetate, lactate, fructose, and glucose. Chloroform degradation is often incomplete, resulting in accumulation of degradation products such as dichloromethane. Anaerobic chlorinated methane degradation can also occur through dehalorespiration. Lee et al (2012) reported complete degradation of chloroform by a dehalorespiration pathway.

Chloroform is a common contaminant found at over 700 of 1,430 national priority list sites in the United States of America (Agency for Toxic Substances and Disease Registry 2014). It is often found at sites with multiple contaminants (Grostern et al 2010). In Canada, halogenated hydrocarbons are found at 157 federal contaminated sites, many of which also have other contaminants (Treasury Board of Canada Secretariat 2019). The presence of contaminant mixtures complicates remediation because each contaminant has unique properties which affect its toxicity, movement, and degradation. Although soil excavation and landfilling can remove all contaminants from a given contaminated site, they fail to remove contamination from the soil, thus rendering it waste for disposal, often in landfills. To conserve soils, remediation techniques or combinations of techniques should be used to remove each contaminant from the site. Each remediation technique used must be effective on its target contaminant(s) in the presence of the

other contaminants. Little research has been conducted on chloroform co-contaminant scenarios and nearly all of it focusses on the effects of chloroform contamination on bioremediation of other contaminants such as perchloroethene. Further research is thus required to improve our understanding of chloroform bioremediation in realistic settings involving complex contaminant mixtures.

The objective of this research was to determine the potential for soil indigenous or inoculation microorganisms to anaerobically degrade chloroform in soil containing multiple contaminants. Three laboratory experiments were conducted using soils from two research sites for active bioremediation. In experiments 1 and 3, chloroform biodegradation by soil microorganisms from the primary research site was assessed under various anaerobic conditions to determine biostimulation treatment potential. In experiment 2, chloroform biodegradation by soil microorganisms from a secondary research site was assessed under various anaerobic conditions to determine bioaugmentation treatment potential. The anaerobic conditions or treatments were canola oil, acetate, nitrate, and sulfate, with lactate treatment added in experiment 3. Canola oil, acetate, and lactate are electron donors and carbon sources for microbiological use in cometabolic reductive dechlorination. Nitrate and sulfate are terminal electron acceptors to provide nitrate and sulfate reducing conditions.

2. MATERIALS AND METHODS

2.1. Research Sites

Soils were collected from the University of Alberta's former Eilerslie Waste Management Facility in Edmonton, Alberta (Appendix) and a secondary site approximately 40 km away in Fort Saskatchewan, Alberta. The approximately 0.25 ha Eilerslie site is located within the Province of Alberta's Edmonton Transportation Utility Corridor with surroundings zoned for agriculture (Alberta Infrastructure 2003, AMEC Earth and Environmental 2010). A major road is located immediately north and residential areas are north and east. Whitemud Creek is approximately 450 m north. The site processed laboratory waste from 1972 to 2007 (Stantec Consulting Limited 2013); activities included solvent incineration, acid dilution and neutralization, and temporary waste storage. Prior to 1983, a waste water pond leaked (University of Alberta 1984). Primary contaminants include chloroform, dichloromethane, and chloride; an additional 15 soil and 34 ground water parameters were identified in exceedance of Alberta Tier 1 soil and ground water remediation guidelines for fine textured soils and agricultural land use (Alberta Environment and Parks 2016). Surficial geology at the Eilerslie site consists mainly of clay and

clay till (Stantec Consulting Limited 2010). Much of the contamination is considered under anaerobic conditions because of its depth relative to the water table and restricted oxygen flow in fine textured soils. The secondary research site in Fort Saskatchewan was industrial with multiple long term contaminant issues. Both research sites are located in the Central Parkland natural subregion (Natural Regions Committee 2006).

2.2. Microcosms

Soil for the experiments was collected by direct push drilling. Soil for experiment 1 was collected from the Ellerslie site in 2014. A composite sample from the 5 to 6 m depth of 8 bore holes within the former waste water pond area was prepared. This depth zone represents the most contaminated soil at the site and the most likely habitat for microorganisms capable of degrading chlorinated organic compounds. Soil for experiment 2 was collected from the Fort Saskatchewan site in 2015. Soil cores were collected, wrapped in plastic to minimize exposure to oxygen and microbiological contamination, and transported to an anaerobic chamber at the University of Alberta where at least 1 cm soil was removed from any surface that touched a non sterile surface such as the core liner or plastic wrap (Kieft 2014). A composite soil sample from the 1.5 to 3 m depth was prepared. Soil for experiment 3 was collected from the Ellerslie site in 2016. Soil from the 5 to 6 m depth of 3 bore holes within the pond area were processed as for experiment 2.

Cultures were set up in 158 mL sterile clear glass bottles, each containing 15 g field wet soil and 100 mL sterilized nutrient medium. The medium preparation method was adapted from Lee et al (2012). A 3 L solution contained 7.5 g NaHCO_3 , 4.5 g NH_4Cl , 1.8 g NaH_2PO_4 , 0.3 g $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, 0.3 g $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, 3.0 mL trace element solution A 1000x, 3.0 mL trace element solution B 1000x, 3.0 mL vitamin solution, and 75 mg resazurin. Trace element solution A contained 1 mL 3 M HCl, 150 mg $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$, 19 mg $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, 10 mg $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$, 7.0 mg ZnCl_2 , 0.6 mg H_3BO_3 , 3.6 mg $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$, 2.4 mg $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$, and 0.2 mg $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ in 99 mL water (Loffler et al 2005). Trace element solution B contained 1.2 mg Na_2SeO_3 , 1.6 mg $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$, and 100 mg NaOH in 200 mL water. The vitamin solution contained 2.0 mg biotin, 2.0 mg folic acid, 10 mg pyridoxine hydrochloride, 5.0 mg riboflavin, 5.0 mg thiamine, 5.0 mg nicotinic acid, 5.0 mg pantothenic acid, 0.1 mg vitamin B12, 5.0 mg p-aminobenzoic acid, and 5.0 mg thiocetic acid in 100 mL water adjusted to pH 7.5 using small amounts of NaOH and/or HCl (Wolin et al 1963). A stock solution of 72.6 mg cysteine in 15 mL water was sterilized and 500 μL added to each bottle. Bottles were capped with sterile butyl stoppers and aluminum seals. Bottles were flushed with 20 % CO_2 80 % N_2 for 30 seconds then vacuumed 30 seconds,

alternating for 3 minutes per bottle to remove oxygen from the headspace. Bottles were incubated in the dark at room temperature for at least five weeks.

Following incubation, sterile control bottles were autoclaved. The headspace of each bottle was flushed with 20 % CO₂ 80 % N₂ for 5 minutes before amendment addition. Each bottle was amended to 2.36 mM chloroform and 78.4 μM methylcyclohexane, an internal standard. Treatment concentrations were selected based on stoichiometry of reactions with organic carbon. Experiment 1 bottles were amended to 4.17 mM canola oil, 6.78 mM acetate, 2.71 mM nitrate, or 1.18 mM sulfate. Calculations for experiments 2 and 3 were refined and bottles amended to 4.74 mM canola oil, 4.72 mM acetate, 1.89 mM nitrate, or 1.18 mM sulfate. A 4.72 mM lactate treatment was added to experiment 3. Canola oil, acetate, and lactate were added as potential carbon sources for reductive dechlorination of chloroform. Nitrate and sulfate were added to initiate nitrate and sulfate reducing conditions, respectively, and act as potential electron acceptors in degradation of chloroform as a carbon source. Each treatment had three replicate bottles and two sterile control bottles in experiments 1 and 2 and three sterile control bottles in experiment 3. Three control treatment bottles in each experiment did not receive amendments except chloroform and methylcyclohexane.

Headspace analysis for chloroform, dichloromethane, methane and methylcyclohexane was completed following amendment addition and at approximately three week intervals thereafter using a gas chromatograph with a flame ionization detector. Pressure was measured for each bottle. Analysis frequency was reduced after several months of monitoring with no observation of chloroform biodegradation or degradation product formation.

2.3. Data Analyses

The amount of chloroform remaining was calculated as $(\text{chloroform}_i / \text{methylcyclohexane}_i) / (\text{chloroform}_s / \text{methylcyclohexane}_s) \times 100 \%$; where chloroform is the peak area for chloroform on the gas chromatography spectrum, methylcyclohexane is the peak area for methylcyclohexane on the gas chromatography spectrum, i is a live microcosm bottle, and s is the mean of the sterile microcosm bottles for the same treatment as i. The remaining chloroform for control bottles, which did not have sterile equivalents, was calculated using sterile acetate bottles in experiments 1 and 2 and the mean of sterile sulfate and sterile nitrate bottles in experiment 3, since analyses occurred on the same calendar days, respectively. Acetate, sulfate, and nitrate were not expected to chemically and/or physically impact either the chloroform or the methylcyclohexane.

Repeated measures statistical analysis was completed using the lme function of the nlme package in R. Correlation structures corAR1, corCAR1, and corARMA were tested with and without adjustment for heterogeneous variances. The model with the lowest Akaike information criterion value was selected. The corAR1 correlation structure was selected with heterogeneous variances for experiments 1 and 2 and homogeneous variances for experiment 3. Normal distribution of residuals was assessed using the Shapiro Wilk test. Post hoc Tukey comparisons of means were performed when significant effects were identified.

3. RESULTS

No large changes in headspace chloroform remaining over time were identified in any of the three experiments. Formation of potential degradation products dichloromethane or methane was not observed. Analysis of mean headspace chloroform remaining over time for experiment 1 indicated that time ($p = 0.0001$) and its interaction with treatment ($p = 0.0003$) were significant (Figure 3.1). Treatment was not significant ($p = 0.1454$). Comparisons of time within treatment showed chloroform decreased over time for the control. Control mean headspace chloroform remaining was 98.1 % at day 0 and decreased to 88.8 % by the end of the experiment with each value being significantly different. For all other treatments, chloroform did not change significantly over time. In experiment 2, treatment ($p = 0.4021$), time ($p = 0.4495$), and their interaction ($p = 0.6584$) were not significant (Figure 3.2). In experiment 3, data for acetate sterile bottles 1 and 2, lactate sterile bottle 2, lactate bottle 4, and nitrate bottle 5 were omitted because of differences in methylcyclohexane peak areas relative to other bottles from the same treatment. Since methylcyclohexane is meant to be a stable internal standard, bottles with differences were considered outliers. Treatment was significant ($p = 0.0309$), while time ($p = 0.7164$) and its interaction with treatment ($p = 0.1949$) were not (Figure 3.3). Residuals were not normally distributed ($p = 0.0004$); however, the shape of the residuals histogram showed the concept of mean was meaningful. When treatments were averaged over all times, the acetate treatment had greater chloroform remaining than the canola oil treatment.

4. DISCUSSION

In experiment 1, the apparent decrease in headspace chloroform remaining over time for the control may be due to a difference in internal standard peak areas. The sterile acetate bottles appeared to have large methylcyclohexane peak areas relative to the control bottles, particularly at the beginning of the experiment. This would cause the calculated headspace chloroform

remaining for the control to be inflated at the beginning of the experiment and to appear to decrease over time. The apparent chloroform decrease is likely not biologically significant or meaningful. This assessment is supported by the lack of formation of degradation products. In experiment 3, the difference in headspace chloroform remaining between the acetate and canola oil treatments could be due to different amounts of chloroform or methycyclohexane adsorption on the microcosm bottles. The lack of dichloromethane and methane formation again indicates a lack of chloroform degradation.

Anaerobic chloroform biodegradation has been successful in several studies. Cometabolic reductive dechlorination was formerly thought to only occur at low chloroform concentrations; at higher concentrations, chloroform was toxic to the microbial community (Chou and Spoo 1997). More recent information indicates otherwise. Shan et al (2014) reported cometabolic fermentation of over 16.75 mM chloroform to mainly carbon monoxide, carbon dioxide, formate, and propionate. Dehalorespiration based bioremediation methods are typically preferable over those based on cometabolic processes because they are more efficient and sustainable (Grostern et al 2010). Grostern et al (2010) reported dehalorespiration of up to 500 μ M chloroform to dichloromethane by a *Dehalobacter* culture grown for 1,1,1-trichloroethane degradation. Lee et al (2012) reported complete biodegradation of chloroform concentrations as high as 360 μ M by *Dehalobacter* species. Chloroform was respired to dichloromethane, then fermented to hydrogen, carbon dioxide, and acetate. Although the chloroform concentration used in this research was higher than that used by Grostern et al (2010) or Lee et al (2012), it should not be unrealistic for microorganisms using chloroform as a growth medium to maintain activity at this increased concentration.

A lag time before chloroform biodegradation began would not be surprising. Lee et al (2012) reported lag times of 90, 141, and 155 days after amendment with 10 μ M chloroform and 25 μ M 1,2-dichloroethane. Their soil was from a site contaminated with multiple chlorinated organic compounds. All three experiments in this research ran well beyond 155 days with no indication of chloroform biodegradation. It is unlikely yet possible for biodegradation to begin in the future.

Chloroform biodegradation in this research may have been restricted by the source soils or experimental conditions. Field conditions may have prevented development of a microbial community capable of degrading chloroform. According to Chou and Spoo (1997), chlorinated organic compounds, aromatic compounds, and trace elements have potential to inhibit chloroform biodegradation. These contaminants are present at the Ellerslie site, possibly at concentrations high enough to be toxic to microorganisms capable of degrading chloroform. The

Fort Saskatchewan site is also impacted by multiple contaminants. Results from microbial community characterization of the Eilerslie site (Chapter IV) indicated that no bacteria associated with biodegradation of chlorinated organic compounds such as *Dehalobacter* and *Dehalococcoides* were enriched in the contaminated pond area. Natural attenuation of chloroform has not been reported for either site. If chloroform degrading microorganisms were present in the microcosms, conditions could have been inappropriate to encourage chloroform biodegradation. For example, the chloroform concentration may have been too high, electron donors or receptors may not have been ideal, or the nutrients may have been insufficient.

Based on the results of these experiments, bioremediation would not be effective for the Eilerslie site. Since chloroform bioremediation was not identified in experiments 1 or 3, biostimulation using an indigenous microbial community is unlikely. Since chloroform bioremediation was not identified in experiment 2, bioaugmentation using Fort Saskatchewan soil as an inoculation source is not recommended. Bioaugmentation using a different inoculation source such as a commercial culture may be possible. Results from this experiment highlight the need to develop remediation strategies which are effective on or in the presence of multiple contaminants. Although chloroform biodegradation has been successful in multiple studies, it may not be a solution for sites impacted by contaminant mixtures.

5. CONCLUSIONS

No evidence of chloroform biodegradation was found in the three anaerobic microcosm experiments conducted. The soil microbial communities may have lacked chloroform degrading microorganisms due to toxicity of co-contaminants or experimental conditions may not have been appropriate to support biodegradation. Biostimulation and bioaugmentation will not be effective remediation techniques for the Eilerslie site.

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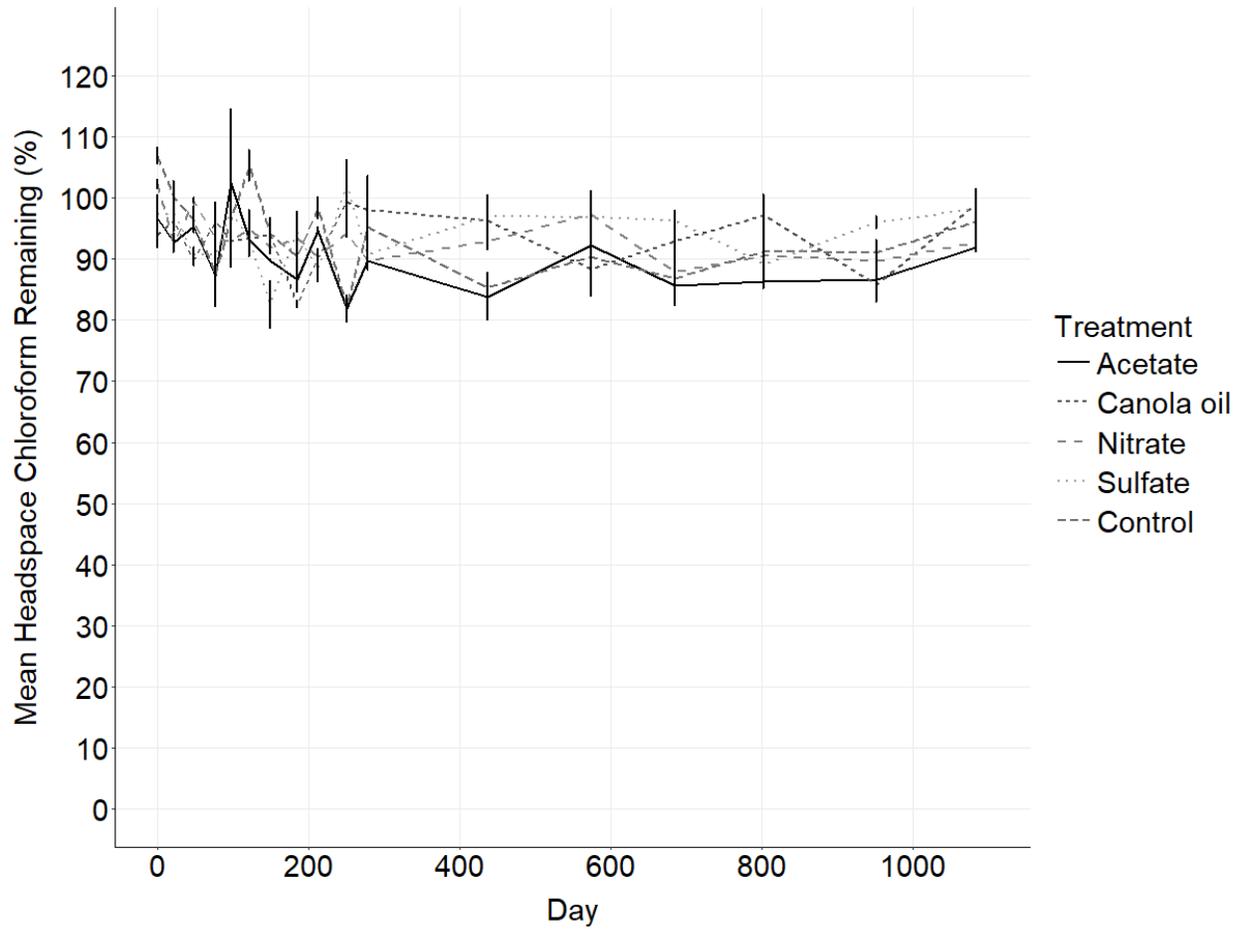


Figure 3.1. Experiment 1 change in chloroform over time. Error bars indicate standard error of the mean.

