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# Detection of trace elements in Svalbard reindeer (*Rangifer tarandus* *platyrhynchus*) faeces in Longyearbyen, Adventdalen and Kapp Linné

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## Abstract

The Arctic is considered a deposition and accumulation region for several possible toxic pollutions, including toxic non-essential elements, that can enter the Arctic environment via long-range transport from other anthropogenic sources in the world. The main aim of this study was to compare levels of essential and non-essential elements in Svalbard reindeer faeces from two geologically different locations at Svalbard with assumed different contamination loads. Longyearbyen and Adventdalen are both inhabitable areas with high levels of human activity including ongoing mining activity, airport activity and exhaust from cars, snow mobiles and boats. The distance from the road and the coal power plant in Longyearbyen and Adventdalen was also measured to examine if animals feeding closer to these possible sources had higher levels of toxic elements. Kapp Linné, which is situated 80km from Longyearbyen, has comparatively much lower impact from human activity, but is more exposed to oceanic input and precipitation. The distance from the ocean close to Kapp Linné was therefore measured to examine possible sources of oceanic input on the faeces.

Svalbard reindeer faeces were collected in Longyearbyen, Adventdalen and Kapp Linné between 2013 and 2017. A total of 97 samples were analysed and a total of 55 elements were detected. A clear difference in element concentrations was found in Adventdalen and Longyearbyen compared to Kapp Linné for both essential and non-essential elements as well as some differences within areas in Kapp Linné. Cu, Fe, Zn, Al, Cr and Tl were significantly higher in Adventdalen and Longyearbyen, whereas Ca, Mg, S, Cd, Pb and Se were significantly higher in Kapp Linné. Concentration differences within Kapp Linné included significantly higher levels of Se in the north and significantly lower levels of Hg in the South area.

The present study clearly shows different element levels between the locations. Most of these differences can be explained either by the different geology in Adventdalen and Kapp Linné or by different sources of contamination, including both local and long-range sources. Kapp Linné experiences more precipitation during the year compared to Adventdalen and Longyearbyen and toxic elements such as Hg, Cd and Pb may enter the environment at a higher rate in this area. Indeed, a positive relationship was identified between snow-depth and faecal levels of Pb and Hg. Once in the Arctic environment, elements can be taken up by soil and plants such as moss and lichen and thus affect both the biota and the reindeer feeding at contaminated areas.

No significant correlation between levels of As, Hg, Cd or Al were seen for reindeer feeding close to the road in Adventdalen, while Cd was significantly correlated with the distance to the coal power plant, with more Cd further away from the power plant. Cd was also positively correlated with distance to the ocean near Kapp Linné, with more Cd in faeces collected close to the ocean.



## Sammendrag

Arktis regnes som en deponerings- og akkumuleringsregion for flere mulige giftstoffer, inkludert ikke-essensielle metaller, som kan transporteres til det arktiske miljøet via luftstrømmene fra forurensningskilder andre steder i verden. Denne avhandlingens overordnede mål var å sammenligne konsentrasjoner av essensielle og ikke-essensielle metaller i ekskrementer fra svalbardreinsdyr fra Adventdalen, Longyearbyen og Kapp Linné, Svalbard. Adventdalen og Longyearbyen er utsatt for menneskelig aktivitet i form av pågående gruvedrift, kommersiell flytrafikk og eksos fra bil-, snøscooter- og båttrafikk. Avstanden fra veien og kullpipa ble derfor også målt for å undersøke om reinsdyr som beiter nærmere disse mulige forurensningskildene hadde høyere konsentrasjoner av giftige metaller. Kapp Linné derimot, er et område med antatt minimal påvirkning fra menneskelig aktivitet, men er mer utsatt forurensing som kommer fra regn eller hav. Avstanden fra havet ble derfor målt på Kapp Linné for å undersøke om ekskrementene inneholdt høyere konsentrasjoner av toksiske metaller som tidligere har blitt assosiert med det marine miljøet.

Ekskrementer fra svalbardreinsdyr ble samlet i Longyearbyen, Adventdalen og Kapp Linné mellom 2013 og 2017 for å oppnå en bedre forståelse av sammensetningen av giftige metaller ved de forskjellige lokasjonene på Svalbard. Totalt ble 97 prøver analysert og totalt 55 metaller ble identifisert. Forskjellige konsentrasjoner av metaller ble funnet i Adventdalen og Longyearbyen sammenlignet med Kapp Linné for både essensielle og ikke-essensielle metaller, samt forskjeller innad områdene i Kapp Linné. Cu, Fe, Zn, Al, Cr og Tl var signifikant høyere i Adventdalen og Longyearbyen, mens Ca, Mg, S, Cd, Pb og Se var signifikant høyere i Kapp Linné. Innad på Kapp Linné var Se signifikant høyere i den nordlige delen, mens Hg var signifikant lavere i den sørlige delen av Kapp Linné.

Denne oppgaven viser klart at det er forskjellig metallkonsentrasjoner i de to ulike områdene som ble undersøkt. De fleste av disse forskjellene kan bli forklart av geologien i Adventdalen sammenlignet med Kapp Linné eller av forskjellige forurensningskilder. Kapp Linné opplever mer nedbør i løpet av året sammenlignet med Longyearbyen og Adventdalen, og giftige metaller som Hg, Pb og Cd kan entre det arktiske miljøet i større grad her. Metaller i det arktiske miljøet kan bli tatt opp i jord og i planter som mose og lav og kan dermed påvirke både biota og reinsdyr som beiter på forurensede områder.