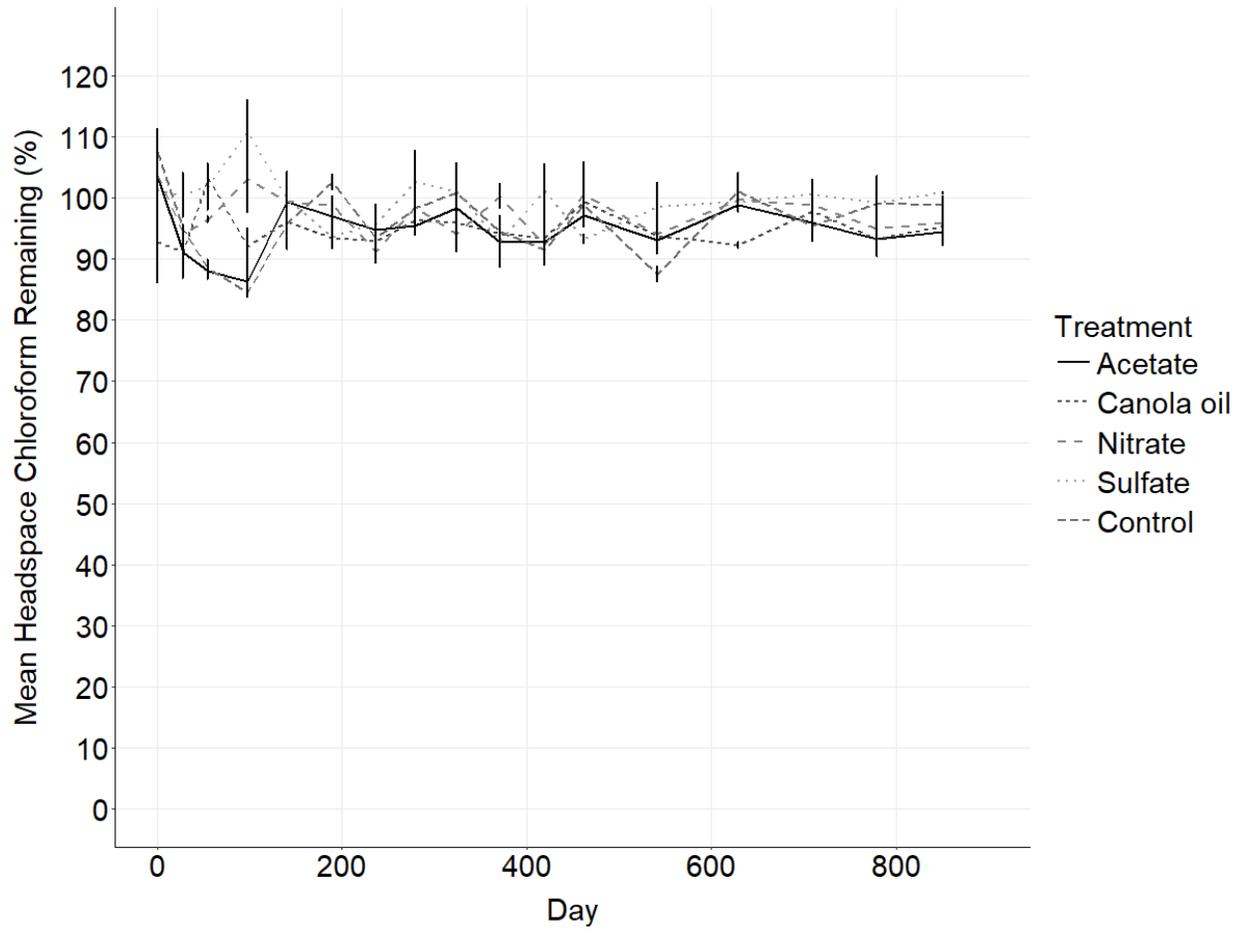


Figure 3.2. Experiment 2 change in chloroform over time. Error bars indicate standard error of the mean.

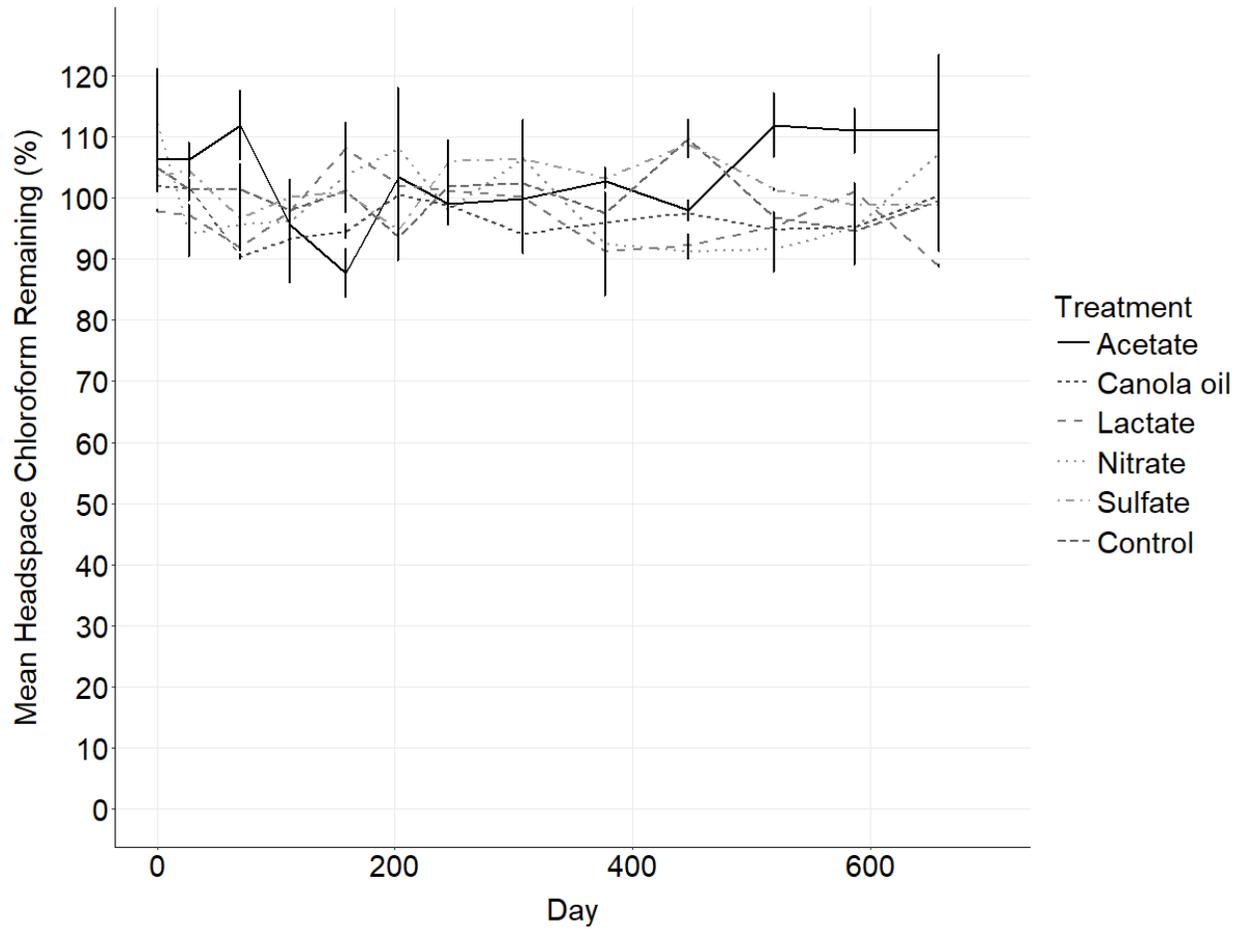


Figure 3.3. Experiment 3 change in chloroform over time. Error bars indicate standard error of the mean.

IV. MICROBIAL COMMUNITY CHARACTERIZATION FOR A SITE CONTAMINATED BY CHLORINATED ORGANIC COMPOUNDS AND SALTS

1. INTRODUCTION

Chloroform (CHCl_3) is a volatile solvent produced naturally by soil and oceanic processes (McCulloch 2003) and anthropogenically for industrial purposes such as production of plastics, refrigerants, pharmaceuticals, pesticides, dyes, and aerosol propellants (Chou and Spoo 1997, Canadian Council of Ministers of the Environment 1999). Dichloromethane (CH_2Cl_2) is a volatile solvent used in paint and furniture strippers, solvents, pesticides, metal and automotive cleaners, and aerosol sprays and in production of foams and photographic film (United States Environmental Protection Agency 1994, Wilson et al 2000). These chlorinated organic compounds can cause neurological, cardiovascular, liver, and kidney damage; chloroform is a probable human carcinogen (United States Environmental Protection Agency 1994, Chou and Spoo 1997, Armstrong and Green 2004, World Health Organization 2004).

Chlorinated organic compounds can be biodegraded under aerobic and anaerobic conditions (Cappelletti et al 2012). Aerobic chlorinated methane degradation typically results in complete conversion of organic chlorine to chloride ions via a cometabolic process in which microorganisms require a separate carbon source. However, Dey and Roy (2011) reported aerobic degradation by *Bacillus* bacteria which used chloroform as a growth substrate. Under anaerobic conditions, cometabolic chlorinated methane degradation is often incomplete, resulting in accumulation of potentially toxic degradation products. However, anaerobic chlorinated methane degradation via dehalorespiration can be complete as reported by Lee et al (2012) in their research on chloroform degradation by *Dehalobacter* bacteria. Biodegradation of chloroform and dichloromethane is useful for active bioremediation techniques and for passive natural attenuation.

Chlorinated methanes are often present at sites with multiple contaminants (Grostern et al 2010). In Canada, halogenated hydrocarbons are found at 157 federal contaminated sites, many of which also have other contaminants (Treasury Board of Canada Secretariat 2019). According to Chou and Spoo (1997), co-contaminants such as chlorinated organic compounds, aromatic compounds, and trace elements can inhibit chloroform biodegradation. Gao et al (2015) reported that salinity and hydrocarbons both impacted the soil microbial community at an oilfield site. Research on microbial communities at sites impacted by chlorinated organic compounds and salinity is lacking. Such research is a necessary step towards improving our

understanding of chloroform bioremediation in realistic settings that often involve complex contaminant mixtures.

The objective of the study was to assess the soil microbial community at a site impacted by multiple contaminants including chloroform, dichloromethane, and salts. Specific objectives were to compare the microbial communities within and outside of the highly contaminated area and identify genera associated with chloroform and dichloromethane degradation.

2. MATERIALS AND METHODS

2.1. Research Site

Soil was collected from the University of Alberta's former Ellerslie Waste Management Facility in Edmonton, Alberta (Appendix). The approximately 0.25 ha Ellerslie site is located within the Province of Alberta's Edmonton Transportation Utility Corridor with surroundings zoned for agriculture (Alberta Infrastructure 2003, AMEC Earth and Environmental 2010). A major road is located immediately north and residential areas are north and east. Whitemud Creek is located approximately 450 m north. The site processed laboratory waste from 1972 to 2007 (Stantec Consulting Limited 2013); activities included solvent incineration, acid dilution and neutralization, and temporary waste storage. Prior to 1983, a waste water pond leaked (University of Alberta 1984). Primary contaminants included chloroform, dichloromethane, and chloride; an additional 15 soil and 34 ground water parameters were identified in exceedance of Alberta Tier 1 soil and ground water remediation guidelines for fine textured soils and agricultural land use (Alberta Environment and Parks 2016). The site is located in the Central Parkland natural subregion (Natural Regions Committee 2006). Surficial geology consists mainly of clay and clay till (Stantec Consulting Limited 2010). Much of the contamination is considered under anaerobic conditions because of its depth relative to the water table and restricted oxygen flow in fine textured soils.

2.2. Soil Sampling And Analyses

Soil was sampled from nine bore holes at the Ellerslie site by direct push drilling in January and February 2016 (Figure 4.1). Three bore holes (16-02-08, 16-10-06, and 16-11-06) were background locations up gradient of the contaminated waste water pond area, three (16-05-09, 16-06-06, and 16-07-06) were within the pond area, and three (16-01-11, 16-08-06, and 16-09-06) were down gradient of the pond area. Bore hole depth was 6 to 11 m, indicated by the final two digits in each bore hole name. In the field, soil cores were cut into segments using an

ethanol sterilized hack saw, then each segment was wrapped in plastic held in place using duct tape to minimize cross contamination and exposure to oxygen. Samples were stored in a freezer at -20 °C until analysis.

Composite soil samples from two microcosm experiments were included to complement research on anaerobic chloroform biodegradation under laboratory conditions. One sample, collected in 2014, contained soil from the 5 to 6 m depth of the Eilerslie pond area. The other, sample collected in 2015, contained soil from a secondary contaminated site in Fort Saskatchewan, Alberta.

Duplicate subsamples for each bore hole were taken every metre from 3 m to the bottom of the bore hole. For bore hole 16-01-11, no samples were taken for 9 or 11 m depths as only a limited number of samples could undergo deoxyribonucleic acid (DNA) extraction. In the laboratory, 1 cm of soil was removed from the core to expose soil which had not touched the core liner. This exposed soil was scraped and 0.5 g was placed into tubes. For composite samples, a random subsample was taken.

Cells were lysed and DNA was extracted using the MP Biomedicals FastDNA™ SPIN Kit for soil according to manufacturer recommendations. Polymerase chain reaction (PCR) amplification of the V3-V4 hypervariable regions of prokaryotic 16S rRNA genes was conducted using New England Biolabs Q5 high fidelity DNA polymerase and primers 515F (5'-GTGCCAGCMGCCGCGTAA-3') and 806R (5'-GGACTACVSGGGTATCTAAT-3'). Fungal internal transcribed spacer (ITS) segments were amplified using primers ITS1 (5'-TCCGTAGGTGAACCTGCGG-3') and ITS2 (5'-GCTGCGTTCTTCATCGATGC-3'). Forward and reverse primers were tagged with adapter, pad, and linker sequences. Barcode sequences (12 mer) were added to reverse primers to pool multiple samples in one Miseq sequencing run. Thermal cycling conditions were 94 °C for 2 min, 30 cycles at 94 °C for 0.5 min, 50 °C for 0.5 min, 72 °C for 1 min extension, and 72 °C for 6 min. PCR products were purified and combined in equimolar ratios with the quantitative DNA binding method to create a DNA pool for sequencing from the adapter. The Illumina MiSeq platform was used to sequence 16S rRNA gene fragments.

2.3. Data Analyses

Sequences were sorted by barcode tag and adapters and primers were removed. Low quality sequences (with expected error threshold = 1) were removed. Operational taxonomic units (OTUs) were defined and assigned to taxa using the UPARSE pipeline process (Edgar 2013).

All data analyses were completed using R software. The community matrix was normalized using the DESeq package. Alpha diversities were calculated using the phyloseq package (McMurdie and Holmes 2013). Non metric multidimensional scaling (NMDS) for ordination was based on the UniFrac phylogenetic distance matrix for community structure. Relative abundance of the various genera was plotted. Microbial communities were then compared between sampling locations and depths.

3. RESULTS

DNA sequencing for bacteria was successful for 46 of 94 soil samples. DNA sequencing was not successful for any depth greater than 6 m. For samples from 3 to 6 m depths, there was no clear association between sample location or depth and sequencing success. DNA sequencing for fungi was successful for 86 of 94 samples. The only sample for which both duplicates failed fungal DNA sequencing was the Fort Saskatchewan microcosm composite sample.

Bacterial richness was highest (mean count 2128) in samples from upgradient of the pond area at the 3 m depth (Figure 4.2). Within each depth, mean richness was lower in samples from the pond area than in samples upgradient or downgradient. The same trend was shown for bacterial Shannon and Simpson diversity indices. Fungal richness was highest (mean count 327) in samples from upgradient of the pond area at 3 m depth (Figure 4.3). Samples from 7 to 10 m depth had relatively low mean richness and variation. Fungal Shannon and Simpson diversity indices for samples from the pond area were slightly lower than in samples from upgradient or downgradient at 5 and 6 m depths.

NMDS showed that soil depth and location may affect the composition of the bacterial community (Figure 4.4). Communities from upgradient and downgradient locations were more similar to each other than they were to communities from the pond area. A gradient by sample depth appears along the secondary axis. NMDS for the fungal community showed substantial overlap (Figure 4.5).

Bacterial communities from the 6 m depth of the pond area had greatest relative abundance of the genera *Geobacter* and *Desulfosporosinus* (Figure 4.6). Relative abundance of *Desulfuromonas* and *Saccharibacteria* genera *incertae sedis* increased at 5 and 6 m depths of the pond area. Relative abundance of *Anaeromyxobacter*, *Microgenomates* genera *incertae sedis*, *Pelotomaculum*, and *Rubrobacter* decreased at 5 and 6 m depths of the pond area. Fungal communities had no clear patterns with sampling location or depth (Figure 4.7).

4. DISCUSSION

Why fungal DNA sequencing was more successful than bacterial DNA sequencing is unknown. The various differences found between the bacterial and fungal communities are likely reflective of their response to soil conditions. The Ellerslie soil bacterial community may be more sensitive than the fungal community as it was less similar between contaminated and uncontaminated locations. Bacterial diversity decreased in the pond area from 3 to 6 m and fungal diversity slightly decreased in the pond area from 5 to 6 m. Decreased diversity could be caused by soil and ground water contamination. Since a complex mixture of contaminants is present in the Ellerslie pond area, it is not known which contaminant or combination of contaminants had the greatest impact on the microbial community. Fritze et al (1996) reported that microbial biomass and microbial activity decreased with soil contamination from a smelter including copper, zinc, nickel, cadmium, lead, iron, and sulfur. Muller et al (2001) found decreased microbial biomass with soil mercury contamination; they did not find a significant decrease in fungal biomass.

Variation in the Ellerslie soil bacterial community composition with soil depth and location is likely due to higher contaminant concentrations within the pond area at 5 to 6 m depths relative to 3 to 4 m depths. The bacteria with increased relative abundances in the pond area are not known to degrade chlorinated methanes (Cappelletti et al 2012). Therefore, their presence may be due to their ability to tolerate rather than their ability to bioremediate chloroform and/or dichloromethane. During decommissioning of the former waste water pond in 1986, some contaminated soil would have been removed and replaced with clean fill soil. The origin of the fill soil is not fully known. Its placement would have introduced a different microbial community and contributed to differences between the soil communities at the Ellerslie site.

Geobacter species, enriched in the pond area, are anaerobic bacteria associated with iron(III) and manganese(IV) reduction (Lovley et al 2011). Although they are not known for chlorinated methane degradation, some participate in anaerobic oxidation of aromatic hydrocarbons, including benzene. Benzene is a co-contaminant at the Ellerslie site so the increased relative abundance of *Geobacter* in the contaminated pond area indicates that benzene biodegradation may be occurring.

Desulfosporosinus and *Desulfuromonas* species are strictly anaerobic bacteria which reduce sulfur (Stackerbrandt et al 1997, Kuever et al 2015). Their increased relative abundances in the pond area indicate that sulfur reducing conditions are likely present in at least some locations or micro sites. This has important implications for bioremediation efforts since it specifies

environmental conditions under which indigenous or inoculation microorganisms capable of contaminant biodegradation must be active. *Desulfuromonas* have been isolated from both saline and freshwater sediments; their increased relative abundance in the pond area could be due to their ability to tolerate salt contamination.

The bacteria with decreased relative abundances in the pond area may be sensitive to one or more of the contaminants present. The Ellerslie soil fungal community composition did not exhibit clear trends with soil depth or with location. This supports the NMDS results from our study showing that fungal communities were similar across the site. Fungi have been linked to chloroform production but not degradation (Hoeskstra et al 1998). The lack of significant populations of known chlorinated methane degraders indicates that natural attenuation of those contaminants is unlikely at this site.

5. CONCLUSIONS

The complex contaminant mixture at the former Ellerslie Waste Management Facility impacted the soil bacterial community more than the fungal community. Decreased diversity in the contaminated pond area may be caused by toxic effects of one or more contaminants. The bacterial community composition was impacted by soil depth and location, likely due to the contaminant concentration gradients. Of the bacteria enriched in the pond area, none are known to degrade chlorinated methanes; however, one may be capable of benzene degradation. Natural attenuation of chloroform and dichloromethane at Ellerslie is likely not a realistic remediation strategy.

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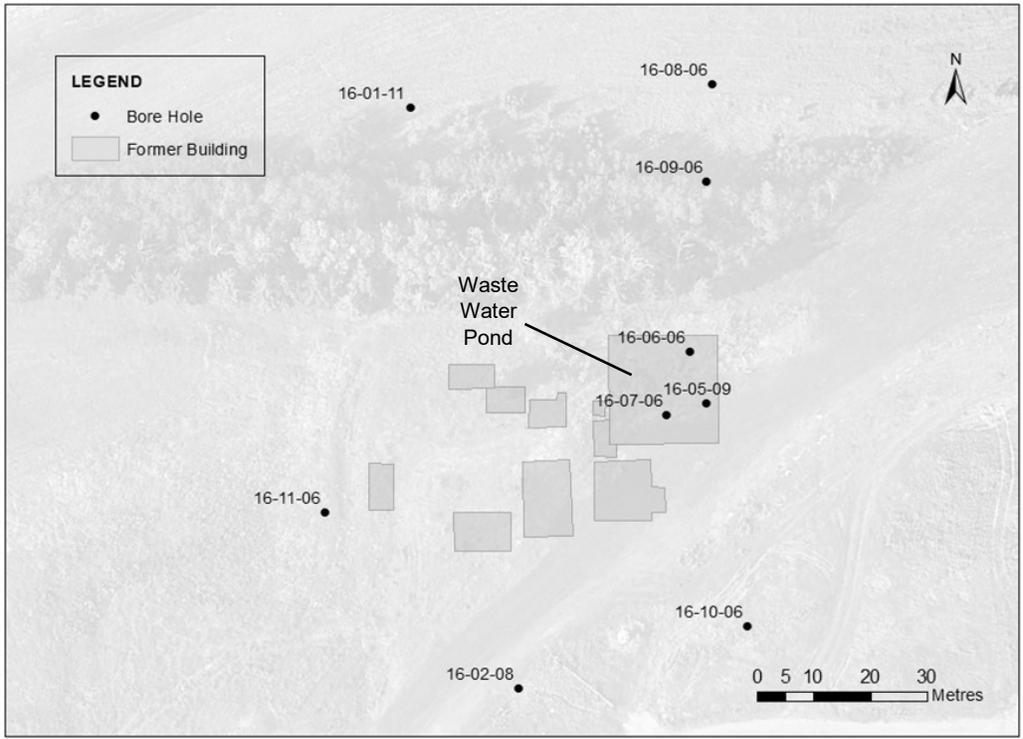


Figure 4.1. Ellerslie Waste Management Facility soil sampling locations.

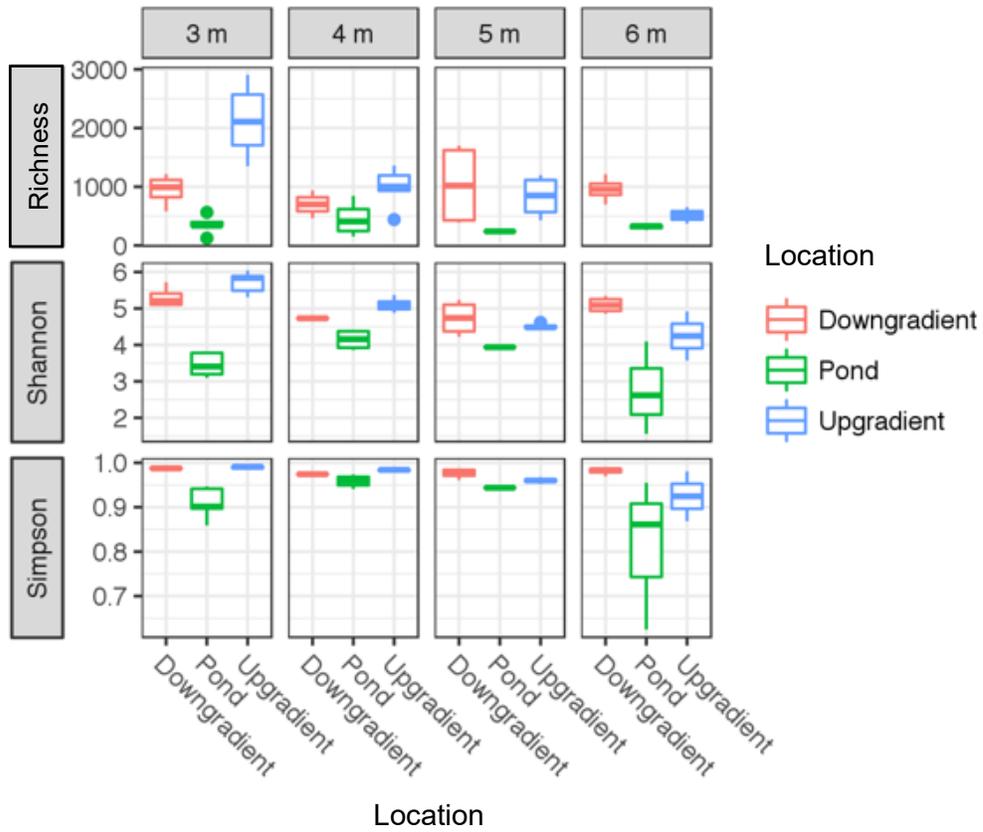


Figure 4.2. Ellerslie soil bacterial community richness and diversity by sampling location and depth.

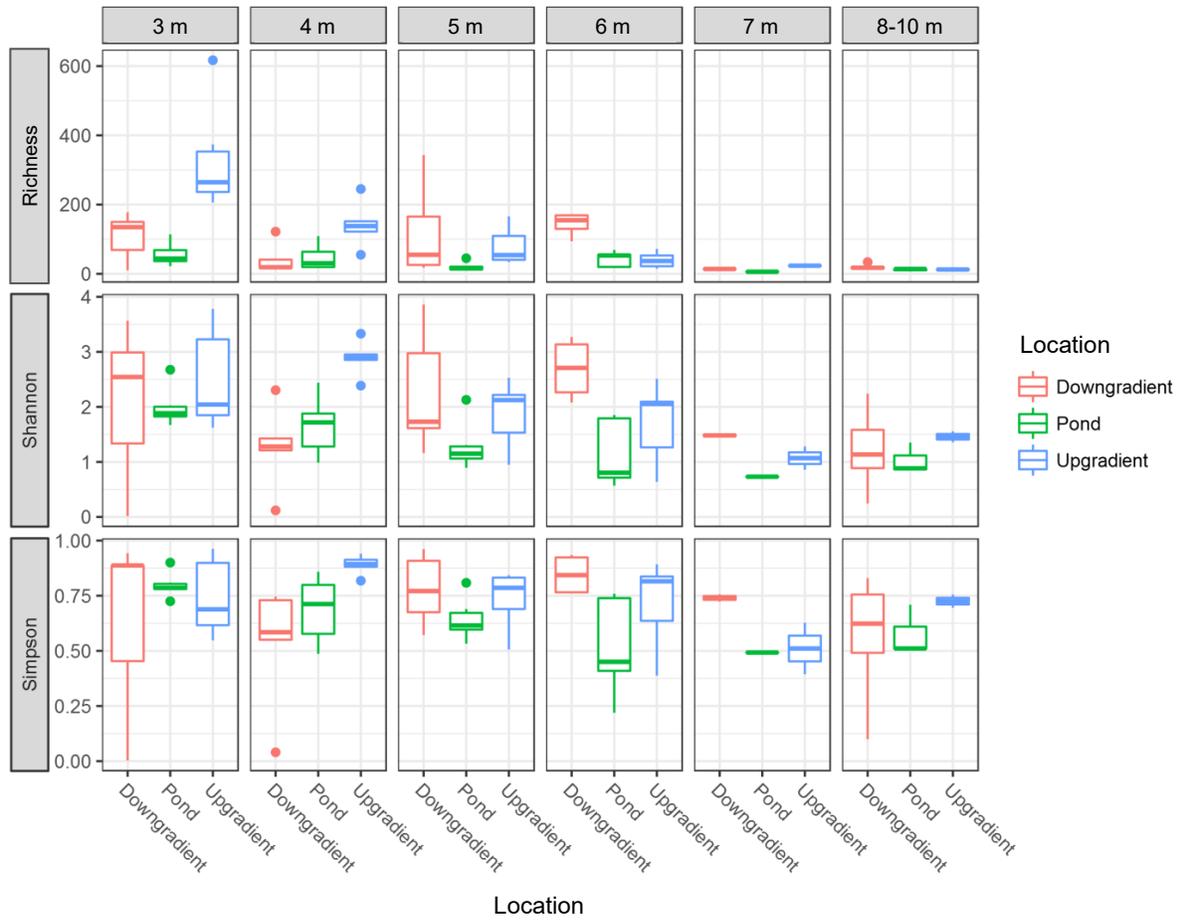


Figure 4.3. Ellerslie soil fungal community richness and diversity by sampling location and depth.

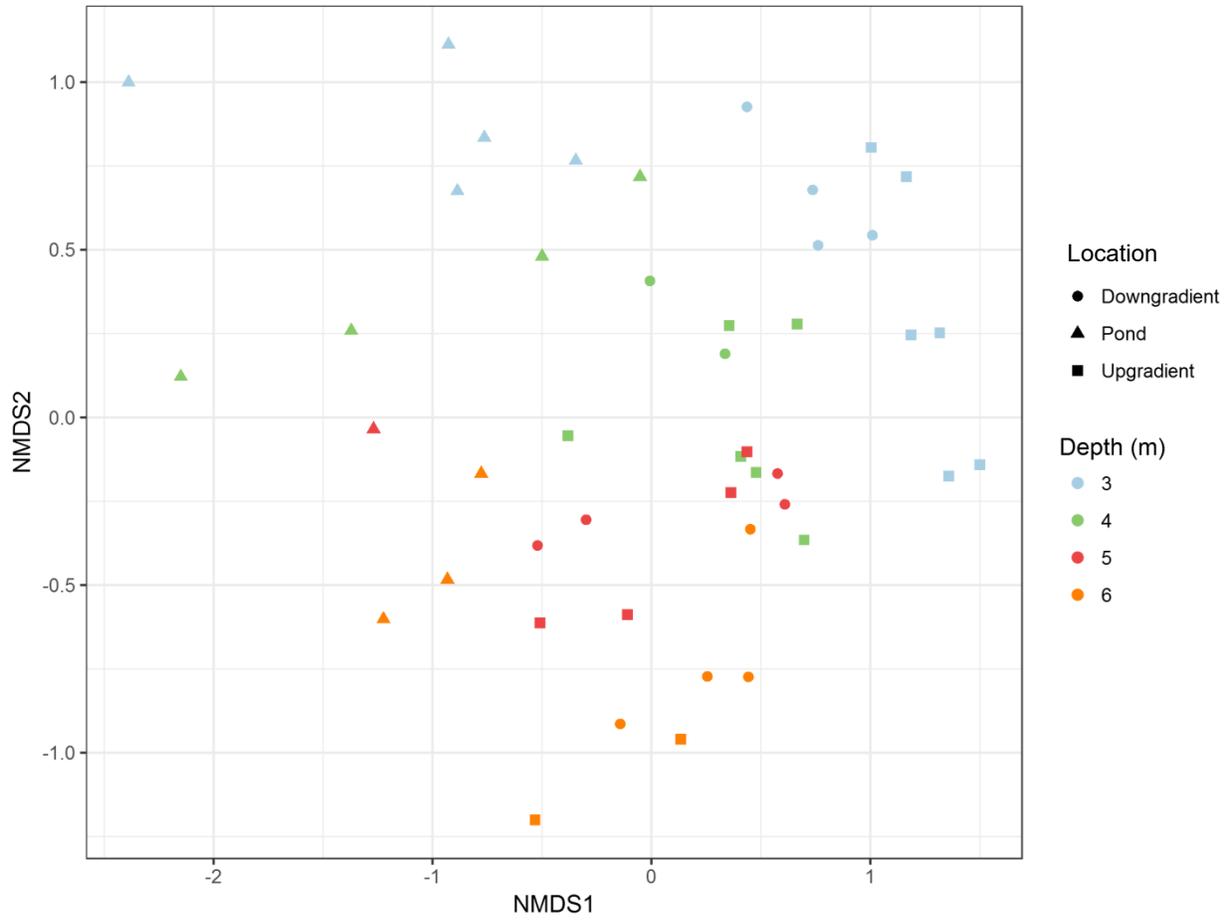


Figure 4.4. Ellerslie soil bacterial community non metric multidimensional scaling analysis.

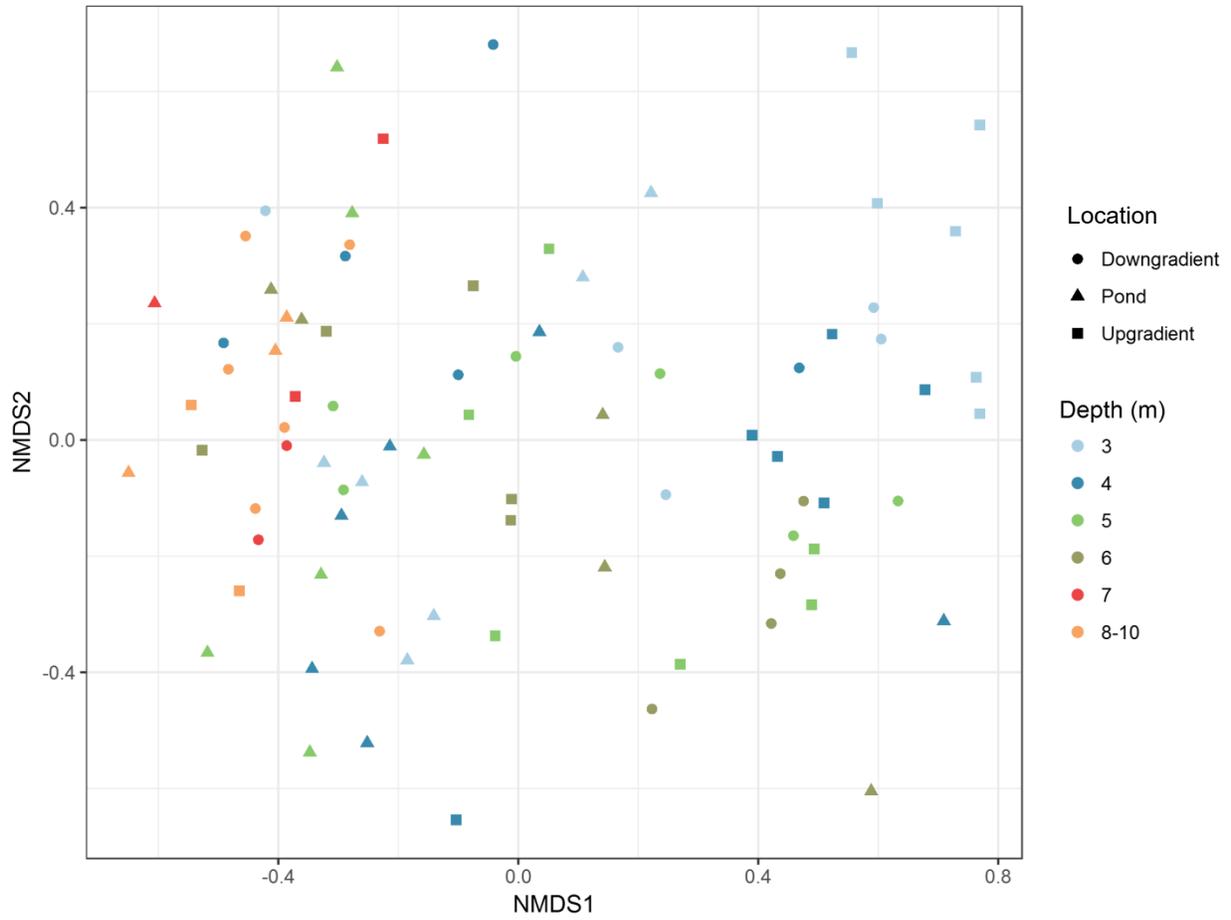


Figure 4.5. Ellerslie soil fungal community non metric multidimensional scaling analysis.

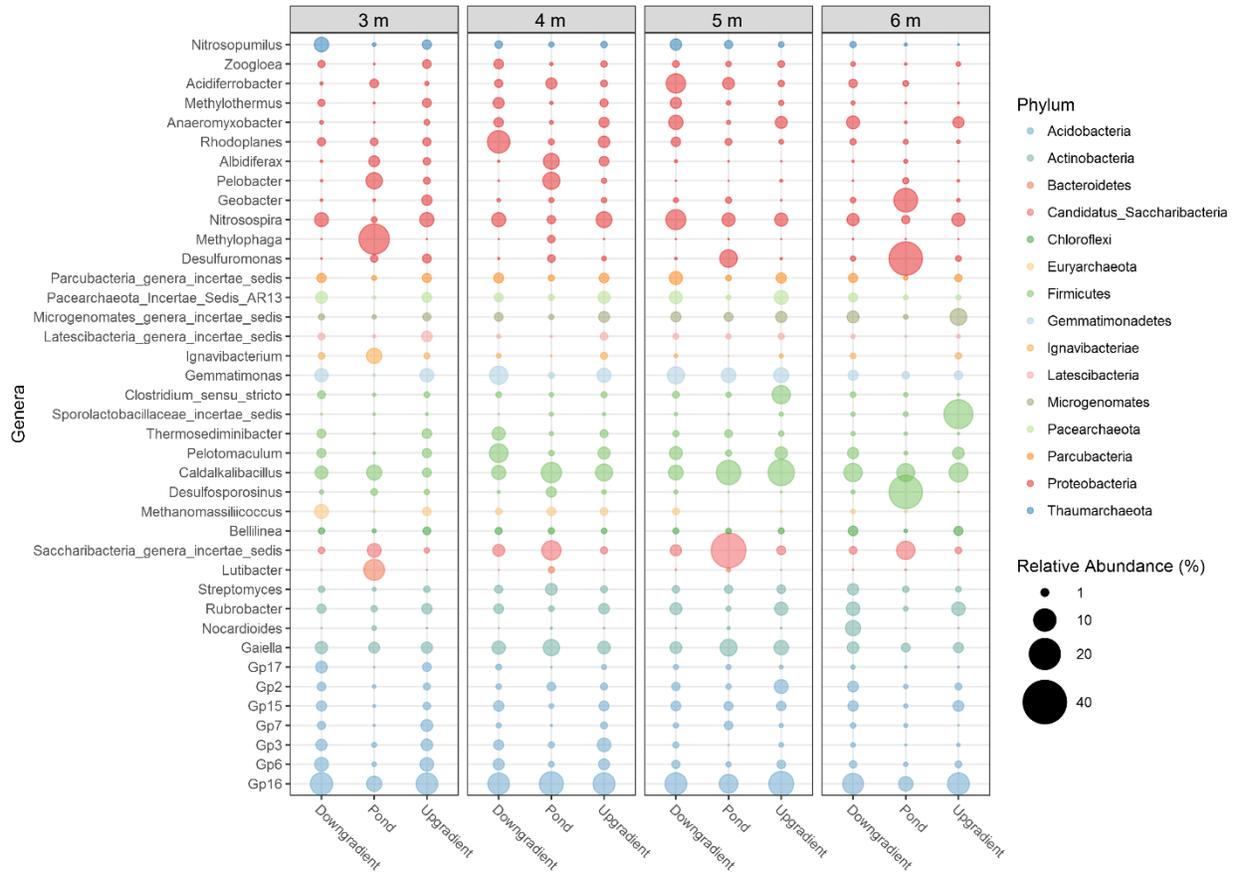


Figure 4.6. Ellerslie soil bacterial community composition.