Ingen signifikant korrelasjon mellom konsentrasjoner av As, Hg, Cd eller Al ble observert for reinsdyr som beitet nære veien i Adventdalen. Cd var signifikant høyere lenger unna kullpipa i Longyearbyen og Adventdalen, og signifikant høyere i ekskrementer samlet nærmere havet.



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## Abbreviations

Al	Aluminium	Nb	Niobium
Au	Gold	Nd	Neodymium
Ag	Silver	Ni	Nickel
As	Arsenic	P	Phosphorus
Ba	Barium	Pb	Lead
Be	Beryllium	Pr	Praseodymium
Bi	Bismuth	Rb	Rubidium
B	Boron	REE	Rare earth element
Ca	Calcium	S	Sulphur
Cd	Cadmium	Sb	Antimony
Ce	Cerium	Sc	Scandium
Co	Cobalt	Se	Selenium
Cr	Chromium	Si	Silicon
Cu	Copper	Sm	Samarium
Cs	Caesium	Sn	Tin
Dy	Dysprosium	Sr	Strontium
Er	Erbium	Tb	Terbium
Fe	Iron	Th	Thorium
Ga	Gallium	Ti	Titanium
Ge	Germanium	Tl	Thallium
Hf	Hafnium	Tm	Thallium
Ho	Holmium	U	Uranium
Hg	Mercury	V	Vanadium
K	Potassium	W	Tungsten
La	Lanthanum	Y	Yttrium
Li	Lithium	Yb	Ytterbium
Lu	Lutetium	Zn	Zink
MeHg	Methylmercury	Zr	Zirconium
Mg	Magnesium		
Mn	Manganese		
Mo	Molybdenum		
Na	Sodium		

# 1. Introduction

The Arctic region is particularly sensitive to pollution, compared to other regions in the world (Sundseth et.al., 2015). Although the Arctic environment is still considered as marginally affected by anthropogenic influences, the combination of local pollution and long-range transport has been shown to contribute to environmental pollution in the Arctic, including the Norwegian archipelago in the Arctic ocean: Svalbard (Marqués et.al., 2017). The Polar regions are considered as deposition and accumulation regions for many long-range transport pollutants due to a process called global transport or the grasshopper effect (Gouin et.al., 2004, Semeena et.al., 2005). Volatile pollutants, including persistent organic pollutants (POPs) and toxic non-essential elements, can evaporate or attach to particles and thus travel in the atmosphere from warmer countries towards colder areas. This happens repeatedly towards the Arctic where they can accumulate due to the cold climate that causes low evaporation rate, decreasing the grasshopper effect and trapping the chemicals in the Arctic (Semeena et.al., 2005, Gouin et.al., 2004).

After atmospheric fallout through condensation, toxic elements can be taken up by the soil or by plants such as moss and lichen (Halbach et.al., 2017, Marqués et.al., 2017, Kallenborn et.al., 2012) and contribute to uptake and deposition in soil and plants, and possible bioaccumulation in food chains (WHO, 2007). In contrast to POPs, elements, including essential elements and toxic non-essential trace elements, are naturally occurring and ubiquitous in the environment (Casarett and Doull, 2013). Thus, their concentrations in the Arctic environment are not only influenced by local or long-range anthropogenic sources of pollution, but also by geological conditions in soil and bedrock (Prestrud et.al., 1993). The Svalbard reindeer (*Rangifer tarandus platyrhynchus*), which is an endemic reindeer sub-species on Svalbard, feeds on ridges, mountain slopes and plateaus, with moss, lichen, shrubs and graminoids as the main food source (Hayashi et.al., 2014), exposing them to toxic non-essential elements originating both from long-range transport and local sources of pollution, and from the local bedrock.

## 1.1 Trace elements

Trace elements are divided into essential and non-essential elements, whereof the non-essential elements are toxic to organisms above certain threshold values and essential elements are required for the organism to maintain bodily function. Nonessential toxic metals can also mimic essential metals and thereby gain access to key cellular functions. This can cause

bioaccumulation of toxic metals without any known biological function (Casarett and Doull, 2013). Adequate concentrations of essential elements are important for the health of humans and animals, and thus the information gained about the concentrations of essential elements in the environment and organisms gives important information about health conditions.

### **1.1.1 Essential Trace Elements**

Essential elements are required in certain amounts in the body. They act as catalytic or structural components in larger molecules and have specific functions (Mertz, 1981). In organisms, they are important cofactors in enzymes, like iron (Fe) in haemoglobin, which is essential for the binding of oxygen in blood. Other essential elements are for example calcium (Ca), which is part of bones, and potassium (K), which is important for nerve and heart function (Casarett and Doull, 2013).

Marginal or severe trace element imbalance below the optimal physiological concentrations, however, can cause organisms to suffer from deficiencies (Hester et.al., 2006). This can lead to different risk factors for several diseases of health importance (Mertz, 1981). The essential trace elements investigated in this thesis are sodium (Na), phosphorus (P), magnesium (Mg), sulphur (S), potassium (K), calcium (Ca), iron (Fe), zinc (Zn) and copper (Cu). An example of an essential element that can cause deficiency if the requirement is not reached is Fe. Because Fe is an important cofactor in haemoglobin in red blood cells, a deficiency can lead to a loss in red blood cell count, also termed anaemia (Balentine et.al., 2017). Another important element for animals is Mg. Mg is the fourth most abundant mineral in the body and is required for DNA and RNA synthesis, reproduction and protein synthesis. A deficiency in Mg could result in unwanted neuromuscular, cardiac or nervous disorders (Gröber et.al., 2015).