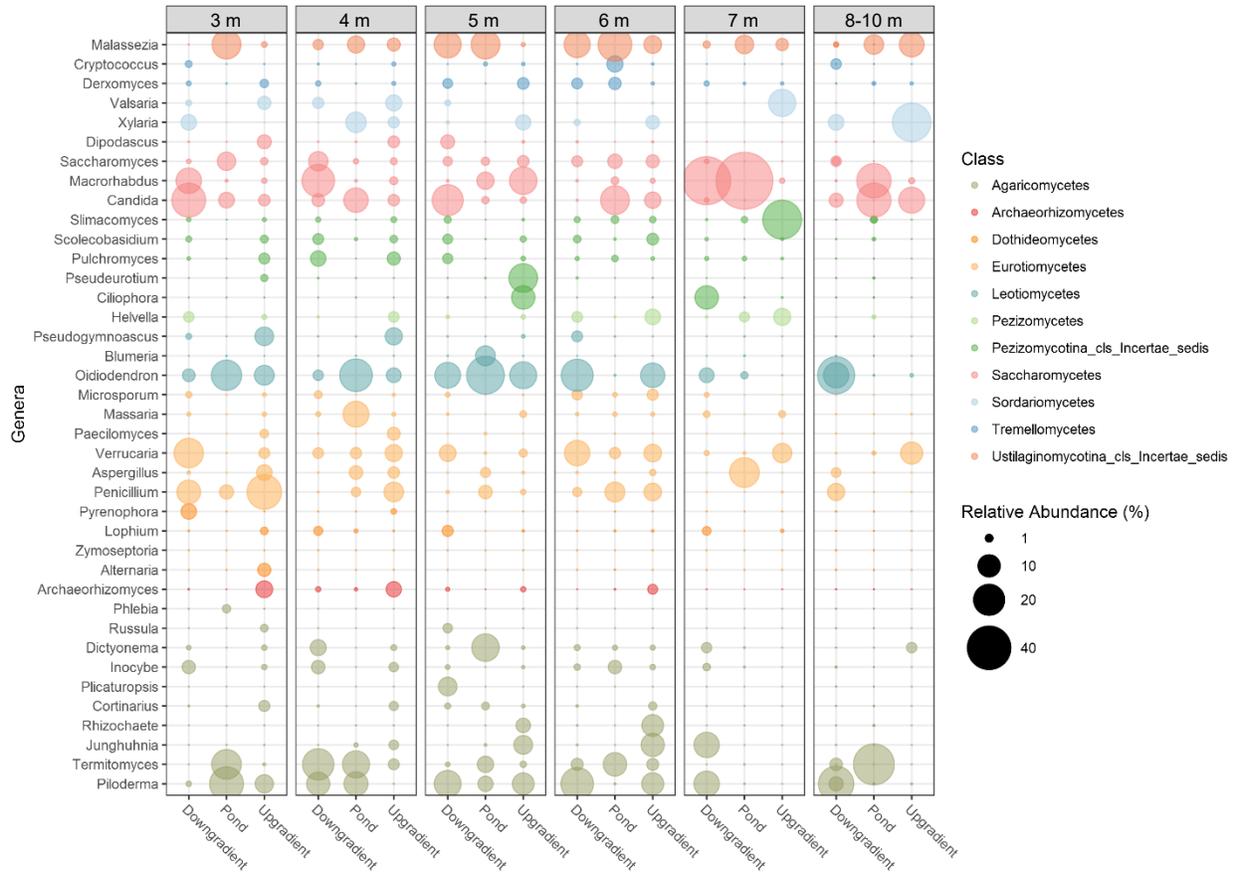


Figure 4.7. Ellerslie soil fungal community composition.

V. REMEDIATION OF CHLOROFORM IN SOILS BY CHEMICAL REDUCTION USING ZERO VALENT IRON IN ANAEROBIC BOTTLES

1. INTRODUCTION

Chloroform (CHCl_3) is a volatile solvent produced naturally by soil and oceanic processes (McCulloch 2003), and anthropogenically for industrial purposes such as production of plastics, refrigerants, pharmaceuticals, pesticides, dyes, and aerosol propellants (Chou and Spoo 1997, Canadian Council of Ministers of the Environment 1999). Since chloroform is a probable human carcinogen and causes cardiovascular, liver, and kidney damage (Chou and Spoo 1997, World Health Organization 2004), environmental limits have been set. Alberta Tier 1 remediation guidelines for chloroform in fine textured soils are $0.0029 \text{ mg kg}^{-1}$ soil and 0.0018 mg L^{-1} ground water (Alberta Environment and Parks 2016). The international drinking water guideline is 0.3 mg L^{-1} (World Health Organization 2011). Chloroform is a common contaminant found at over 700 of 1,430 national priority list sites in the United States of America (Agency for Toxic Substances and Disease Registry 2014).

Remediation of contaminated media using zero valent iron (Fe^0) has shown great potential since its emergence in the 1990s (Gillham and O'Hannesin 1994). Zero valent iron is abundant, inexpensive, non toxic, and relatively easy to produce (Fu et al 2014). It is an effective reducing agent for chemical remediation; electrons are transferred from zero valent iron to oxidized contaminants in a redox reaction which degrades the contaminant to a non toxic or less toxic form. When zero valent iron is used in the presence of dissolved oxygen, hydrogen peroxide and hydroxyl radicals are produced which can oxidize reduced contaminants. Sub colloidal nano scale zero valent iron particles are advantageous in remediation due to their high specific surface area available for reaction (Xiaoqin et al 2006). Their disadvantage is cost and tendency to form aggregates. Zero valent iron on negatively charged support materials can reduce aggregation and interaction with net negatively charged soil minerals, thus increasing dispersion. Noble or catalytic metals such as palladium, nickel, platinum, and silver combined with zero valent iron can increase reaction rate.

Zero valent iron treatment is considered applicable to a variety of contaminants including chlorinated organic compounds, nitroaromatic compounds, trace elements, nitrate, dyes, and phenol (Xiaoqin et al 2006, Fu et al 2014). Much research has been conducted in recent years on the use of zero valent iron for remediation of chlorinated organic compounds. However, most studies investigate remediation of chlorinated ethenes such as perchloroethene and

trichloroethene. Relatively little research information is available on remediation of chloroform with zero valent iron. However, research on carbon tetrachloride degradation using zero valent iron has shown a degradation pathway which includes chloroform as an intermediary degradation product.

Feng and Lim (2005) reported successful reduction of carbon tetrachloride using nano scale zero valent iron, nano scale zero valent iron and nickel, micro scale zero valent iron, and micro scale zero valent zinc. The nano scale amendments had higher surface areas and quicker carbon tetrachloride removal. Treatment with micro scale amendments resulted in accumulation of dichloromethane. Zhang et al (2010) studied carbon tetrachloride degradation using micro scale zero valent iron and reported that addition of organic ligands, such as ethylenediaminetetraacetic acid, citric acid, tartaric acid, malic acid, and oxalic acid increased the rate of carbon tetrachloride degradation. The organic ligands chelated iron(III), which promoted exposure of active sites throughout the experiment. Shackelford et al (2005) used a combination of zero valent iron and kaolinite clay for in situ treatment of a site contaminated with carbon tetrachloride, perchloroethene, trichloroethene, and dichloromethane. Clay was added to slow the movement of contaminated ground water through the soil and encourage homogeneous distribution of zero valent iron. After one year, over 99 % of total chlorinated organic compounds were removed.

The objective of this research was to investigate the potential for chloroform remediation in soils by anaerobic chemical reduction using zero valent iron. Specific objectives were to identify chloroform concentration reduction and degradation product formation and assess reaction timeframe under various conditions.

2. MATERIALS AND METHODS

2.1. Research Site

Soil was collected from the former University of Alberta Ellerslie Waste Management Facility in Edmonton, Alberta (Appendix). The approximately 0.25 ha Ellerslie site is located within the Province of Alberta's Edmonton Transportation Utility Corridor with surroundings zoned for agriculture (Alberta Infrastructure 2003, AMEC Earth and Environmental 2010). A major road is located north and residential areas are north and east. Whitemud Creek is approximately 450 m north. The site processed laboratory waste from 1972 to 2007 (Stantec Consulting Limited 2013); activities included solvent incineration, acid dilution and neutralization, and temporary

waste storage. Prior to 1983, a waste water pond leaked (University of Alberta 1984). Primary contaminants were chloroform, dichloromethane, and chloride; another 15 soil and 34 ground water parameters were identified in exceedance of Alberta Tier 1 soil and ground water remediation guidelines for fine textured soils and agricultural land use (Alberta Environment and Parks 2016). The site is located in the Central Parkland natural subregion (Natural Regions Committee 2006). Surficial geology at the site consists mainly of clay and clay till (Stantec Consulting Limited 2010). Much of the contamination is considered under anaerobic conditions because of its depth relative to the water table and restricted oxygen flow in fine textured soils.

2.2. Reaction Bottles

Soil for the experiment was collected from the contaminated former waste water pond area at the Eilerslie site in 2014 by direct push drilling. The soil was air dried, mechanically ground, and then passed through a 2 mm sieve. A composite sample was prepared by mixing soil from depths of 3 to 7 m below surface. Soils from these depths were silty clay loam texture with 32 to 38 % clay and 2 to 9 % sand.

Reaction containers were prepared in an anaerobic chamber using 38 mL glass bottles. Each bottle contained 10 mL distilled water and either 5 mL glass beads, 2.5 mL glass beads and 2.5 mL soil, 2.5 mL glass beads and 2.5 mL zero valent iron, or 2.5 mL soil and 2.5 mL zero valent iron. The zero valent iron was Ferox-Flow™, a micro scale cast iron powder from a commercial source (Hepure Technologies Incorporated 2014). All materials were autoclaved to prevent chloroform degradation by microbial activity. Each bottle was sealed with a butyl stopper and aluminum cap, then each bottle was amended with 20 µL chloroform for an initial concentration of 1,973 mg chloroform L⁻¹ slurry. Bottles were stored on a mechanical shaker in the dark to encourage even mixing and prevent chloroform degradation by photoreaction.

Headspace analysis for chloroform, dichloromethane, and methane was completed using a gas chromatograph with a flame ionization detector. A 50 µL headspace sample was collected for analysis at 20 specific time points (days 0, 0.25, 0.5, 1.1, 1.5, 2, 3, 4, 5, 6, 7, 10, 13, 32, 54, 79, 115, 153, 191, and 230). Dichloromethane and methane were the expected chloroform degradation products. Calibration was completed using bottles containing 10 mL distilled water, 5 mL glass beads, and either 987 mg L⁻¹ chloroform, 880 mg L⁻¹ dichloromethane, and 10 µL L⁻¹ methane; 1,973 mg L⁻¹ chloroform, 1,760 mg L⁻¹ dichloromethane, and 42 µL L⁻¹ methane; 2,960 mg L⁻¹ chloroform, 2,640 mg L⁻¹ dichloromethane, and 87 µL L⁻¹ methane; or 3,947 mg L⁻¹ chloroform, 3,520 mg L⁻¹ dichloromethane, and 178 µL L⁻¹ methane.

2.3. Data Analyses

Calibration curves were prepared for chloroform, dichloromethane, and methane by plotting gas chromatography peak areas against their concentrations. Sample concentrations were calculated using the calibration lines of best fit. If no peak was found or if the calculated concentration was less than zero, the concentration was then assumed to be zero. For each experimental treatment, mean chloroform, dichloromethane, and methane concentrations were plotted over time. Repeated measures analysis of variance was unsuccessful since chloroform, dichloromethane, and methane concentrations did not respond linearly to changes in time. Non linear models were not used because they would differ between treatments and comparison would not be appropriate. Instead, trends were compared visually.

3. RESULTS

Plots of headspace chloroform, dichloromethane, and methane over time showed clear differences between the treatments. Chloroform was removed, and the degradation products dichloromethane and methane were produced in both treatments that included additions of the zero valent iron.

Mean headspace chloroform concentration decreased rapidly in treatments including zero valent iron (Figure 5.1). Chloroform was completely removed by days 0.5 and 2 in the soil zero valent iron and zero valent iron treatments, respectively. Mean headspace chloroform concentration appeared to decrease slowly in the soil and control treatments; the most rapid decrease occurred in the first two weeks.

Headspace dichloromethane was detected in the soil zero valent iron treatment by day 0.25; it had been completely removed by day 2 (Figure 5.2). Headspace dichloromethane was detected in the zero valent iron treatment by day 0.5; it was completely removed by day 2. Maximum mean headspace dichloromethane concentration was 553 and 686 mg L⁻¹ in the soil zero valent iron and zero valent iron treatments, respectively. No headspace dichloromethane was detected in the soil and control treatments.

Headspace methane in the soil zero valent iron treatment was detected by day 0.25, increased until day 32, and then decreased to zero by day 230 (Figure 5.3). Headspace methane in the zero valent iron treatment was detected by day 0.25 and increased or remained throughout the entire experiment. Maximum mean headspace methane concentration was 24 and 112 $\mu\text{L L}^{-1}$ in

the soil zero valent iron and zero valent iron treatments, respectively. No headspace methane was detected in the soil and control treatments.

4. DISCUSSION

Results from this study support anaerobic chloroform reduction using zero valent iron as a promising remediation technique for contaminated soils, including co-contaminated soils. The reaction between chloroform and zero valent iron produces dichloromethane and methane, with rapid and complete chloroform and dichloromethane removal.

Zero valent iron was highly effective in reducing chloroform in the reaction bottles. The chemical reaction occurred quickly; no headspace chloroform was detected in the soil zero valent iron or zero valent iron bottles after 1.5 days. Chloroform in the slurry phase was not measured, but it is presumed to have been completely removed since reaction between chloroform and zero valent iron in this phase would continuously draw chloroform from the headspace. The much slower decrease in headspace chloroform in the soil and control treatments was likely not caused by chloroform degradation. No zero valent iron with which the chloroform could react was present in those treatments. Materials were sterilized to prevent microbial degradation. The decline may have been caused by chloroform sorption to the bottle and to the stopper.

Detection of headspace dichloromethane in both the soil zero valent iron and the zero valent iron treatments indicates that chloroform was reduced to dichloromethane when it reacted with zero valent iron. The reaction pathway $\text{CHCl}_3 + \text{e}^- \rightarrow \cdot\text{CHCl}_2 + \text{Cl}^-$ and $\cdot\text{CHCl}_2 + \text{e}^- + \text{H}^+ \rightarrow \text{CHCl}_2$ was proposed by Feng and Lim (2005). The timing of dichloromethane production and removal shows that it is produced at the same time as chloroform is removed, and is removed quickly as further degradation occurs. Lack of headspace dichloromethane in the soil and control treatments indicates the apparent decrease in chloroform in those treatments was not caused by chloroform degradation.

Detection of headspace methane in both the soil zero valent iron and the zero valent iron treatments indicates that after chloroform was reduced to dichloromethane, it was further reduced to methane. This outcome is desirable in a remediation technique because if chloroform was only degraded to dichloromethane, contamination would still be of concern. Methane production began at the same time as chloroform was being degraded. Headspace methane concentration continued to increase after headspace chloroform and dichloromethane were no longer detected, possibly due to desorption and degradation of chloroform and

dichloromethane which had initially adsorbed to soil and bottle surfaces. It is unknown why the maximum headspace methane concentration was higher in the zero valent iron treatment than in the soil zero valent iron treatment. It is also unknown why the headspace methane was removed over time in the soil zero valent iron treatment. One thought is that the presence of soil in some way contributed to methane adsorption. The lack of headspace methane in the soil and in the control treatments further supports the conclusion that the apparent decrease in chloroform in those treatments was not caused by chloroform degradation.

Successful chloroform reduction using zero valent iron has been previously reported under laboratory and field conditions which focused on carbon tetrachloride (Feng and Lim 2005, Shackelford et al 2005, Zhang et al 2010). However, unlike in this experiment, incomplete removal of the degradation product dichloromethane was found. Shackelford et al (2005) noted that further degradation product removal may occur with time and that dichloromethane can undergo biodegradation. The dichloromethane accumulation in the experiment by Zhang et al (2010) may be at least partially attributed to the short 8 h experimental duration. Chloroform reaction time observed in this experiment is similar to that reported by Feng and Lim (2005). They found chloroform was reduced to 8 % of its original concentration after approximately 50 h when treated with micro scale zero valent iron.

Zero valent iron treatment could be applied at the former Ellerslie Waste Management Facility as an in situ or as an ex situ remediation technique. If used for in situ remediation, zero valent iron could be injected into the contaminated zone where it would react. It could also be installed as a permeable reactive barrier to prevent chloroform from spreading with ground water flow. If used ex situ, zero valent iron could be added to the soil and ground water which had been excavated and collected. This treatment could occur on the site directly, or on a remediation treatment site. A batch reactor would be required to maintain anaerobic conditions so that the zero valent iron would not react with oxygen. Zero valent iron could also be used for remediation of arsenic, chromium, and lead (Fu et al 2014); all of which are co-contaminants at the Ellerslie site. Therefore, zero valent iron has the additional benefit of simultaneously treating multiple contaminants at the Ellerslie site and other similar sites.

5. CONCLUSIONS

Addition of zero valent iron caused the complete reduction of chloroform to methane. Dichloromethane was produced and removed in the process. This anaerobic redox reaction is

quick and thus has great potential to be used as a remediation technique for co-contaminated soil and ground water.

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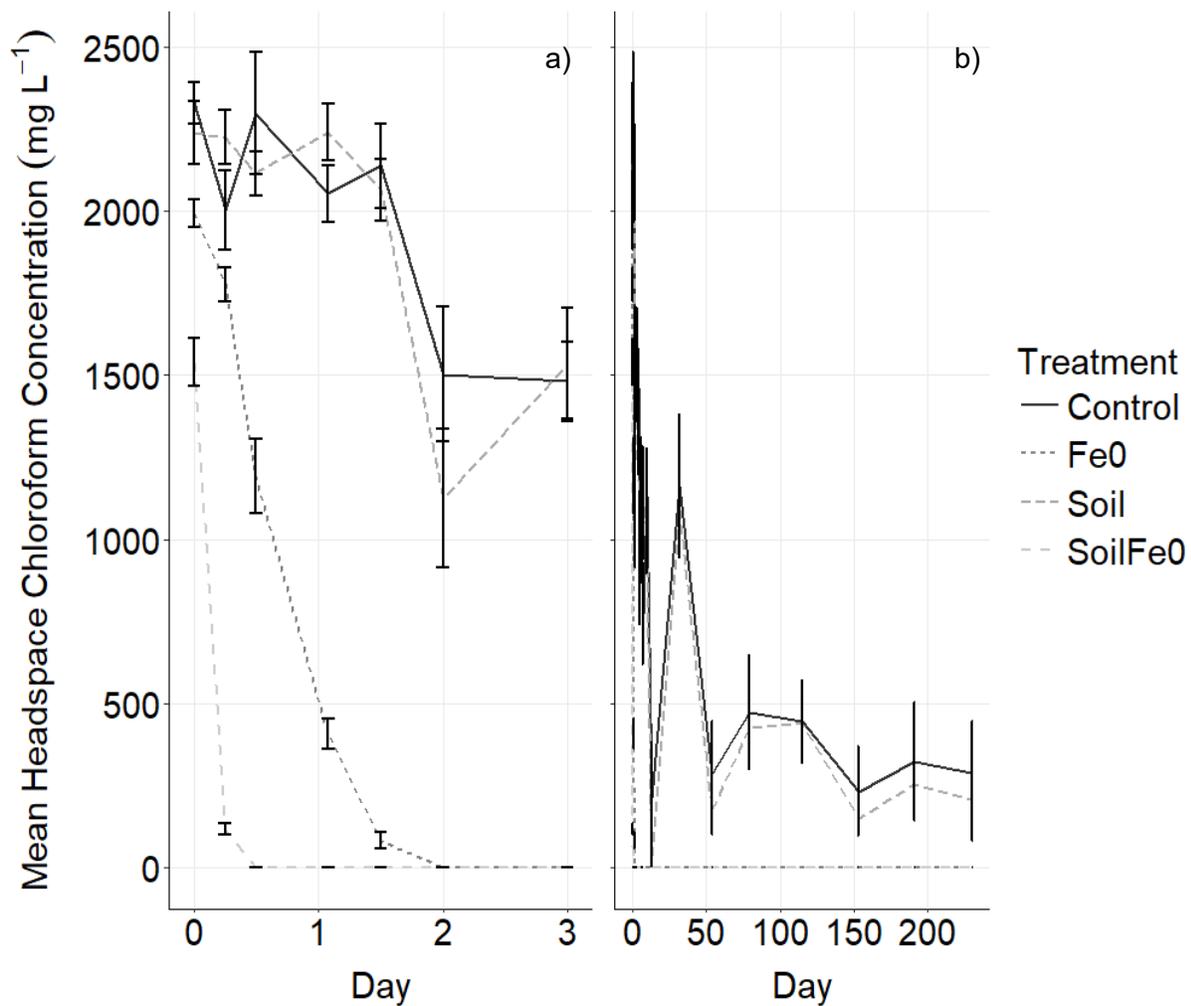


Figure 5.1. Change in headspace chloroform concentration over time for a) the first three days and b) the entire experiment. Error bars indicate standard error of the mean.

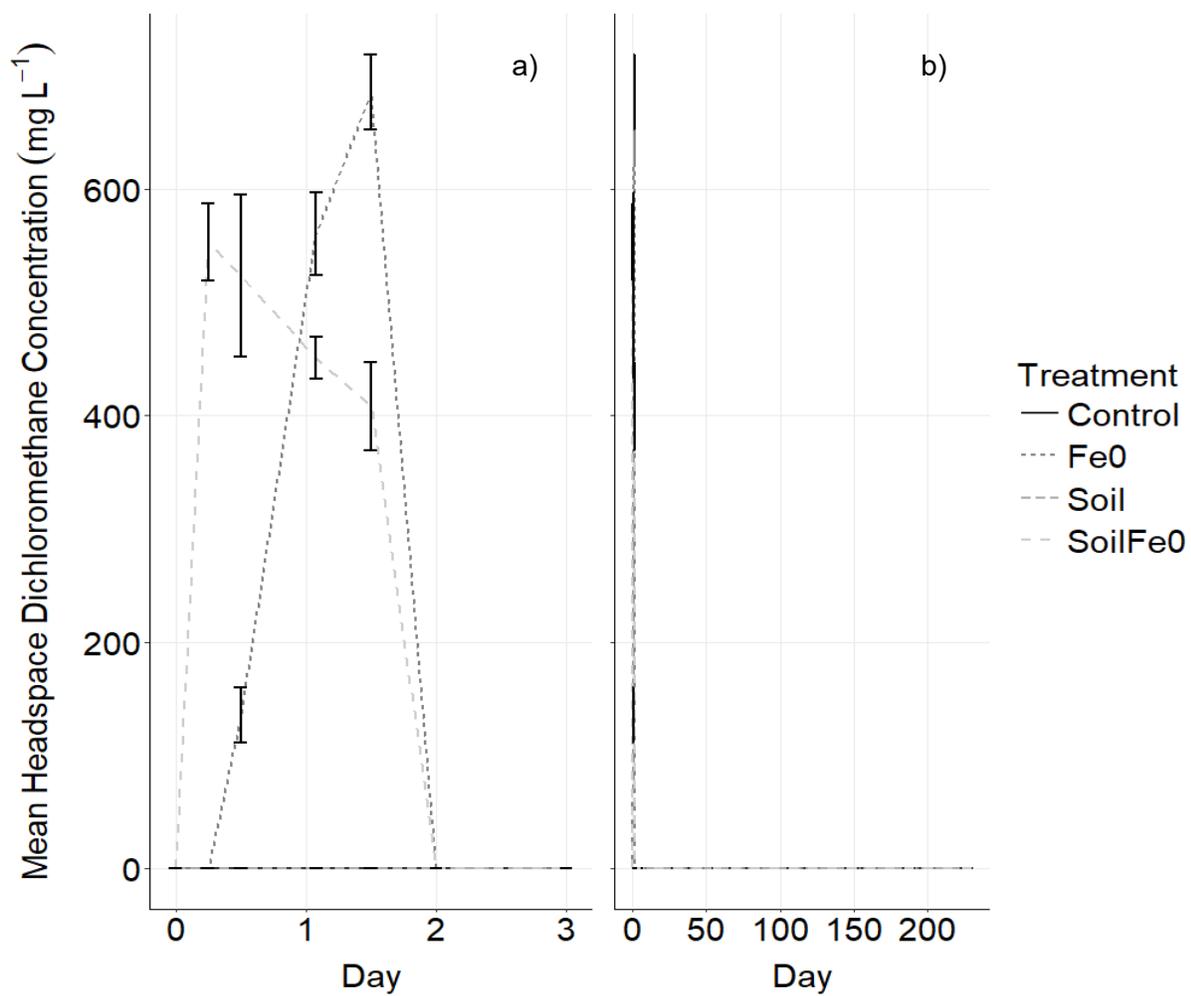


Figure 5.2. Change in headspace dichloromethane concentration over time for a) the first three days and b) the entire experiment. Error bars indicate standard error of the mean.

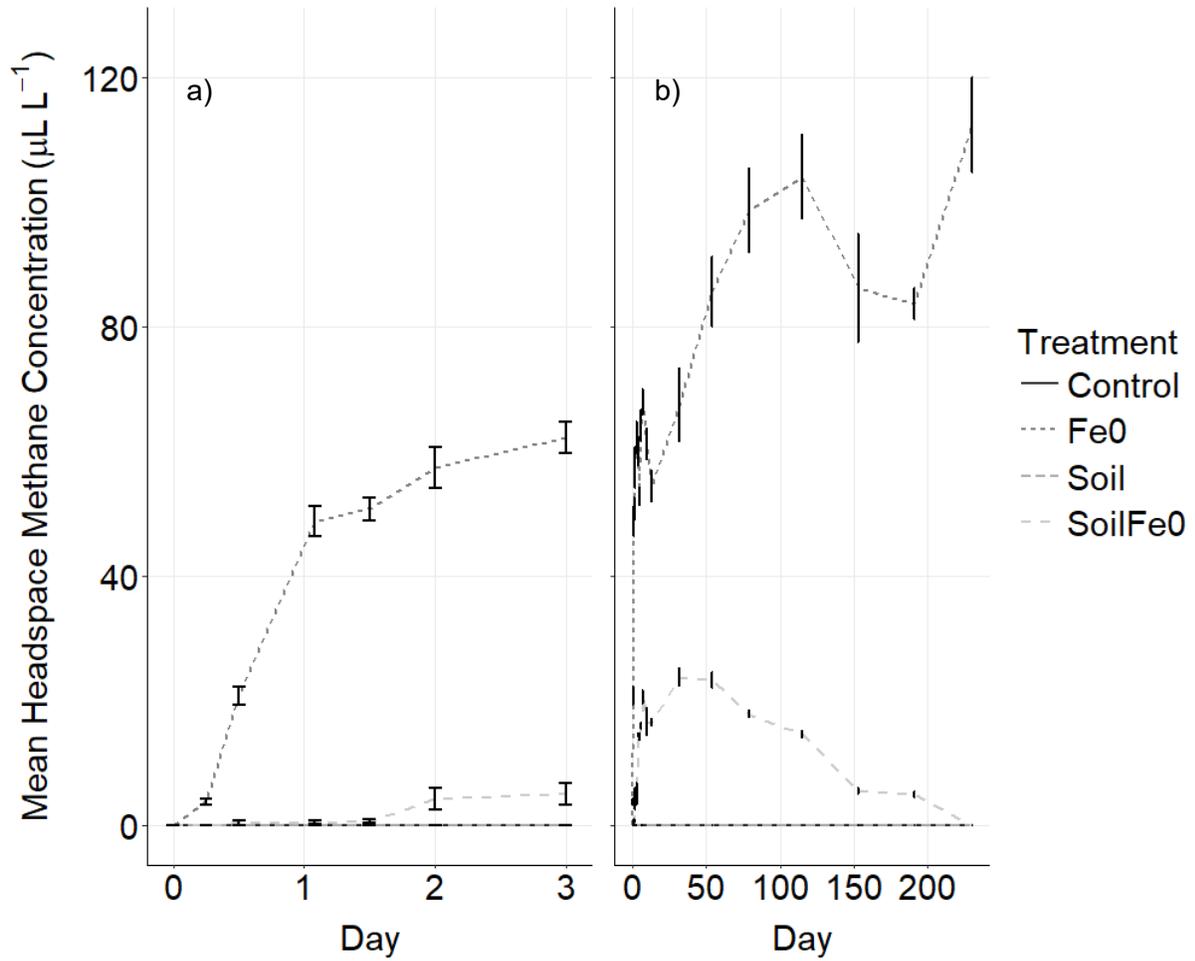


Figure 5.3. Change in headspace methane concentration over time for a) the first three days and b) the entire experiment. Error bars indicate standard error of the mean.

VI. EFFECTS OF CALCIUM NITRATE AMENDMENT ON LEACHING OF FINE TEXTURED SALT AFFECTED SOILS

1. INTRODUCTION

Salt affected soils contain significant quantities of neutral salts that dissolve in water to form cationic and anionic elements or compounds such as sodium (Na^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), chloride (Cl^-), and sulfate (SO_4^{2-}) (Allison et al 1954, Abrol et al 1988). Soils can accumulate salts due to anthropogenic activities such as irrigation, oil and gas production, and road maintenance (Alberta Environment 2001, Jordan et al 2004) or natural processes such as weathering of saline parent materials and evaporation of surface and ground waters containing salts (Allison et al 1954, Jordan et al 2004, Qadir and Oster 2004). Saline soils have electrical conductivity $> 4 \text{ dS m}^{-1}$ and sodic soils have exchangeable sodium percentage > 15 (sodium adsorption ratio > 13) (Jordan et al 2004).

Salinity increases the osmotic pressure with which water is held in the soil, making it more difficult for plants to access (Corwin 2003, Qadir and Oster 2004); it can also cause ion toxicities and nutrient imbalances (Qadir and Schubert 2002, Qadir and Oster 2004). Elevated sodium concentrations in sodic soils cause soil dispersion, degraded soil structure, increased runoff and erosion, and decreased seedling emergence and plant root penetration (Jordan et al 2004, Qadir and Schubert 2002, Qadir and Oster 2004). Salinity reduces ground water quality, limiting its use for irrigation and consumption and potentially negatively impacting aquatic organisms when transported to freshwater systems (Environment Canada 2001). Agricultural guidelines for various salinity parameters are readily available for Alberta and Canada (Table 6.1), but not international jurisdictions.

Leaching is typically used for remediation of salt affected soils, requiring net transport of water and salts down the soil profile. To remediate sodic soils, calcium is added before leaching to replace sodium on the soil particles (Qadir and Oster 2004). Common calcium amendments of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and calcium nitrate ($\text{Ca}(\text{NO}_3)_2$) may be dissolved and added to soil in liquid form to increase depth of addition and reduce treatment time (Alberta Environment 2001). If precipitation is insufficient to wash salts from the soil, irrigation or improvement of soil drainage may be required (Lee et al 2013). In many jurisdictions, if leachate release has potential to cause further damage to living organisms or previously uncontaminated media, it must be collected and properly discarded (Alberta Environment 2001). Soil can be excavated and washed in ex situ salt removal treatment (Sastre-Conde et al 2015).

Fine textured soils typically have small pore spaces and low permeability which can be a limiting factor in time required to leach salts (Callaghan et al 2014). Salt containing water may become trapped when held tightly in small pore spaces or in disconnected pores (Brooks et al 2010). Numerous factors, including clay content, mineralogy, and aggregation affect soil response to salinity and sodicity (McNeal et al 1968, Abu-Sharar et al 1987). Thus, research on individual soils is important in assessing site specific potential for leaching as a remediation strategy.

The objective of this research was to investigate the potential for calcium nitrate amendment in leaching treatment for fine textured salt affected soils. Two experiments were conducted. The objective of experiment 1 was to measure saturated hydraulic conductivity of various fine textured soils and determine the effect of calcium nitrate amendment. The objective of experiment 2 was to assess the impact of repeated leaching rounds on soil and leachate quality for calcium nitrate amended and unamended soils.

2. MATERIALS AND METHODS

2.1. Research Site

Soils were collected from the University of Alberta's former Eilerslie Waste Management Facility in Edmonton, Alberta (Appendix). The approximately 0.25 ha site is located within the Province of Alberta's Edmonton Transportation Utility Corridor with surroundings zoned for agriculture (Alberta Infrastructure 2003, AMEC Earth and Environmental 2010). A major road is located north and residential areas are north and east. Whitemud Creek is approximately 450 m north. Surficial geology at the Eilerslie site consists mainly of clay and clay till (Stantec Consulting Limited 2010). The site is located in the Central Parkland natural subregion (Natural Regions Committee 2006). The site processed laboratory waste from 1972 to 2007 (Stantec Consulting Limited 2013); activities included solvent incineration, acid dilution and neutralization, and temporary waste storage. Prior to 1983, a waste water pond leaked (University of Alberta 1984). Primary contamination concerns are electrical conductivity, chloride, sodium, chloroform, and dichloromethane. Another 14 soil and 32 ground water parameters were identified in exceedance of Alberta Tier 1 soil and ground water remediation guidelines for fine textured soils and agricultural land use (Alberta Environment and Parks 2016).

2.2. Soils

Soils for the experiments were collected from eight bore holes in the former waste water pond area by direct push drilling in 2014. Soils were air dried, mechanically ground, and passed

through a 2 mm sieve. Two composite soil samples were prepared for each bore hole based on sampling depth. Composite samples from the 3 to 7 m depth were expected to have finer texture than those from the 7 to 10 m depth based on previous soil analysis. Half the soils were amended with 8 g kg⁻¹ calcium nitrate based on the amount of calcium required to replace sodium on the most salt affected soil; half were unamended controls.

Before amendment, each composite soil was analyzed for percent sand, silt, and clay using the CSSS 55.3 hydrometer method (Kroetsch and Wang 2008); pH and electrical conductivity using the CSSS 15.2 and 15.3 saturated paste method (Miller and Curtin 2008); sodium adsorption ratio using the CSSS 15.4.4 calculation (Miller and Curtin 2008); calcium, magnesium, potassium, sodium, and sulfur (as sulfate) using the CSSS 15 saturated paste (Miller and Curtin 2008) and EPA 6010 B inductively coupled plasma atomic emission spectrometry (United States Environmental Protection Agency 1996) methods; and chloride using the CSSS 15 saturated paste (Miller and Curtin 2008) and APHA 4500 Cl⁻ E automated ferricyanide colorimetry (American Public Health Association 2011a) methods.

2.3. Saturated Hydraulic Conductivity

Saturated hydraulic conductivity was measured for soils from all 8 bore holes using the falling head permeameter method (Hillel 1998). Two layers of 1 mm² cheese cloth were secured to the bottom of a metal core of 7.7 cm diameter and 2.5 cm height. The core was filled with soil to 1.25 Mg m⁻³ bulk density and placed on a rubber ring and acrylic glass plate. Soil was saturated from the bottom up by placing the core in a pan of distilled water. Once saturated, a rubber ring and acrylic glass plate were placed on top of the core and secured to the bottom plate. The upper acrylic glass plate was attached to a burette filled with distilled water using a plastic tube. Time zero was set when the water level in the burette was at the 0 mL mark. Time was recorded for five consecutive 2 mL decreases in water level. Each treatment had three replicates.

2.4. Leaching

Leaching was conducted for soils from five bore holes with sufficient soil mass, using a laboratory scale soil column experiment. A metal collar was used to secure 1 mm plastic mesh to the bottom of an acrylic glass column of 10.2 cm diameter. Clean, dry sand was added to 2 cm in the column to hold soil without impeding drainage. Soil was added to 5 cm above the sand surface to 1.25 Mg m⁻³ bulk density. The column was attached to a metal stand using a ring clamp and covered with plastic wrap to minimize evaporation. A plastic bag was taped to the bottom to direct leachate into a sample bottle. Fisher Scientific 09-801C filter paper was

trimmed to fit in the column and placed on the soil surface to reduce disturbance from adding water. Soil was saturated by adding 275 mL distilled water to the surface through an inverted Buchner funnel. Once the soil was saturated and no ponded water remained, one pore volume, 216 mL, distilled water was added to the soil surface. The leaching process was repeated daily with subsequent 216 mL aliquots distilled water for eight days and leachate collected separately each time. Each treatment had two or three replicates depending on soil mass available.

Following leaching, one soil sample per column was analyzed for salinity parameters (pH, electrical conductivity, sodium adsorption ratio, calcium, magnesium, potassium, sodium, chloride, and sulfur). Leachate electrical conductivity was measured for each sample using an Oakton PC 300 pH conductivity probe. The first leachate sample for each column, containing any water that drained during the saturation process and the first pore volume of leachate, was analyzed for salinity parameters. Leachate pH, electrical conductivity, bicarbonate, carbonate, hydroxide, and total alkalinity (as calcium carbonate) were determined using the APHA 4500 H⁺ (American Public Health Association 2011b), APHA 2510 electrode (American Public Health Association 2011c), and APHA 2320 titration (American Public Health Association 2011d) methods. Dissolved calcium, magnesium, potassium, and sodium were determined using the APHA 3030 B filtration (American Public Health Association 2004) and EPA 6020 A inductively coupled plasma mass spectrometry (United States Environmental Protection Agency 2007) methods. Chloride, fluoride, nitrate (as nitrogen), nitrite (as nitrogen), and sulfate were determined using the EPA 300.1 ion chromatography method (Hautman and Munch 1997). Hardness and total dissolved solids were determined using APHA 1030 E calculations (American Public Health Association 2011e).

2.5. Data Analyses

Saturated hydraulic conductivity was calculated using the equation $K_{\text{sat}} = [2.3aL / A(t_2 - t_1)] \times (\log(H_1 / H_2) / 100$ where K_{sat} is saturated hydraulic conductivity (m s^{-1}), a is burette area (cm^2), L is soil length (cm), A is soil area (cm^2), t is time (s), and H is height of water above the bottom of the soil core (cm) (Hillel 1998). The mean of the five values for each replicate was calculated. Split plot analysis was completed with depth as main plot factor and amendment as split plot factor using SAS statistical software. A logarithmic transformation was applied and the model was adjusted for heterogeneous variances with depth and amendment.