### **1.1.2 Non-essential trace elements**

Toxic non-essential elements are naturally occurring trace components, but the levels of many of these have increased in the environment due to industrial, agricultural and mining activities (Olmedo et.al., 2013). The general mechanism for non-essential trace elements is to first bind to biomolecules, and then disturb the metal homeostasis and cause formation of reactive oxygen species (ROS). ROS can lead to harm by causing lipid peroxidation, oxidation of DNA and oxidation of proteins (Casarett and Doull, 2013), as well as disrupt functions in vital organs and glands such as heart, brain, kidney, bone and liver by accumulating (Singh et.al, 2011). In this thesis, the non-essential elements investigated are arsenic (As), cadmium (Cd), selenium (Se), thallium (Tl), mercury (Hg), lead (Pb), chromium (Cr) and aluminium (Al).

Hg is an example of a toxic element that can be found in Arctic biota due to long-range transport caused by its long residence time in the atmosphere of about one year (Born et.al., 1991). Hg can be converted biologically to methylmercury (MeHg) in soil and water. This compound is a potent neurotoxic chemical (WHO, 2007), which can cross both the blood-brain barrier of mammals as well as the blood-placenta barrier, and cause either death or damage to the fetus. MeHg can also bind to proteins and has been shown to biomagnify through a diverse range of food webs (Dehn et.al., 2006, Jæger et.al., 2009).

Cd, Pb and As are other toxic elements that have been widely found and investigated in the Arctic (AMAP, 2011). Sources for Cd is for example as a by-product of Zn and Pb smelting and in batteries (Casarett and Doull, 2013), and it is known to cause kidney and bone damage (WHO, 2007). Sources for Pb is for example paint, old pipes and leaded gasoline, and it is known to interfere with enzyme activity and replace calcium in bone (Casarett and Doull, 2013). As is an ubiquitous, naturally occurring metalloid that can cause health problems for humans and wildlife. It has the ability to attach to small particles in the air and travel long distances (Chang et.al., 2014). Fish and seafood is the primary sources of As for humans, and once absorbed, it has been shown to cause formation of ROS and inhibit lipids and enzyme activity (Molin et.al., 2015)

Se and Cr can act as both an essential and a toxic elements to animals (Yakubov et.al., 2014, Boehler et.al., 2014, Mannot et.al., 2014). Se toxicity is not well understood, but it is associated with several neurological conditions, while Cr has been proven to be carcinogenic (Yakubov et.al., 2014). Thallium is a rare element found in the earth's crust that is highly toxic to living organisms. It can remain in the environment for long periods of time and be transported to Arctic regions via long-range transport (Peter et.al., 2005). Al is not a required element for humans or plants, but is the fourth most common element in soil nevertheless (Frankowski, 2016). Not much is known about the toxicity of Al, but studies suggest an effect of Al on the metabolism of essential elements, including Ca, Mg, Cu, Zn and Fe (Bellés et.al., 2001).

Excretion of elements takes place mainly through the kidneys in urine or in faeces through the gastrointestinal mucosa. Both physical and chemical properties of the compounds can affect the excretion of different chemicals. In humans, Pb is mainly excreted through urine (75%), and only a small fraction is excreted in the faeces (16%). MeHg is the most studied Hg compound due to its toxicity. 90% of MeHg is excreted in the urine. Cadmium (Cd) is excreted slowly, while excretion of Se and Tl are not well studied (Vercruysse, 2000).

### **1.1.3 Rare Earth Elements**

Rare earth elements (REEs) are a group of 17 elements (Pourret et.al., 2018) that often occur together in the same ore deposits and exhibit similar chemical characteristics (Obhodaš et.al., 2018). Despite the name, REEs are not that rare on earth, but widely distributed in the earth's crust (Pourret et.al., 2018). In the current study, 13 REEs will be considered; Cerium (Ce), dysprosium (Dy), erbium (Er), holmium (Ho), lanthanum (La), lutetium (Lu), neodymium (Nd), praseodymium (Pr), samarium (Sm), scandium (Sc), terbium (Tb), ytterbium (Yb) and yttrium (Y). As rare earth elements occur together, these elements will be investigated to examine if there are geological differences at the different sampling locations.

## **1.2 Svalbard**

Svalbard, which lies at ~78°N to ~15°E in the Barents Sea, has several marine and terrestrial ecosystems that can be affected by both anthropogenic and local sources (Halback et.al., 2017, Nuth et.al., 2010). Svalbard has a high geological diversity in a relatively small area. Svalbard was below sea level for a long period of its history, and mud, sand, gravel etc. was continuously deposited and later turned into parts of the special geology that Svalbard consists of today. The landscape is naked, with little soil and no forests or agriculture (Elevold et.al., 2007). Longyearbyen is the largest settlement in Svalbard with approximately 2000 inhabitants (Longyearbyen Lokalstyre, 2012). The city is located in Adventdalen, which is a 40-kilometer-long valley on the west coast of Spitsbergen (Halbach et.al., 2017). Human activity in Longyearbyen includes an all-year open airport, a small coal-fired power plant, traffic from cars and snow mobiles and a coal mine called Gruve 7, which is situated in Adventdalen, 16 km east of Longyearbyen. Gruve 7 has an annual coal production of 60 000 tonnes mainly used for local energy and heating (Granberg et.al., 2017). Runoff from both Gruve 7 and other closed mines have been characterized as acid drainage with high levels of toxic elements such as Ni, Cu and As (Granberg et.al., 2017).