Changes in soil salinity parameters before and after leaching were calculated as initial values subtracted from final values. Split plot analysis was conducted. Calcium was not included as

changes in concentration were confounded due to the presence of calcium in the amendment. Soil pH, magnesium, and potassium were not included because they were of little to no contamination concern when compared to remediation guidelines or soil quality test guides (Horneck et al 2011, AGVISE Laboratories 2012, Alberta Environment and Parks 2016). For change in electrical conductivity, the model was adjusted for heterogeneous variances with depth and treatment. For change in sodium adsorption ratio, sodium, chloride, and sulfate, homogeneous variances were used. Concentrations below detection limit were taken as the value of the detection limit. Since statistical analyses were conducted for five soil parameters, $\alpha = 0.05/5$ was used.

Leachate electrical conductivity was assessed by split plot repeated measures analysis with leaching round as the factor over which measurements were repeated. A logarithmic transformation and spherical covariance structure were applied. A significant three way interaction was assessed by a test of effects by leaching round slice.

Leachate salinity parameters for the first round of leaching were assessed by split plot analysis. Calcium, nitrate, and nitrite were not included in the analysis as changes in concentration were confounded due to the presence of calcium and nitrate in the amendment. Leachate pH, bicarbonate, alkalinity, magnesium, potassium, and fluoride were not included in the analysis because they were of little to no contamination concern when compared to ground water remediation or drinking water guidelines or well water test guides (World Health Organization 2009, Alberta Health 2014, Alberta Environment and Parks 2016). Hardness and total dissolved solids were not included because they were represented by calcium and magnesium and electrical conductivity, respectively. Carbonate and hydroxide were not included because their values were all below detection limit. For leachate electrical conductivity, the model was adjusted for heterogeneous variances with depth and treatment. For leachate chloride, a logarithmic transformation was used and the model was adjusted for heterogeneous variances with depth. For leachate sodium and sulfate, homogeneous variances were used. Since statistical analyses were conducted for four leachate parameters from the first leaching round, $\alpha = 0.05/4$ was used.

For each statistical analysis, normal distribution of residuals was assessed using the Shapiro Wilk test and visual assessment of conditional studentized residual plots. In some cases residuals were not normally distributed; however, the shape of the conditional studentized residuals histogram showed that the concept of the mean was meaningful. Post hoc Tukey comparisons of means were performed when significant effects were identified.

3. RESULTS

3.1. Initial Soil Properties

Soils sampled from the 3 to 7 m depth were finer textured than those from the 7 to 10 m depth (Table 6.2). Soils from the 3 to 7 m depth had silty clay loam and silty clay textures with 7.0 to 18.0 % sand, 42.8 to 55.2 % silt, and 35.5 to 40.7 % clay. Soils from the 7 to 10 m depth had loam and clay loam textures with 37.7 to 46.3 % sand, 29.3 to 34.1 % silt, and 24.0 to 28.6 % clay. Soils from the 3 to 7 m depth were generally more variable and more salt affected than soils from the 7 to 10 m depth (Table 6.3). Soils from the 3 to 7 m depth had electrical conductivities 2.9 to 12.2 dS m⁻¹ and sodium adsorption ratios 1.1 to 18.5. The soils with high electrical conductivities had high concentrations of chloride (1,330 to 3,500 mg kg⁻¹) and the soils with high sodium adsorption ratios had high concentrations of sodium (1,540 to 2,070 mg kg⁻¹). Soils from the 7 to 10 m depth had electrical conductivities 3.1 to 4.5 dS m⁻¹ and sodium adsorption ratios 1.7 to 3.1.

3.2. Saturated Hydraulic Conductivity

Soil saturated hydraulic conductivity was 1.30×10^{-8} to 9.07×10^{-7} m s⁻¹ (Figure 6.1). Depth ($p < 0.0001$), amendment ($p = 0.0001$), and their interaction ($p < 0.0001$) were significant. Mean saturated hydraulic conductivity was highest for soils from the 7 to 10 m depth; mean saturated hydraulic conductivity was $6.80 \pm 0.29 \times 10^{-7}$ and $7.37 \pm 0.28 \times 10^{-7}$ m s⁻¹ for calcium nitrate amended and unamended soils, respectively. For the 3 to 7 m depth, soils to which calcium nitrate was added had significantly higher mean saturated hydraulic conductivity ($1.29 \pm 0.14 \times 10^{-7}$ m s⁻¹) than unamended soils ($4.12 \pm 0.46 \times 10^{-8}$ m s⁻¹).

3.3. Leaching

Soil electrical conductivity was 2.9 to 12.2 dS m⁻¹ before leaching and 0.7 to 3.2 dS m⁻¹ after leaching. None of depth ($p = 0.0848$), treatment ($p = 0.1199$), or their interaction ($p = 0.8107$) were significant. Soil sodium adsorption ratio was 1.1 to 18.5 before leaching and 0.2 to 0.4 after leaching. None of depth ($p = 0.1686$), treatment ($p = 0.4065$), and their interaction ($p = 0.6287$) were significant. Soil sodium was 88 to 1,600 mg kg⁻¹ before leaching and 5 to 16 mg kg⁻¹ after leaching. Treatment ($p = 0.0017$) was significant while depth ($p = 0.1397$) and its interaction with treatment ($p = 0.2575$) were not. Mean decrease in soil sodium concentration was slightly greater for unamended soils (395 ± 97 mg kg⁻¹) than for calcium nitrate amended soils (394 ± 97 mg kg⁻¹). Soil chloride was 11 to 2,750 mg kg⁻¹ before leaching. Values, taken as

detection limits, were 8 to 18 mg kg⁻¹ after leaching. None of depth ($p = 0.0813$), treatment ($p = 0.2932$), and their interaction ($p = 0.4403$) were significant. Soil sulfate was 1,200 to 2,390 mg kg⁻¹ before leaching and 81 to 1,000 mg kg⁻¹ after leaching. None of depth ($p = 0.9472$), treatment ($p = 0.1341$), and their interaction ($p = 0.9312$) were significant.

Leachate electrical conductivity was 1.3 to 22.5 dS m⁻¹ (Figure 6.2). The three way interaction depth by treatment by leaching round ($p = 0.0001$) was significant. Other significant effects were the depth by leaching round interaction ($p < 0.0001$), the treatment by leaching round interaction ($p < 0.0001$), treatment ($p = 0.0241$), and leaching round ($p < 0.0001$). The depth by treatment interaction ($p = 0.5149$) and depth ($p = 0.2650$) were not significant. The test of effect slices indicated significant differences only for the first leaching round ($p < 0.0001$).

Leachate electrical conductivity for the first leaching round was 3.7 to 53.4 dS m⁻¹. Treatment ($p < 0.0001$) was significant while depth ($p = 0.0588$) and its interaction with treatment ($p = 0.8094$) were not. Mean leachate electrical conductivity was higher for calcium nitrate amended soils (14.1 ± 0.7 dS m⁻¹) than for unamended soils (6.9 ± 0.9 dS m⁻¹). Leachate sodium for the first leaching round was 192 to 3,790 mg L⁻¹. None of depth ($p = 0.1317$), treatment ($p = 0.0867$), and their interaction ($p = 0.1276$) were significant. Leachate chloride for the first leaching round was 17 to 7,980 mg L⁻¹ (Figure 6.3). Depth ($p = 0.0064$), treatment ($p < 0.0001$), and their interaction ($p = 0.0048$) were significant. Calcium nitrate amended soils from the 3 to 7 m depth had the highest mean leachate chloride concentration ($3,324 \pm 875$ mg L⁻¹) followed by unamended soils from the 3 to 7 m depth ($2,282 \pm 571$ mg L⁻¹), calcium nitrate amended soils from the 7 to 10 m depth (63 ± 12 mg L⁻¹), and unamended soils from the 7 to 10 m depth (51 ± 8 mg L⁻¹). Leachate sulfate for the first leaching round was 1,610 to 4,890 mg L⁻¹. Treatment ($p = 0.0055$) was significant while depth ($p = 0.5168$) and its interaction with treatment ($p = 0.9664$) were not. Mean leachate sulfate concentration was lower for calcium nitrate amended soils ($2,249 \pm 88$ mg L⁻¹) than for unamended soils ($3,051 \pm 147$ mg L⁻¹).

4. DISCUSSION

Saturated hydraulic conductivities of the fine textured soils used in this research were low, as expected, but could be increased in finer textured soils through calcium nitrate amendment. The saturated hydraulic conductivities of the coarser textured soils may not have been impacted by calcium nitrate amendment because they had lower initial sodium adsorption ratios. The slow drainage did not appear to restrict salt removal and leaching treatment was successful in reducing soil salinity regardless of calcium nitrate amendment. For 3 to 7 m soils before

leaching, electrical conductivity was rated good for one bore hole, fair for one, poor for one, and unsuitable for two. Sodium adsorption ratio was rated good for two bore holes, fair for one, poor for one, and unsuitable for one. After leaching with or without calcium amendment, electrical conductivity and sodium adsorption ratio were rated good for all five bore holes assessed. For 7 to 10 m soils before leaching, electrical conductivity was rated fair and sodium adsorption ratio rated good for all five bore holes. After leaching without calcium amendment, electrical conductivity and sodium adsorption ratio were rated good for all five bore holes assessed. After leaching with calcium amendment, electrical conductivity was rated fair for one replicate from each of three bore holes and good for all other samples; sodium adsorption ratio was rated good for all five bore holes.

Salt removal by leaching has been successful in several studies on fine textured soils. Tanton et al (1995) reported efficient salt removal from intact fine textured soil cores with water application rates of 2 and 108 mm day⁻¹ even though much of the water bypassed micropores and moved through macropores. They noted that salts must have moved through micropores by diffusion and through macropores by convection. Callaghan et al (2017) reported successful in situ salt removal from fine textured soils and attributed greater salt removal during drier conditions to increased movement of salts from micropores to macropores. Callaghan et al (2014) reported decreased hydraulic conductivity of intact smectite bearing soil cores during leaching which they attributed to clay swelling.

Treatment significantly affected change in soil sodium but the small difference in means, 1 mg kg⁻¹, is not considered meaningful because it is much smaller than the standard error of the mean. The difference is also much smaller than moderate soil sodium concentrations of 81 to 120 mg kg⁻¹ (AGVISE Laboratories 2012) so it would not be expected to be biologically meaningful. The lack of significant treatment and depth effects on soil salinity properties occurred because all treatments had similar amounts of salt removal after eight rounds of leaching. Comparing change in soil salinity after each round of leaching could be useful in detecting differences; however, the experimental design was restricted by soil availability and did not allow for additional sampling.

The greatest salt removal occurred during the first round of leaching. Calcium nitrate amendment appears to have increased salt leaching from soils. Calcium nitrate amendment would directly increase leachate electrical conductivity because calcium nitrate is a salt. Calcium nitrate amendment would also replace sodium on soil surfaces with calcium, reducing soil dispersion and improving soil structure. This could increase soil saturated hydraulic conductivity

and salt flushing. Leachate chloride results from the first leaching round indicate that the amendment improved salt flushing since the amendment did not contain chloride yet leachate chloride concentration was higher for amended soils. In contrast, leachate sulfate was lower for amended soils. However, this could be caused by its reaction with calcium to form gypsum (Jobson 2019).

Fine textured salt affected soils at the Ellerslie site could be treated in situ by installation of an interceptor trench down gradient of the contaminated zone. Contaminated water collected in the trench could be disposed of or treated in an evaporation pond (Naeth et al 2019). The soil saturated hydraulic conductivity values determined in this research would be important in calculating the time required for ground water to travel from the contaminated former waste water pond area to the interceptor trench. Calcium nitrate could be added to the soil to increase flow. Amendment rate could be calculated based on the moles of sodium present in the soil as was done in this research. Application in liquid form would be preferable so that the calcium could penetrate to the depth of contamination (Alberta Environment 2001). Calcium nitrate amendment could impact co-contaminants at the Ellerslie site by acting as a nutrient for bioremediation of organic contaminants. Nitrate leaching would not be a concern because leachate would be collected. Calcium nitrate amendment may interfere with zero valent iron treatment of chlorinated organic compounds because nitrate can be reduced by zero valent iron and form iron oxides on the metal's surfaces (Liu and Wang 2019). Leaching could also be conducted ex situ on excavated soils in combination with a leachate collection system.

5. CONCLUSIONS

Calcium nitrate amendment was successful in increasing saturated hydraulic conductivity of the finer textured soils in this research. Leaching was successful in removing salts from the soils regardless of treatment. It is uncertain whether calcium nitrate increased salt flushing from soils. Leaching is a possible remediation technique for the Ellerslie site. It could be applied in situ using an interceptor trench or ex situ on excavated soils.

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Table 6.1. Canadian soil and water salinity guidelines for agricultural land use.

Parameter	Medium	Canadian Guideline*	Alberta Tier 1 Guideline†
Electrical conductivity (dS m ⁻¹)	Topsoil	2	<2 good 2-4 fair 4-8 poor >8 unsuitable
	Subsoil		<3 good 3-5 fair 5-10 poor >10 unsuitable
Sodium adsorption ratio	Ground water		1
	Soil	5	<4 good 4-8 fair 8-12 poor >12 unsuitable
Sodium (mg L ⁻¹)	Ground water		200
Chloride (mg L ⁻¹)	Ground water		100
	Irrigation water	100 or higher depending on species	

* Canadian Council of Ministers of the Environment (2007)

† Alberta Environment and Parks (2016)

Table 6.2. Particle size distribution and texture of composite soil samples.

Depth (m)	Bore Hole	Sand (%)	Silt (%)	Clay (%)	Texture
3-7	14-05	13.0	48.3	38.6	Silty clay loam
	14-06	7.0	55.2	37.8	Silty clay loam
	14-29	11.4	49.6	39.0	Silty clay loam
	14-32	16.6	42.8	40.7	Silty clay
	14-33	9.0	55.0	36.0	Silty clay loam
	14-34	10.8	51.8	37.4	Silty clay loam
	14-35	11.4	51.8	36.8	Silty clay loam
	14-38	18.0	46.5	35.5	Silty clay loam
7-10	14-05	43.8	30.3	26.0	Loam
	14-06	44.0	30.9	25.1	Loam
	14-29	41.4	30.0	28.6	Clay loam
	14-32	40.7	32.9	26.5	Loam
	14-33	43.0	33.0	24.0	Loam
	14-34	37.7	34.1	28.2	Clay loam
	14-35	46.3	29.3	24.3	Loam
	14-38	43.0	30.2	26.8	Loam / clay loam

Table 6.3. Initial salinity properties of composite soil samples.

Depth (m)	Bore Hole	PH	Electrical Conductivity (dS m ⁻¹)	Calcium (mg kg ⁻¹)	Magnesium (mg kg ⁻¹)	Potassium (mg kg ⁻¹)	Sodium (mg kg ⁻¹)	Sodium Adsorption Ratio	Chloride (mg kg ⁻¹)	Sulfate (mg kg ⁻¹)
3-7	14-05	7.5	11.5	1020	221	17	1560	11.5	3170	1980
	14-06	7.7	11.9	690	157	35	2070	18.5	2470	3100
	14-29	7.7	4.2	586	118	12	445	4.4	287	2230
	14-32	7.6	2.9	558	111	15	108	1.1	38	1840
	14-33	7.6	3.6	525	112	17	211	2.2	146	1880
	14-34	7.6	10.2	836	160	17	1540	12.8	2480	2240
	14-35	7.5	5.7	904	176	16	238	1.9	1330	1470
	14-38	7.6	12.2	959	224	26	1590	12.0	3500	1920
7-10	14-05	7.8	3.7	499	208	32	297	2.8	46	2500
	14-06	7.8	4.3	480	214	38	317	3.0	64	2530
	14-29	7.8	3.8	506	201	31	275	2.6	34	2480
	14-32	7.6	3.7	524	194	22	264	2.5	21	2440
	14-33	7.7	4.5	488	210	39	323	3.1	23	2620
	14-34	7.7	3.6	470	186	32	262	2.6	83	2190
	14-35	7.8	3.1	438	173	26	162	1.7	36	1970
	14-38	7.7	3.9	452	184	32	257	2.6	26	2320

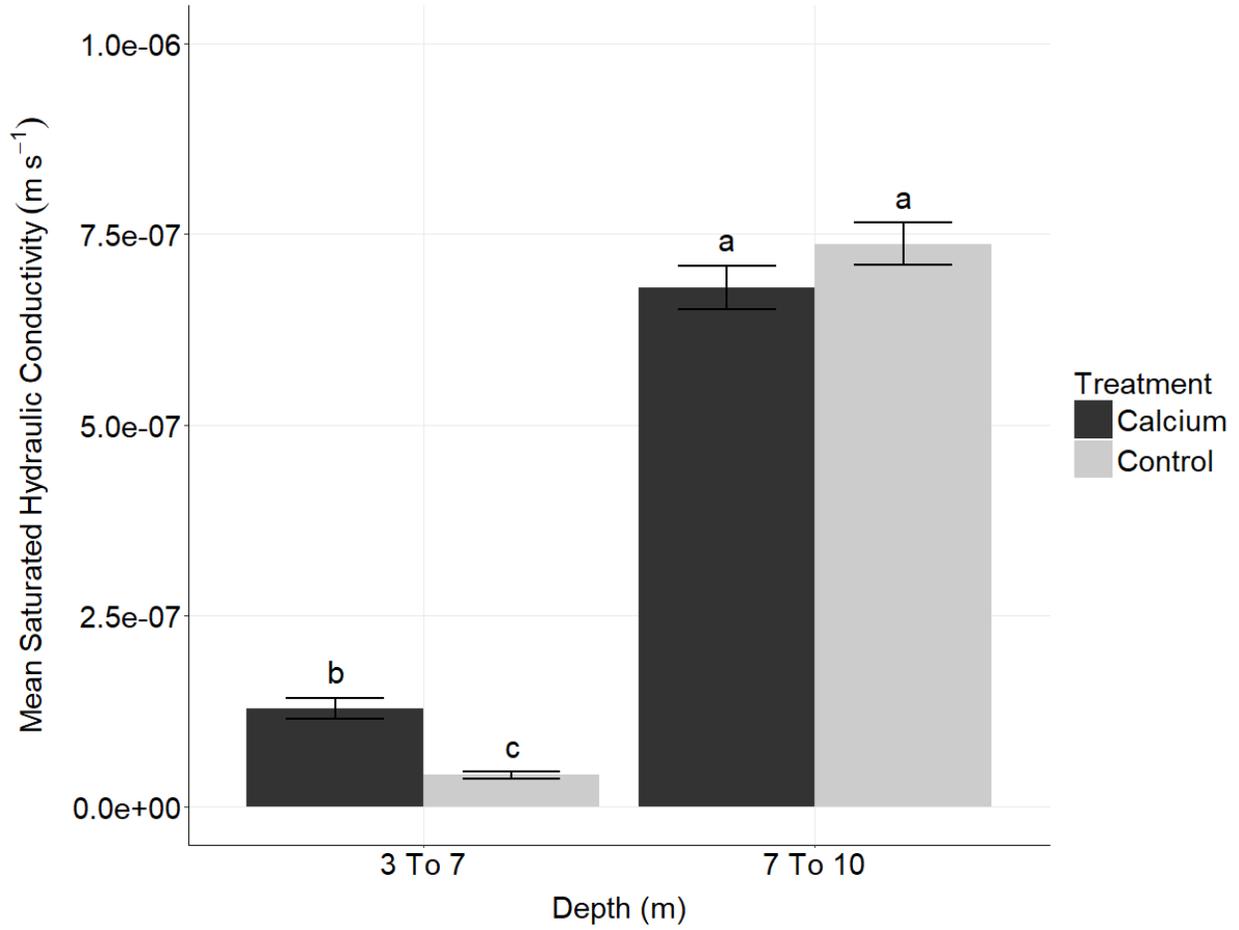


Figure 6.1. Soil saturated hydraulic conductivity. Error bars indicate standard error of the mean. Means with the same letter are not significantly different.