Kapp Linné is located on the north of Nordenskiöldkysten on the west coast of Svalbard, south of the entrance to the Isfjord (Barr, 2009). The coast consists of beaches and steep cliffs with flat and barren tundra behind. The area is known for diverse geology, with both sedimentary and metamorphic rocks (Stange, 2013). Isfjord Radio was built in 1933 and provided radio communication between several settlements on Svalbard and Norway, as well as guiding the ships safely to the area (Johannessen, 2017). Today, Isfjord Radio is used as a hotel with tourists arriving on snow mobiles in the winter and by boat in the summer. Several Svalbard reindeer

feed and breed in this relatively undisturbed area, (Stange, 2013). The goal is therefore to compare levels of elements in and close to an inhabited area with ongoing mining activity and high levels of other human activities to an area with presumed minimal impact from human activity.

### **1.2.1 Elements in the Arctic biota**

According to the Arctic Monitoring and Assessment Programme (AMAP), a large amount of heavy metals in the Arctic arrives via long-range atmospheric transport from lower latitudes. This is mainly due to human emissions or from natural sources such as volcanoes and weathering of rock naturally rich in for example Hg. Wildlife species that are at the limit of their tolerance to stressors, for example due to starvation, will be particularly affected by heavy metals in biota (AMAP, 2011). The Arctic environment at Svalbard is affected by long-range pollutants originating from western Europe and Russia, and to a lesser extent in North America (Grodzinska et.al., 1991). The main sources for toxic non-essential element emissions are fossil fuel combustion, non-ferrous metal production and waste incineration (AMAP, 2011). Elements that form volatile compounds, such as sulphur or nitrogen, or are present at a lower particle radius, may be readily released into the atmosphere from burning of coal and other industrial processes (Kabata-Pendias, 2011).

Local anthropogenic sources of pollution in Svalbard include coal production, combustion release of smoke from the coal power plant, motor vehicle exhaust, airport activities and tourism, including snow mobile and boat activities (Granberg et.al., 2017). During the tertiary period (65-2 million years ago), large swamps were formed at Svalbard, giving rise to the coal deposits around Longyearbyen, Sveagruva and Barentsburg (Kjærnet, nd). Coal can be defined as an organic rocklike natural product that is a result of the decay and maturation of floral remains over geological time (Speight, 1994). Currently (2018), two coal mines are still active at Svalbard; one in Longyearbyen and one in Barentsburg. The main types of pollution from the coal mining activities include elements such as Fe, Cu, Mg, Cd, As, Ni and Hg, and coal mining has therefore long been widely identified as a possible source of pollution on Svalbard (Granberg et.al., 2017). Local non-anthropogenic sources of toxic elements on Svalbard include bedrock, soil and sea spray (Kabata-Pendias, 2011).

### **1.2.2 Svalbard biota**

In Svalbard, plant growth and distribution are limited by low mean summer temperatures (Van der Wal et.al., 2014), but also constrained by large temperature fluctuations, a short growth

season, nutrient deficiency, wind exposure, permafrost and soil movement. The most important factors determining plant distribution are temperature, bedrock type, soil texture and topography. The permafrost melts 30-150 cm every summer, which makes it possible for plants to grow in the shallow frost-free layer of the soil. Granitic bedrock results in an acidic soil type with low pH, while sedimentary bedrock contains Ca and supports nutrient rich soil with higher pH (Overrein et.al., 2015). An acidification in soil can lead to an increase in mobility of metals bound to soil (Abollino et.al., 2003). The diversity of plants on Svalbard is closely related to the climate and it would become strongly affected should the climate conditions change (Overrein et.al., 2015). Two plants that are widely distributed at Svalbard are moss and lichen.

Moss and lichen have commonly been used as a bioindicator of heavy metal pollution (Harmens et.al., 2010). Mosses have a great capacity to absorb and retain heavy metals from precipitation over their entire surface, due to their lack of root system or cuticle layer (Harmens et.al., 2008). Thus, when pollutants are transported to the Arctic, the mosses will obtain pollutants in the precipitation and dry deposition, and the concentration in the moss tissue will reflect the atmospheric deposition. Moss analysis is easier and cheaper than conventional precipitation analysis while also providing a time-integrated measure of the spatial patterns of heavy metal deposition from the atmosphere to terrestrial systems (Harmens et.al., 2008) and are therefore good bioindicator species for heavy metal deposition.

Lichen consists of a symbiotic association of fungi and algae and are widely used as a bioindicator of heavy metal pollution in the Arctic (Wegrzyn et.al., 2016), especially if they have a wide geographic range. Lichen have chitin cell walls, which do not have any barriers for protecting them from contaminants entering through the whole simple body, also called the thallus. The symbiotic fungi and algae have cation exchange properties and grows on rocks in cold, dry and poor nutrient habitats, such as habitats in the Arctic. Lichen are perennial, have a slow growth rate and are largely dependent on atmospheric deposition for their metabolism (Bargagli et.al., 2000). Lichen can accumulate pollutants both from ground and from the air due to uptake of soluble elements rapidly over the entire surface (Bargagli et. al., 2000). Non-essential element atoms can thus enter either via air or via water, and accumulate (Wegrzyn et.al., 2016). Soil particles and aerosols can also be absorbed at the surface and contribute to a higher concentration of elements (Bargagli et.al., 2000). Levels of elements in the soil are the sum of the different concentrations they derive from different sources; such as bedrock, glacial rivers, plant debris, atmospheric dry deposition and precipitation (Wegrzyn et.al., 2016).