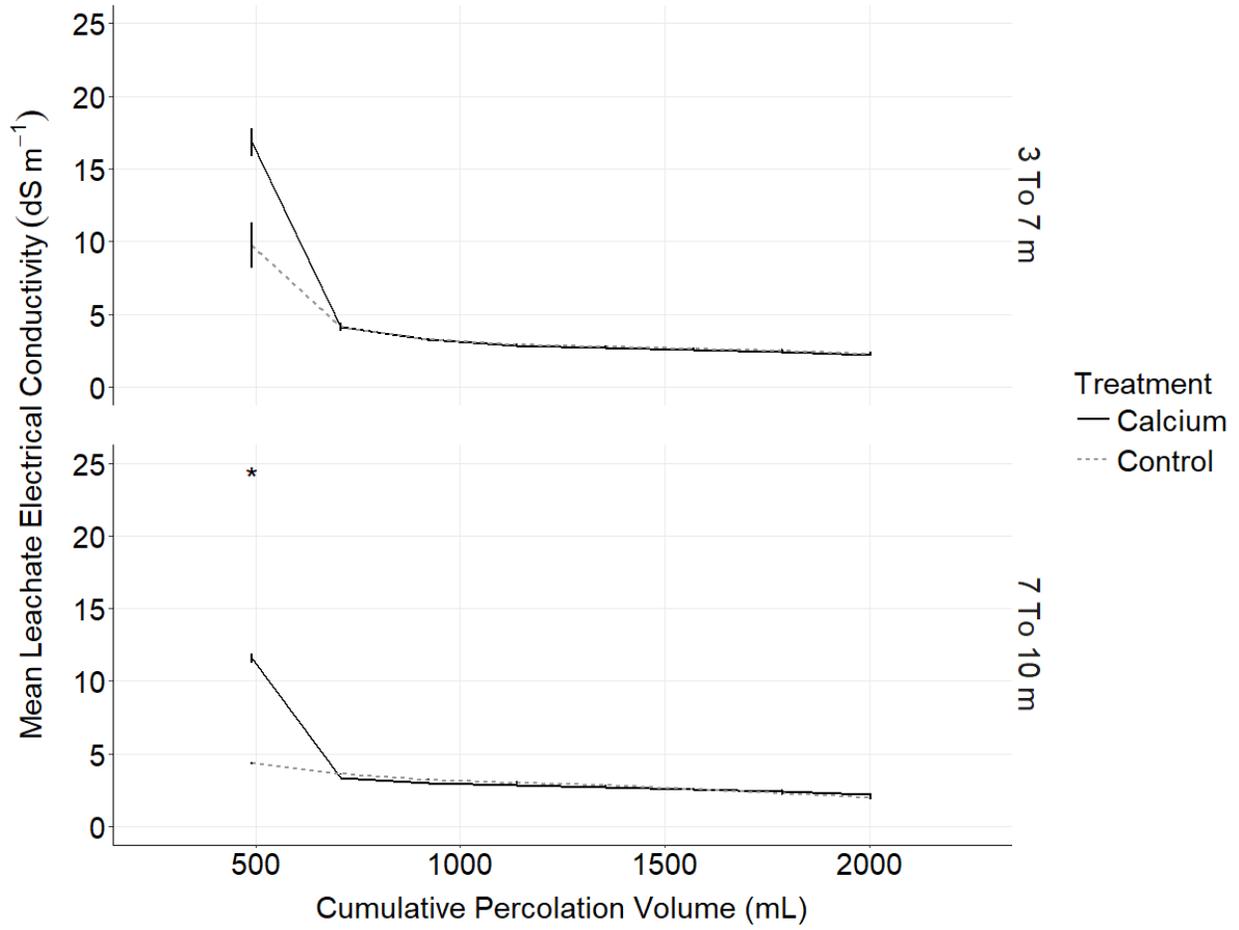


Figure 6.2. Leachate electrical conductivity over time. Error bars indicate standard error. Asterisk indicates leaching round at which significant differences were found.

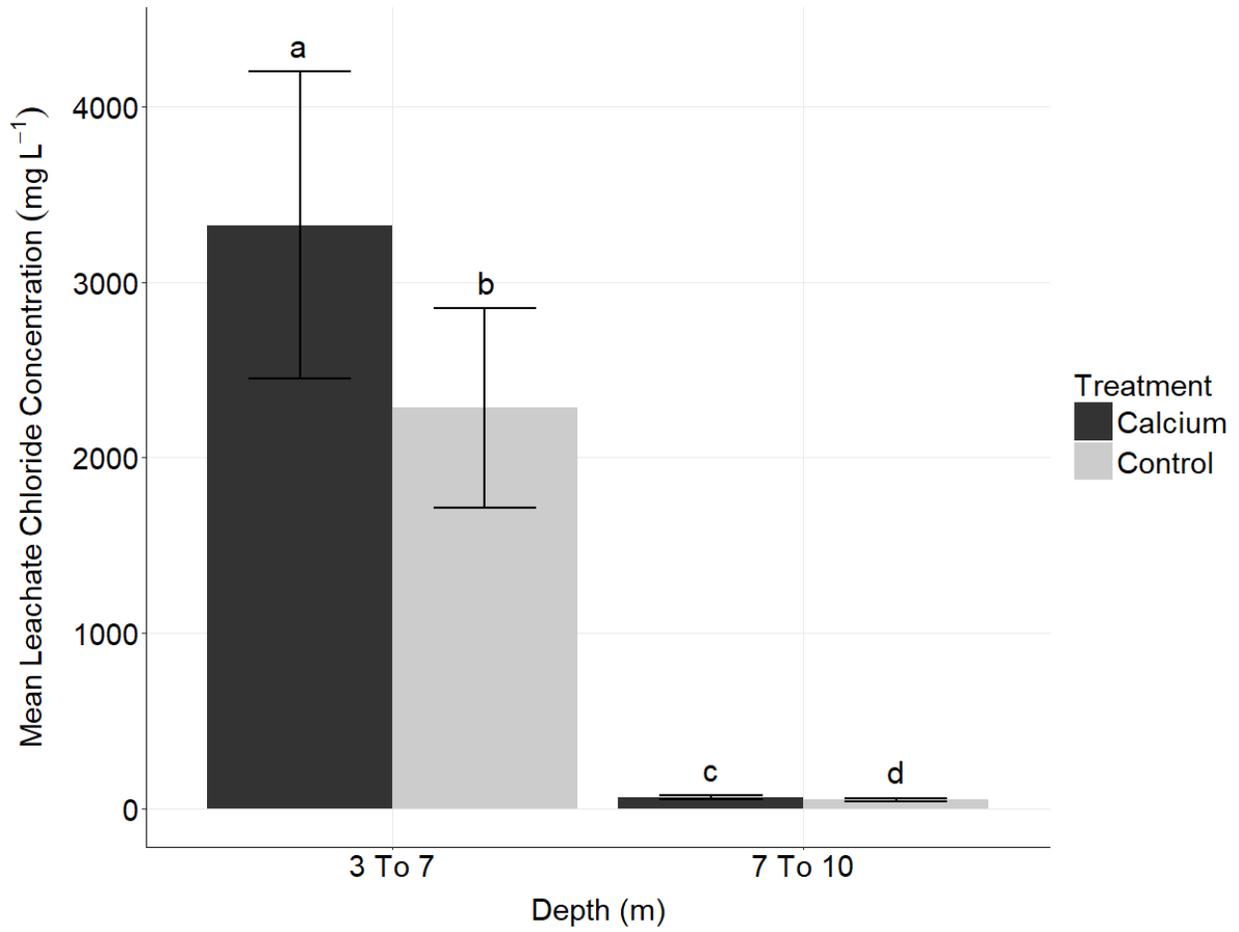


Figure 6.3. First round leachate chloride concentration. Error bars indicate standard error. Means with the same letter are not significantly different.

VII. SYNTHESIS

1. RESEARCH SUMMARY

Environmental contamination is a widespread and serious issue frequently associated with industrial activity. This research was conducted to address soil and ground water contamination at the former Ellerslie Waste Management Facility in Edmonton, Alberta (Appendix). The site operated from 1972 to 2007 to process laboratory waste including solvents, acids, and dyes (Stantec Consulting Limited 2013). Prior to 1983, the polyvinyl chloride membrane of a waste water pond leaked (University of Alberta 1984), resulting in soil and ground water contamination which has persisted to the present. Objectives were to assess spatial and temporal trends in soil and ground water contamination, characterize the microbial community, and investigate the potential of bioremediation, chemical reduction, and leaching for site remediation. The Ellerslie site provided an excellent research opportunity as it had fine textured soils and was impacted by a complex mixture of contaminants. Most remediation research focusses on coarser soils and single contaminants.

1.1. Contaminant Dynamics

Soil contamination was characterized using data from 2009, 2010 and 2014. A total of 18 soil parameters were in exceedance of Albert Tier 1 guidelines for a fine textured setting and agricultural land use (Alberta Environment and Parks 2016). Salinity and chlorinated organic compounds were of high concern based on frequency and magnitude of detection. Electrical conductivity (335 samples), dichloromethane (205 samples), and chloroform (116 samples) had the greatest number of exceedances. Dichloromethane (103,846 times guideline), chloroform (16,552 times guideline), and benzene (435 times guideline) had the greatest maximum magnitude of exceedance relative to guidelines. Three dimensional mapping of ten selected contaminants showed that contamination was spatially associated with the pond area. The salinity parameters chloride, sodium, and electrical conductivity had highest values in, north, and east of the pond area. The trace element parameters cadmium and nickel had exceedances only in the pond area; uranium had no exceedances. The organic compound parameters dichloromethane and benzene had highest values in, east, and west of the pond area; the highest chloroform values were spread across the total site. Carbon tetrachloride was only present on the west side of the site. The volume of soil with electrical conductivity $> 5 \text{ dS m}^{-1}$ was approximately $12,580 \text{ m}^3$.

Ground water contamination was characterized using data from even years from 1988 to 2016 plus 2015 and 2017. Relative to Albert Tier 1 guidelines for a fine textured setting and agricultural land use (Alberta Environment and Parks 2016), 37 parameters had exceedances. Salinity and chlorinated organic compounds were of high concern. Using data from 2014 to 2017, parameters with the greatest mean annual number of exceedances were electrical conductivity (27.8 wells), uranium (27.5 wells), and manganese (13.8 wells). Parameters with the greatest mean annual proportion of wells with exceedances (in brackets) were sulfate (1.000), electrical conductivity (0.991), and uranium (0.983). Parameters with the greatest maximum magnitude of exceedances relative to guidelines were dichloromethane (159,600 times guideline), chloroform (7,278 times guideline), and cadmium (4,750 times guideline). Temporal trends analyzed for 31 parameters varied by contaminant and well with no clear overall trend emerging. The directions of significant trends were typically not consistent within a contaminant or well. Two dimensional mapping of ten selected contaminants using data from 1992, 2004, 2014, and 2017 showed that contamination was spatially associated with the pond area. The salinity parameters, chloride, sodium, and electrical conductivity, had highest values in the pond footprint and extending approximately north east likely due to movement with ground water flow. The trace elements cadmium, nickel, and uranium had no clear spatial trends. The organic contaminants benzene, chloroform, and dichloromethane had highest concentrations in the pond area. Carbon tetrachloride was focused on the west side of the site like it was for soil.

1.2. Bioremediation

The potential for anaerobic chloroform biodegradation using indigenous or inoculation microorganisms as a remediation technique was investigated in three microcosm experiments. The amendments canola oil, acetate, lactate, nitrate, and sulfate were used. No meaningful changes in headspace chloroform were identified in any of the experiments. Expected degradation products of dichloromethane and methane were not detected. The lack of chloroform degradation could have been caused by a lack of microorganisms capable of degrading chloroform, the presence of other contaminants in the soil, or the relatively high chloroform concentration used.

1.3. Microbial Community

The soil microbial community at the Ellerslie site was investigated by comparing the communities from bore holes up gradient, down gradient, and in the pond area. The bacterial

community was likely impacted by contamination, the fungal community less so impacted. At each soil depth, samples from the pond area had lower mean bacterial richness and diversity. Bacterial communities from the up gradient and down gradient locations were more similar to each other than to communities from the pond area. Bacterial communities from the 5 or 6 m depth samples in the pond area had greater relative abundances of *Geobacter*, *Desulfosporosinus*, *Desulfuromonas*, and *Saccharibacteria* genera *incertae sedis*. These genera are not thought to degrade chlorinated organic compounds. However, *Geobacter* may be capable of benzene degradation. Fungal diversity may have been lower in the pond area at 5 and 6 m depths below surface. Fungal communities showed more similarity between sampling locations than did bacterial communities.

1.4 Chemical Reduction

The potential for anaerobic chemical reduction of chloroform was investigated using nano scale zero valent iron. Treatment with zero valent iron completely removed nearly 2 g L⁻¹ chloroform in reaction bottles including and excluding soil from the Ellerslie site. Headspace chloroform was no longer detected after 0.5 and 2 days in the soil zero valent iron and zero valent iron treatments, respectively. Chloroform was degraded to dichloromethane and eventually methane. Dichloromethane was no longer detected after 2 days in any treatment.

1.5. Leaching

Leaching of salt affected fine textured soils from the Ellerslie site was studied using column experiments in the laboratory. The finer textured soil from 3 to 7 m below surface had 35.5 to 40.7 % clay, electrical conductivity 2.9 to 12.2 dS m⁻¹, and sodium adsorption ratio 1.1 to 18.5. The coarser textured soil from 7 to 10 m below surface had 24.0 to 28.6 % clay, electrical conductivity 3.1 to 4.5 dS m⁻¹, and sodium adsorption ratio 1.7 to 3.1. Treatment with calcium nitrate increased mean saturated hydraulic conductivity for 3 to 7 m soils from $4.12 \pm 0.46 \times 10^{-8}$ m s⁻¹ to $1.29 \pm 0.14 \times 10^{-7}$ m s⁻¹. Calcium nitrate amendment did not significantly impact mean saturated hydraulic conductivity for the 7 to 10 m soils which had values of $6.80 \pm 0.29 \times 10^{-7}$ and $7.37 \pm 0.28 \times 10^{-7}$ m s⁻¹ for calcium nitrate amended and unamended soils, respectively. Soil electrical conductivity, sodium adsorption ratio, sodium, chloride, and sulfate were reduced substantially after eight rounds of leaching. Analysis of leachate electrical conductivity indicated that the first round of leaching removed the most salt. Calcium nitrate amendment resulted in increased leachate electrical conductivity during the first round of leaching, which would be at least partially caused by calcium nitrate which is a salt itself. Increased leachate chloride with

calcium nitrate amendment indicated that increased salt flushing likely occurred; decreased leachate sulfate may have been due to gypsum formation.

1.6. Conclusions

Overall, the Ellerslie site was confirmed to have a complex mixture of contaminants which were generally spatially associated with the pond area. Biodegradation of chloroform was likely not occurring on site and anaerobic chemical reduction with zero valent iron would be a much more effective treatment. There is potential for remediation of salt affected soils using a leaching based approach.

2. RESEARCH APPLICATIONS

Results from this research are important for development and application of a site management and remediation plan. The contaminant trend characterization study provided an inventory of soil and ground water contaminants that will need to be addressed. This information is crucial for remediation planning. The selected remediation strategy must address the complex contaminant mixture. Since there were no clear temporal trends, natural attenuation will not likely be feasible. Characterization of spatial trends illustrated the distribution of various contaminants, providing information to guide remediation efforts such as soil excavation, amendment application, or interceptor trench installation.

Results from the anaerobic chloroform biodegradation experiment do not indicate that bioremediation would be an effective treatment technique for the former Ellerslie Waste Management Facility. Additionally, microbial community characterization did not identify significant abundances of microorganisms known to degrade chlorinated organic compounds. Bioaugmentation could still be possible but habitat suitability for inoculation microorganisms would need to be considered.

Results from the anaerobic chloroform chemical reduction experiment showed that treatment with zero valent iron is a much more promising remediation strategy than bioremediation. The transformation of chloroform to methane was complete within days. Zero valent iron could be applied at the Ellerslie site using a batch reactor for excavated soil or a permeable reactive barrier or injection wells for in situ treatment.

Results from the column experiments indicate that the first round of leaching with water flushes the most salts from the soil. Calcium nitrate amendment increases saturated hydraulic conductivity of the finer textured soils and it may improve leaching of salts. Leaching could be

applied at the Ellerslie site by way of soil washing for excavated soil or an interceptor trench for in situ treatment.

The treatments studied in this research were selected based on their ability to be applied in situ with relatively low maintenance. If a relatively long period of time such as a decade was available for remediation, the Ellerslie site could be treated using one or more of the above methods. An interceptor trench could be installed to capture salt affected ground water. Installation would cost approximately \$ 100,000 to 150,000 and annual water treatment or disposal costs could be as high as \$ 15,000 per year (Naeth et al 2019). Ground water containing chlorinated organic compounds would also collect in this trench and could necessitate additional infrastructure to prevent emissions of volatile contaminants and additional water treatment or disposal costs. An alternative option would be to treat chlorinated organic compounds with zero valent iron as a permeable reactive barrier before they reach the interceptor trench. The estimated cost of this approach is \$ 175,000.

Since this research began, a pipeline was installed through the site. Future plans for development include a second pipeline and a light rail transit line. Areas near the site will have a light rail transit station and a parking lot. Some construction has begun near the site. If development necessitates rapid site remediation, different applications of the principles included in this research or methods not included in this research may be more desirable. Chlorinated organic compounds could be treated using zero valent iron if applied by pressure injection, in a biopile, or in a batch reactor with estimated costs of \$ 225,000, \$ 375,000, and \$ 500,000, respectively (Naeth et al 2019). Thermal desorption could treat chlorinated organic compounds in excavated soils at an estimated cost of \$ 1.2 million. Ex situ soil washing could be used to treat salt affected soils at an estimated cost of \$ 20 to 36 million. A typical dig and dump treatment, including soil excavation, sorting, land filling, and replacement, would cost in excess of \$ 25 million.

Overall, the information gained by this research is critical for development and execution of a management and remediation plan for the Ellerslie site. Results can also be extrapolated to similar sites with multiple contaminants or fine textured soils.

3. RESEARCH LIMITATIONS

The primary limitation of the contaminant trend characterization study was the number of concentrations below an analytical detection limit which was above the applicable Alberta Tier 1

guideline; these values were omitted from spatial trend analysis. For example, soil chloroform and carbon tetrachloride had 374 and 413 values removed, respectively, which hindered determination of spatial distribution. Spatial trend analysis was limited by the scarcity of soil data available for depths greater than 10 m below ground surface, the relatively small amount of ground water data available for some parameters in some years, and the changing location and number of ground water samples over time. Temporal trend analysis was limited by the monotonic nature of the Mann Kendall test, with trends which changed direction over time not able to be identified.