### **1.3 Svalbard reindeer**

Svalbard reindeer are a small subspecies of reindeer (*Rangifer tarandus*) and is endemic to the high-Arctic Archipelago of Svalbard (Joo et.al., 2014, Pedersen, n.d.). Being the only endemic mammalian herbivore at Svalbard (Wegener et.al., 1998) is challenging because snow and ice cover most of the vegetation for eight months of the year and the grazing season for Svalbard reindeer is short (Joo et.al., 2014). However, the reindeers manage to accumulate fat during spring and summer and use these reserves to restore their depleted body reserves (Tyler, 1986). Svalbard reindeer have a varied diet which includes almost all types of vegetation in the summer time when food is easily accessible. They are adapted to survive the harsh and variable climate in the Arctic by having low energy demands, an outstanding ability to use their body reserves when the access to food is limited, as well as a thick fur to insulate from low temperatures and wind (Pedersen, n.d., Stange, 2014).

#### **1.3.1 Reindeer faeces**

Faecal matter is the remaining material after food is digested and is composed of proteins, fats, bacterial biomass, carbohydrates and inorganic material. Reindeer faeces have previously been used to investigate population size and biomass, sex structure, habitat use and diet composition (Putman, 1984, Morden et.al., 2011). In addition, faecal matter from Svalbard reindeer can be used to investigate environmental concentrations of POPs and trace elements at Svalbard. Wang et.al (2015) reported polybrominated diphenyl ethers (PBDE), which are flame retardants, in faeces from reindeer feeding in Ny-Ålesund (Wang et.al., 2015). Faecal matter is sensible to collect because it is readily-available and easily-collected source of information, and ecological and toxicological relevant exposure data can be gathered in a non-intrusive way for the animal (Putman, 1984). In addition, Svalbard reindeer will save as much energy as possible, and therefore they do not cover long distances in search of food. This, and the fact that faeces are excreted daily, means that the element composition in the faeces will reflect the element composition in their surrounding environment.

## **1.4 Aim of the thesis**

The available data on element concentrations in polar herbivores is still limited and research concerning elements in Svalbard reindeer is especially scarce. The main objective of this thesis was therefore to investigate concentrations of elements in Svalbard reindeer faeces in Longyearbyen, Adventdalen, and Kapp Linné from 2013 to 2017. These areas were compared due to high human activity including mining, traffic and all year open airport in Longyearbyen and Adventdalen. Kapp Linné, on the other hand, is a pristine area much less influenced by human activity with no mining, airport or local traffic.

Coal is transported in trucks driving on the road in Longyearbyen and through Adventdalen, and coal dust can be transported in the air and accumulate in vegetation close to the road and thus affect animals feeding close to the road. In addition, the pipe from the coal power plant is located in Longyearbyen, which also emits coal smoke that can affect nearby vegetation. Reindeer feeding in different distances from the road and the coal power plant were investigated in Longyearbyen and Adventdalen to examine local pollution effects on the vegetation. Animals in Kapp Linné are more exposed to the ocean and some reindeer have also been observed eating kelp. To examine possible effects of oceanic input of elements in Svalbard reindeer, relationships between the elemental faecal concentrations and the distance from the ocean were examined in Kapp Linné.

Collection of faeces is a good non-intrusive way for assessing metal contamination in the surrounding environment of the animal. Levels of toxic elements in the faeces will reflect concentration of the elements in the local environment and may thus be applied as an integrated proxy of elemental concentrations and help monitor the contamination load in the Arctic environment. The concentration will most likely reflect local bedrock elemental concentration, as well as anthropogenic inputs from both local sources of pollution and from long-range transport of pollution to the local Arctic environment. Two groups of elements were investigated in this study; essential trace elements (Ca, Cu, Fe, Mg, S and Zn) and non-essential trace elements (Al, As, Cd, Cr, Pb, Hg, Se and Tl). In addition, general differences in the pattern of REEs were investigated between the two main areas.

The main objectives of this study were to:

1. Establish whether there are differences in elemental concentration in reindeer feeding at geologically different locations. Both between Adventdalen/Longyearbyen and Kapp Linné and between different areas within Kapp Linné.
2. Establish whether animals feeding closer to the road and the coal power plant in Longyearbyen and Adventdalen, and the ocean in Kapp Linné are more contaminated by toxic non-essential elements compared to animals feeding further away.
3. Establish whether toxic elements known to travel to the Arctic via long-range transport are found in higher concentrations in faeces collected in deep snow.

## 2. Materials and methods

### 2.1 Faecal sampling

Faeces were sampled at multiple locations on the high-Arctic Svalbard archipelago, Norway, in 2013, 2014, 2015 and 2017. Three main areas were selected for their suspected difference in element concentrations; Adventdalen and Longyearbyen for their assumed high levels of toxic elements due to human activity, and Kapp Linné due to its assumed low levels of toxic elements due to low levels of human activity. Kapp Linné was further divided into three areas, with three different sampling parties going to the north, the south and the west of the area, respectively, to collect samples. Adventdalen was also further divided into Adventdalen, Endalen and Todalen. Snow mobiles were used to drive around at the selected areas searching for reindeer. The animals were observed, and faeces were only collected if the animal were identified. By collecting fresh faeces, several other factors could be investigated and controlled for as well. The age (adult/calf), the sex of adults, snow depth and the GPS coordinates for the location at the sampling site were recorded and used when analysing the samples. The samples were placed in plastic zip bags, marked with sample number, date of collection, location, sex of the animal and latitude and longitude.

To prevent contamination of the samples, the faeces were collected with nitrile gloves that were changed between each collection and the samples were stored in zip-lock plastic bags and stored frozen in Longyearbyen until they were transported to Trondheim and stored in the freezer at NTNU. The samples from 2013, 2014, 2015 and some from 2017 were chosen and shipped to Trondheim in 2017 for further analysis. A total of 3 samples were taken from Longyearbyen, with 2 of the samples in the city centre and 1 sample taken approximately 1 km outside the centre. A total of 62 samples were collected from Adventdalen, with 53 from Adventdalen, 5 from Endalen and 4 from Todalen. A total of 32 samples were taken at Kapp Linné, with 15 samples from the north, 8 from the west and 9 from the south. All the samples were collected between January and May. Collection sites are shown in figure 2.1 and 2.2. Total dataset are shown in Appendix C, Table C.1.

By using the GPS coordinates, the distance to the ocean, to the coal power plant and to the road could be measured by using <http://toposvalbard.npolar.no/>. The distance was measured by using the shortest way in linear distance. Distance to the ocean, coal power plant and road was measured in Adventdalen and Longyearbyen samples, while only the distance to the ocean was

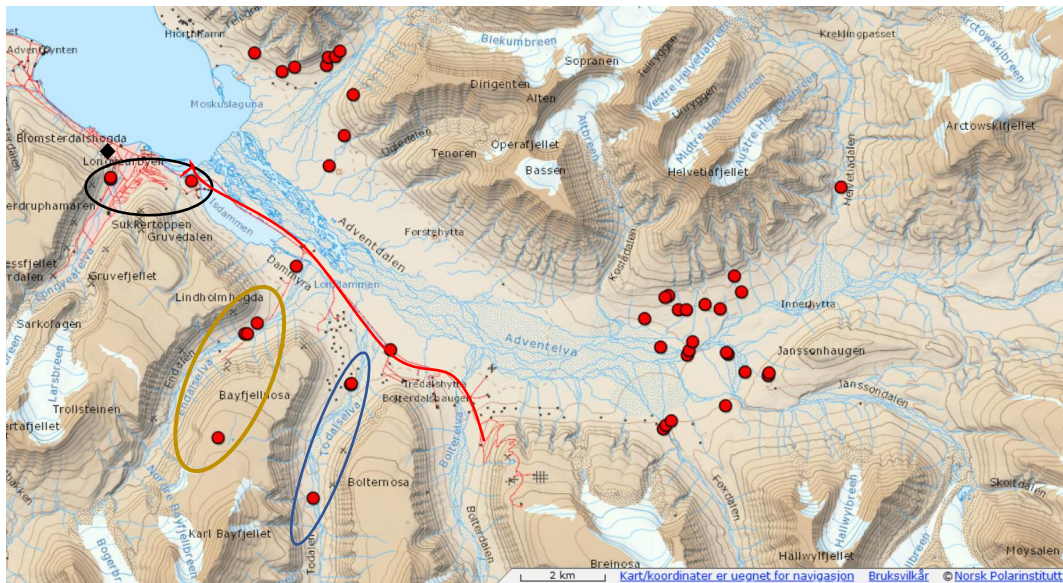
measured in the samples from Kapp Linné due to the lack of roads, coal power plants or other anthropogenic contaminant sources.

### 2.1.1 Sampling sites

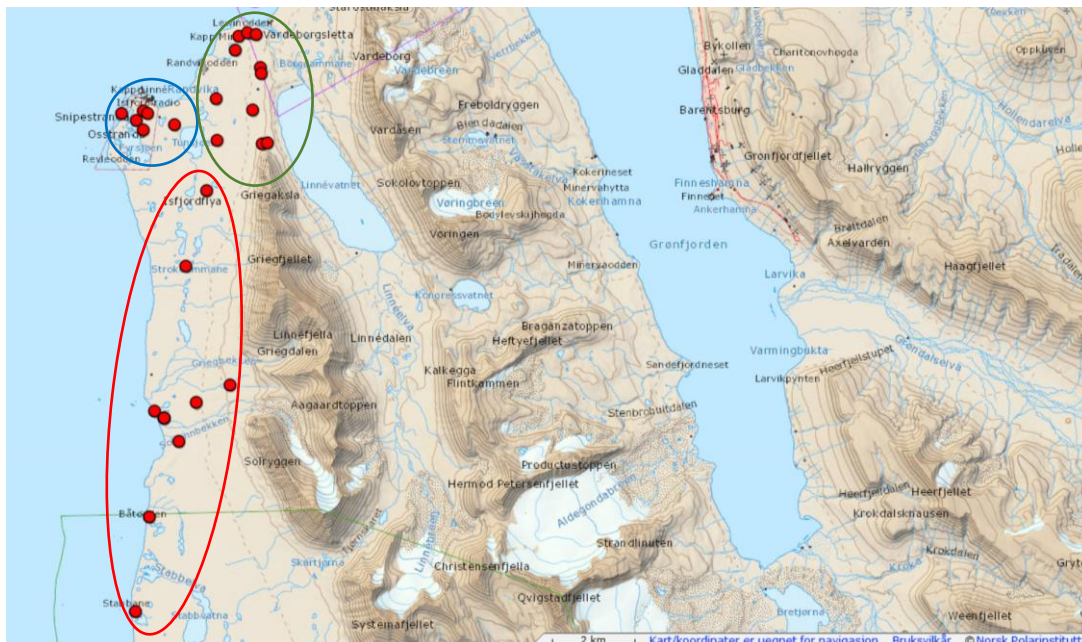
Sampling was conducted on Svalbard (Figure 2.1) with sampling sites in Adventdalen, Longyearbyen (Figure 2.2) and Kapp Linné (Figure 2.3). The different subgroups, the road and the coal power plant are shown.