The first microcosm experiment testing the potential for anaerobic chloroform biodegradation was limited by the non-aseptic soil sampling. This was corrected in the second and third experiments, yet chloroform degradation was still not detected. The microbial community study was limited by the number of soil samples for which deoxyribonucleic acid sequencing was successful. This was likely partially related to low bacterial counts in samples from lower depths.

The anaerobic chloroform chemical reduction experiment was limited by the number of sampling times at the beginning of the study. The most frequent sampling possible using the repeated measures experimental design was at six hour intervals due to the time necessary for sample analysis. More frequent sampling would allow for calculation of reaction kinetics and more accurate determination of treatment time.

The primary limitation of the leaching experiments was the lack of soil measurements after each round of leaching. The small amount of soil available meant that it could only be analyzed at the end of all eight rounds of leaching.

4. FUTURE RESEARCH

There are numerous opportunities for future research on the contamination and remediation of soil and ground water at the Ellerslie site. Further soil sampling and analysis with lower detection limits for certain parameters would allow for more accurate contaminant mapping. However, the drilling and laboratory costs would be high. Instead, a study could be conducted to determine relationships between soil and ground water concentrations of selected contaminants. Then, ground water maps could be used to estimate soil maps for each parameter. Continued soil and ground water monitoring could be useful in determining the effect of pipeline infrastructure on contaminant movement.

Further research of potential remediation methods would be beneficial in developing the most efficient remediation plan for the site. Continued research on bioremediation could find an effective microbial based treatment. More specifically, experiments could be conducted using different inoculation sources and amendments. If a community capable of the desired contaminant degradation is identified, it could be grown and optimized for on site application.

Chemical reduction research could be expanded to include various types of zero valent iron with different particle sizes, surface areas, surface modifications, or support materials. The combination of zero valent iron amendment for treatment of chlorinated organic compounds and calcium nitrate amendment for treatment of salinity should be investigated to determine changes in their effectiveness. The combination of zero valent iron amendment and microbial degradation could be studied.

An assessment could be completed to determine the risk posed by soil and ground water contamination when all contaminant and site characteristics are taken into consideration. It could consider public opinion of various remediation options. The results could be used to determine the required speed of remediation or in further development and modification of site specific remediation guidelines.

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APPENDIX: DETAILED RESEARCH SITE DESCRIPTION

1. LOCATION

The University of Alberta's former Ellerslie Waste Management Facility is located at 210 127 Street South West in Edmonton Alberta, within SE 25-51-25 W4M (Figure A.1). The University of Alberta leased that quarter section, SE 24-51-25 W4M, and NE 24-51-25 W4M from the Province of Alberta in 1961 for 50 years. The three quarter sections form the University of Alberta's Ellerslie Research Station. Since the 2011 lease expiration, the land has been leased on an annual basis (Dvorsky 2015). The University of Alberta must return the property to the Province in a condition similar to that present before the land was leased.

The former Ellerslie Waste Management Facility was approximately 0.25 ha (AMEC Earth and Environmental 2010). The area surrounding the site is zoned for agricultural purposes. The entire SE 25 quarter section is part of the Province of Alberta's Edmonton Transportation Utility Corridor (Alberta Infrastructure 2003, Dvorsky 2013). Anthony Henday Drive is located immediately north of the former Ellerslie Waste Management Facility. Residential areas are located north across Anthony Henday Drive and east across 127 Street South West. Whitemud Creek is approximately 450 m north of the site; it is also west of the site. Blackmud Creek is east and north of the site.

2. OPERATION

2.1. Infrastructure And Activities

The University of Alberta's Ellerslie Research Station is used for field research and teaching related to agriculture, environmental science, and forestry. The Ellerslie Waste Management Facility operated from 1972 to 2007 under Alberta Environment licence 91-WL-124 and approval 20370-01-00 and Canadian Nuclear Safety Commission licence CNSC 130677 to process liquid waste generated by University of Alberta research and education laboratories and outside sources (Alberta Environment 1991, AMEC Earth and Environmental 2010, Stantec Consulting Limited 2013).

Site layout changed over the years as infrastructure was added or removed. At the end of operation, there were eight buildings (AMEC Earth and Environmental 2006) (Figure A.2). The acid neutralization building was built in the early 1990s for acid treatment. It was 25 m², with a metal exterior and concrete slab floor. The vial crusher and drum storage building was built in

the mid 1980s for processing vials of fluorescent dyes. It was 25 m², with a metal exterior, concrete slab floor, and curbing to retain spills. The storage tank building was built in the mid 1980s and housed two above ground storage tanks of 65,000 L and 20,000 L capacity, respectively. It was 90 m², with a metal exterior and concrete floor approximately 0.9 m below grade. The radioactive waste storage building was built in the early to mid 1970s and held solid and liquid radioactive waste. It was 50 m², with a metal exterior and concrete slab floor. The chemical waste storage building was built in the early 1980s and used for waste sorting, storage, and packaging. It was 40 m², with a metal exterior and concrete slab floor. The storage building was built in the late 1970s and held drums. It was 25 m², with a metal exterior and concrete slab floor. An office building was built in the mid 1980s and a maintenance equipment shed was built in the early 1990s. The office was 20 m² with a metal exterior and wood floor; the shed was 50 m² with a metal exterior. All buildings except the storage building and the maintenance equipment shed were connected to municipal water supply and heated by natural gas furnaces and heaters. Sanitary sewage from the office building was stored in Ellerslie Research Station lagoons north west of the research site until 2003 when the lagoons were decommissioned and the office building was connected to the municipal sanitary sewer system during construction of Anthony Henday Drive (AMEC Earth and Environmental 2008a). Two 10,000 L above ground solvent storage tanks within a concrete dyke for secondary containment were located near the vial crusher and drum storage building (AMEC Earth and Environmental 2006). Approximately 50 % of the site surface was covered by buildings or pavement (AMEC Earth and Environmental 2010). The site was surrounded by a chain link fence.

Some waste was treated on site while some was packaged for off site disposal. Acids were diluted and neutralized using sodium hydroxide or sodium carbonate in a 1,360 L polyethylene reaction tank (AMEC Earth and Environmental 2010, Stantec Consulting Limited 2013). Plastic and glass vials were shredded, rinsed, and temporarily stored before being sent for off site disposal. Reactive waste was treated using an open top explosives incinerator (Alberta Environment 1991). Chlorinated (chloroform, dichloroethane, trichloroethane, dichloromethane, tetrachloroethene) and non chlorinated (methyl ethyl ketone, acetone, methanol, benzene, xylenes) solvents are presumed to have been treated using an open top explosives incinerator from 1972 to 1986 (Stantec Consulting Limited 2013). Solvents were treated using a liquid waste incinerator from 1986 to 1991. In the following years, solvents were sent for off site disposal. The Ellerslie Waste Management Facility did not accept explosives or bio medical waste (Alberta Environment 1991). Incinerator ash, decayed solid radioactive waste, asbestos, mercury, exotic waste, metal salts, isocyanates, oxidizers, sulfur, and solid organics were

packaged for disposal at the Clover Bar Landfill in Edmonton, the Alberta Special Waste Treatment Centre in Swan Hills, and Laidlaw Hazardous Waste Landfill in Ontario.

A waste water pond was used from 1972 to 1986 to collect and store liquids (AMEC Earth and Environmental 2010, Stantec Consulting Limited 2013). It was built on compacted clay and lined with a 16 mm polyvinyl chloride membrane. The waste water pond was replaced by the waste water and solvent storage tanks (AMEC Earth and Environmental 2008b, Stantec Consulting Limited 2013). Material transferred to the waste water storage tank included waste water from acid neutralization, rinse water from vial processing, and storm water runoff (AMEC Earth and Environmental 2010, Stantec Consulting Limited 2013). The solvent storage tanks were used for fluorescent dyes, such as fluorescein and rhodamine, and solvents. From 2006 to 2007, when the facility was being closed, solvents were stored in 205 L barrels (AMEC Earth and Environmental 2010).

The former Ellerslie Waste Management Facility was decommissioned in 2007 under Alberta Environment approvals 20370-01-00 and 20370-02-00 and Canadian Nuclear Safety Commission licence WNSL-W5-3702.0/2011 (Stantec Consulting Limited 2010). All buildings were removed at that time other than the office building which remained until October 2012. The site is currently being used for land reclamation research by the University of Alberta in partnership with the Government of Alberta under Alberta Environment approval 20370-02-00. Laboratory waste management is now handled at the University of Alberta's Cloverbar Hazardous Waste Management Facility located at the City of Edmonton's Clover Bar Waste Management Centre (Stantec Consulting Limited 2013).

2.2. Accidental Releases

Two accidental releases have been reported. A University of Alberta report (1984) indicated that the waste water pond liner had cracked prior to 1983 and that this crack led to leakage from the pond. The pond was decommissioned in 1986 with the liner and approximately 100 m³ of rock and sludge transferred to an off site landfill (University of Alberta 1986). The pond was backfilled with soil obtained from the construction of the waste water storage tanks and off site locations. Soil and ground water contamination with organic and inorganic substances has persisted following decommissioning. Stantec Consulting Limited (2010) estimated 12,000 to 34,000 m³ soil would need to be treated to meet Alberta Tier 1 Soil and Groundwater Remediation Guidelines for fine grained material in an agricultural setting. The area of chloride impacted

ground water was estimated at 40 m by 60 m. Semi annual ground water monitoring has been conducted since approximately 1988 (AMEC Earth and Environmental 2006).

In 1999, approximately 4,800 L of liquid was released when solvent storage tank valves were left open (AMEC Earth and Environmental 2006). Approximately 1,700 L overflowed from the secondary containment system and entered the parking lot where approximately 1,500 L entered a catch basin. The liquid mostly contained non chlorinated solvents and water. Dichloromethane was also present. Captured liquid and impacted snow and ice were transferred to the solvent storage tanks. Approximately 8.8 m³ impacted soil was remediated on site by aeration on a concrete pad and returned to the parking lot.

3. ENVIRONMENTAL SETTING

3.1. Climate

Edmonton has a continental climate characterized by warm summers and cold winters with no dry season (Peel et al 2007). Mean daily temperature ranges from -10.4 °C in January to 17.7 °C in July (Environment Canada ND). Maximum daily temperature is highest in July at 23.1 °C and minimum daily temperature is lowest in January at -14.8 °C. Mean annual precipitation, dominated by summer rainfall, is 455.7 mm. Maximum rainfall is 93.8 mm in July and maximum snowfall is 24.5 cm in January. Wind is most frequently from the west, with mean speed ranging from 10.3 km h⁻¹ in January to 13.8 km h⁻¹ in May. Mean date of last spring frost is May 9 and mean date of the first fall frost is September 22. Mean frost free period is 135 days.

3.2. Hydrogeology

Regional bedrock consists of sandstones, shales, bentonitic beds, and small coal lenses of the Horseshoe Canyon or Wapiti Formations (Ceroici 1979, Cooper 2000). Bedrock surface is approximately 663 to 670 m above sea level. Surficial geology consists of silt and clay deposits from glacial Lake Edmonton (Kathol and McPherson 1975). Significant sand and gravel thicknesses are expected along the buried Beverly Channel Valley, although the channel's thalweg is approximately 6 to 7 km north west of the site. Regional topography is undulating to hummocky (Natural Regions Committee 2006).

Test drilling at the site for installation of ground water monitoring wells and collection of soil samples has exposed mainly fine textured material with a brown grey clay overlying a dark brown clay till (AMEC Earth and Environmental 2010, Stantec Consulting Limited 2010). Surficial clay was a massive unit with very few varved horizons. Fill which appeared to overlie a

soil horizon was observed in some bore holes within the waste water pond. The buried soil horizon was assumed to be an artifact of back filling when the pond was covered. A sand unit was identified at an approximate depth of 10.5 to 12.5 m below ground (AMEC Earth and Environmental 2010). Its size and continuity are unknown. Bedrock surface has not been identified in any testing to date. The deepest test holes were completed to an approximate depth of 15 m. Topography on site is low relief with elevation from approximately 683 to 688 m above sea level (Stantec Consulting Limited 2010).

Ceroici (1979) predicted potential ground water yields of 6.5 to 33.0 m³ d⁻¹ from bedrock aquifers of the region. Higher yields may be available from sand and gravel aquifers associated with buried channel thalwegs. The direction of regional ground water movement is north and north west towards the regional topographic low represented by the North Saskatchewan River. Shallow ground water movement beneath the facility is approximately northwards with the highest ground water levels typically located in the south west corner of the facility. Hydraulic conductivity on site, estimated from monitoring well response test data, ranges from 2.1 x 10⁻⁹ to 1.2 x 10⁻⁷ m s⁻¹. Estimated linear ground water velocity ranges from < 0.1 to 0.7 m y⁻¹.

3.3. Soil

Edmonton is located within Alberta's Central Parkland natural subregion (Natural Regions Committee 2006). Predominant Central Parkland soils include Orthic Black Chernozems, Orthic Dark Gray Chernozems, and Dark Gray Luvisols. Solodized Solonetz and Solod Solonetzic soils are common. Orthic and Humic Gleysols are found mainly in wetland areas and low lying areas.

Soil in the SE 25-51-25 W4M area has been classified as Malmo silty clay loam, an Eluviated Black Chernozem formed on slightly saline lacustrine parent material (Bowser et al 1962). The soil is fairly well to well drained. It has a very dark brown to black silty clay loam Ah horizon with granular to weak prismatic structure over a thin dark grayish brown to grayish brown silty clay loam Ae or Ahe horizon with medium platy to coarse weak blocky structure. The dark brown to brown clay Bt horizon has weak columnar to subangular blocky structure. The dark grayish brown clay Ck and C horizons have massive structure.

3.4. Vegetation

Most of the non urban Central Parkland natural subregion is used for cultivation or livestock grazing, although small areas of native parkland remain in areas not suitable for agriculture due to their soil or topography (Natural Regions Committee 2006). Native vegetation is dominated

by plains rough fescue prairie in the warmer, drier south east and aspen forest and parkland in the cooler, wetter north west. Approximately 10 % of the Central Parkland is wetland with marshes, willow shrub lands, and seasonal ponds in the south and treed fens in the north west.

Various native plant species are commonly found on Black Chernozem soils; grasses and forb dominate with some small inclusions of woody shrub species in imperfectly and moderately well drained areas and wetland species in small wetlands such as sloughs (Natural Regions Committee 2006). Common grasses include *Festuca hallii* (Vasey) Piper (plains rough fescue), *Hesperostipa curtiseta* (Hitchcock) Barkworth (western porcupine grass), *Elymus trachycaulus* (Link) Gould ex Shinnars (slender wheat grass), and *Helictotrichon hookeri* (Scribner) Holub (Hooker's oat grass). Common perennial forbs include *Anemone patens* L. (prairie crocus), *Artemisia frigida* Willdenow (prairie sagewort), *Linum lewisii* Pursh (wild blue flax), *Galium boreale* L. (northern bedstraw), and *Geum triflorum* Pursh (three flowered avens). Common shrubs include *Symphoricarpos occidentalis* Hooker (western snowberry), *Elaeagnus commutata* Bernhardt ex Rydberg (silver berry), *Rosa acicularis* Lindley (prickly rose), and *Amelanchier alnifolia* (Nuttall) Nuttall ex M Roemer (saskatoon).

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Figure A.1. Location of former Ellerslie Waste Management Facility in Edmonton, Alberta.

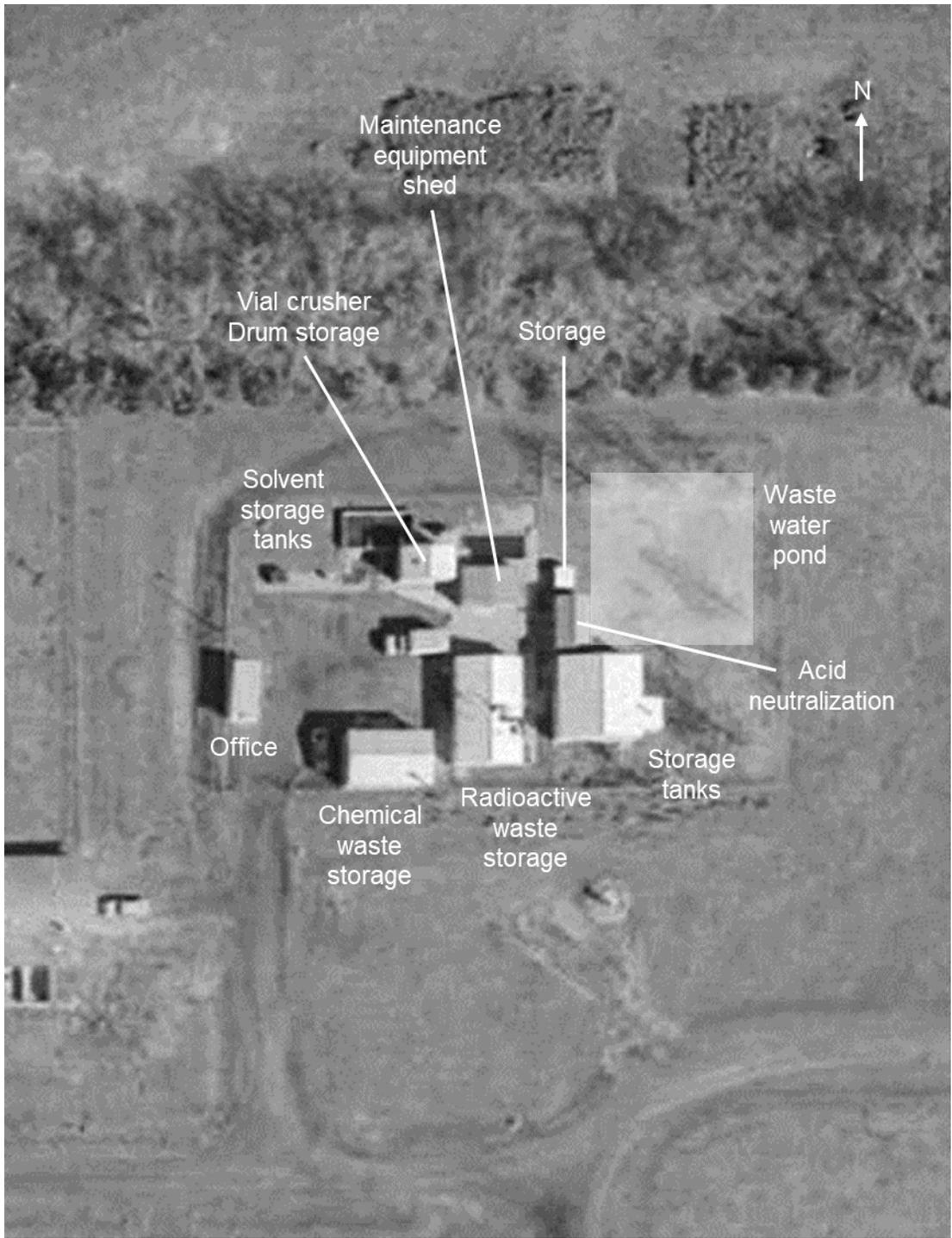


Figure A.2. Former Ellerslie Waste Management Facility layout. Scale not available.