**Figure 2.1 Map of Spitsbergen showing the sampling sites Longyearbyen and Kapp Linné (Norwegian Polar Institute).**



**Figure 2.2** Sampling in Longyearbyen (black circle,  $n=3$ ) and Adventdalen ( $n=62$ ). Adventdalen was further divided into Adventdalen ( $n=53$ ), Endalen (yellow circle,  $n=5$ ) and Todalen (blue circle,  $n=4$ ). Each red dot indicates one or more reindeer sample. The road is marked in red, and the pipe is marked by a black diamond outside Longyearbyen (<http://toposvalbard.npolar.no/>).



**Figure 2.3** Sampling site at Kapp Linné. Each red dot indicates one or more reindeer sample. Kapp Linné was further divided into subgroups, the north group (green circle,  $n=15$ ), the east group (blue circle,  $n=8$ ) and the south group (red circle,  $n=9$ ) (<http://toposvalbard.npolar.no/>).

## 2.2 Preparation and ICP-MS

To prepare the samples for inductively coupled plasma mass spectrometry (ICP-MS), the samples were freeze-dried for 23 hours in an Alpha 1-2 LDplus Entry Laboratory Freeze Dryer. The lid on the samples was pierced to prevent the samples from exploding under the pressure. Teflon tubes (18 mL) were washed two times with ultra-pure water and a maximum 500 mg of the dried reindeer faeces was added to each tube followed by addition of 60 mg concentrated nitric acid (HNO<sub>3</sub>). All samples were handled with care in a clean and non-contaminated environment and transferred to the Teflon tube either by pouring or by using a plastic spoon. Blanks with HNO<sub>3</sub> and ultra-pure water were used as quality assurance to assess any possible contaminations from the sample preparation.

For digestion, the samples were placed in a high-pressure microwave system (Milestone UltraClave) for two and a half hours to prepare the samples for further laboratory work. The digested samples were then diluted with ultra-pure water until they weighted between 59 and 63 grams (g.). A new tube was then washed with the solution once before filled completely with the solution. These tubes were marked and made ready for ICP-MS.

ICP-MS is a well-established method to detect and quantify elements in various matrices, and it has the ability to detect elements at low limits and scan for many elements at the same time to obtain isotopic information (Thomas, 2008). ICP-MS was performed by Syverin Lierhagen from the Department of chemistry at NTNU. A total of 58 metals were analysed, whereof 55 were above the limit of detection. These included aluminium (Al), antimony (Sb), arsenic (As), barium (Ba), beryllium (Be), bismuth (Bi), boron (B), cadmium (Cd), caesium (Cs), calcium (Ca), cerium (Ce), chromium (Cr), cobalt (Co), copper (Cu), dysprosium (Dy), erbium (Er), gallium (Ga), germanium (Ge), gold (Au), hafnium (Hf), holmium (Ho), iron (Fe), lanthanum (La), lead (Pb), lithium (Li), lutetium (Lu), magnesium (Mg), manganese (Mn), mercury (Hg), molybdenum (Mo), neodymium (Nd), nickel (Ni), niobium (Nb), phosphorus (P), potassium (K), praseodymium (Pr), rubidium (Rb), samarium (Sm), scandium (Sc), selenium (Se), silicon (Si), silver (Ag), sodium (Na), strontium (Sr), sulphur (S), terbium (Tb), thallium (Tl), thorium (Th), thulium (Tm), tin (Sn), titanium (Ti), uranium (U), tungsten (W), vanadium (V), ytterbium (Yb), yttrium (Y), zink (Zn) and zirconium (Zr).

Instrumental detection limits (IDLs) are calculated from the concentration yielding to 25% of relative standard deviation and the correction for the baseline. Limit of detection (LOD) was

calculated from IDLs for the different elements and was chosen to be the highest value between IDL and three times the standard deviation of the blanks.

## **2.3 Statistical analysis**

SIMCA (version 13, Umetrics, Umeå, Sweden) was used to perform principal component analyses (PCA), and R and R studio was used to investigate significance and to create plots that show the concentration of elements in the samples.

### **2.3.1 Principal Component Analysis**

PCA plots were used to investigate the differences in element composition in Longyearbyen, Adventdalen and Kapp Linné at Svalbard, as well as differences within Adventdalen and Kapp Linné. PCA is a multivariate technique that analyses a data table in which observations are described by several inter-correlated quantitative dependent variables. It finds the principal components of the dataset and transforms the data into a new coordinate system, where the first principle component (PC1) describes the highest level of variance, and the second principle component (PC2) describes the second highest level of variance and so on (Abdi et.al., 2010, Hamilton, L., 2014). Variables that cluster together or are in the same area in the plot are likely positively correlated, whereas variables that are positioned oppositely are negatively correlated. By using Hotellings T<sup>2</sup>, with a 95% confidence interval that forms an ellipse around the score plot, possible outliers can be examined.

### **2.3.2 R and R Studio**

R and R Studio were used to perform Student T-tests and to make box plots and scatter plots for the different elements in the samples. For the T-tests, the level of significance was set at  $p < 0.05$ . The Shapiro test was used to test for normal distribution. The data obtained were not always normally distributed, however, a log-transformation did not improve the data to fit a normal distribution. Therefore, two-paired T-tests were used when a normal distribution was given, providing p-values, t-values and degrees of freedom (df). Mann-Whitney U tests were used when a normal distribution was not given, providing w-values. Spearman correlation was used to test the significance for the trendlines in the scatterplots, and intercept and slope of the trendline is presented.

Because there were only three samples from Longyearbyen and no statistical difference was shown between Longyearbyen and Adventdalen samples, these were treated as the same for the box plots. The elements were divided into two groups; essential elements and non-essential elements.



## **3 Results**

Total sample size was 97 faecal samples, including 3 samples from Longyearbyen, 62 samples from Adventdalen and 32 samples from Kapp Linné. A total of 55 elements were above the limit of detection, but only Ca, Cu, Fe, Mg, S, Zn, Al, As, Cd, Cr, Pb, Hg, Se and Tl were examined further in detail. Differences in sex, age (calf/adult) and sampling year were not included due to few samples and an unbalanced design.

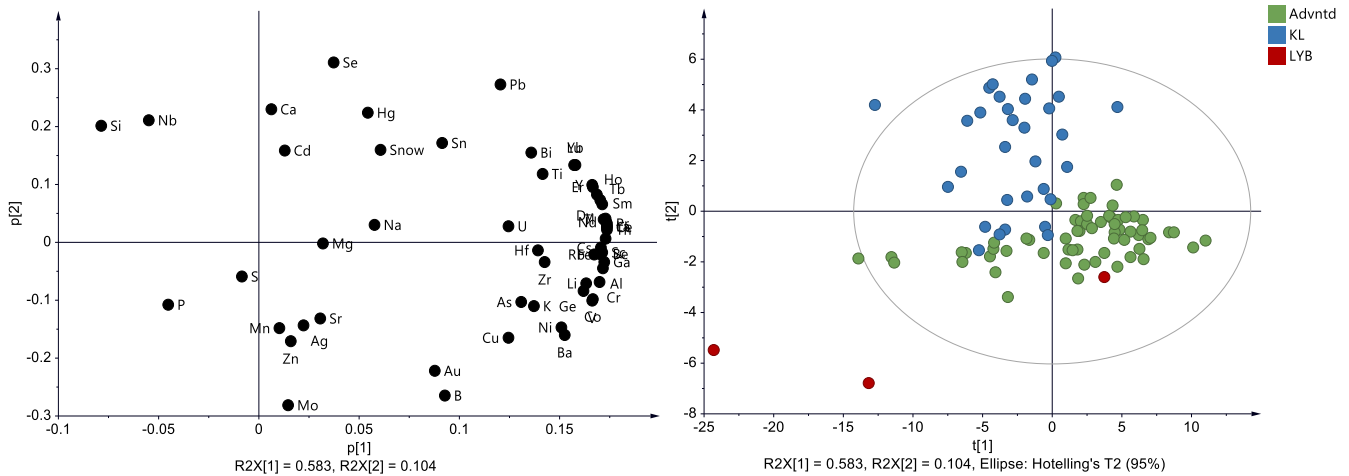
### **3.1 Comparison between Adventdalen, Longyearbyen and Kapp Linné**

To investigate the gross differences in elemental concentrations in Svalbard reindeer faeces between the two main locations: Longyearbyen/Adventdalen and Kapp Linné, the samples from each of these locations were pooled and compared between the areas.

#### **3.1.1 Principal Component Analysis**

The PCA of all reindeer faeces samples, including the different elements found, location, snow depth and distance to ocean, pipe and road resulted in a model with five significant principal components explaining ca. 92% of the variation. The scores PC1, PC2, etc., are new variables summarizing the X-variables. The plots show the possible presence of outliers, groups, similarities and other patterns in the data.

PC1 explained 58.3% of the variation, whereas PC2 explained 10.4%, PC3 explained 9.41%, PC4 explained 5.89%, and PC5 explained 3.23% of the variation. The variables that cluster together are most likely positively correlated to each other, while variables that are further away from each other are most likely negatively correlated.



**Figure 3.1** PCA loading and score plot for all elements in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces samples in Adventdalen (green, n=62), Kapp Linné (blue, n=32) and Longyearbyen (red, n=3). Snow represents the snow depth. For the loading plot, the figure shows principal component 1 (PC1) and 2 (PC2), where PC1 explains 58.3% and PC2 10.4% of the variation. For the score plot, the figure shows principal component 1 (PC1) and 2 (PC2), where PC1 explains 58.3% and PC2 10.4% of the variation. Hotellings T2 ellipse shows four outliers.

Figure 3.1 shows the element composition in Svalbard reindeer faeces from all locations. The PCA loading plot (left) shows that rare earth elements and some other elements, such as Al and Cr, are clustered together in the right site of the PCA plot. The rest of the elements are more scattered in the PCA plot. The score plot (right) shows large differences between locations. The samples from Adventdalen (green) seem to be clustered together, with one sample marked as an outlier according to Hotellings T2 ellipse. One sample from Longyearbyen (red) is located together with the samples from Adventdalen, indicating that this sample is positively correlated to the Adventdalen samples. The two other samples from Longyearbyen are marked as outliers. The samples from Kapp Linné (blue) are also clustered together with one outlier, indicating a different element composition in Kapp Linné compared to Adventdalen and Longyearbyen. When considering the loading and score plot, it is indicated that faeces from Adventdalen and Longyearbyen had higher levels of elements such as Cu, Fe, Zn, Al, Cr and Ti, while faeces from Kapp Linné had higher levels of Ca, Mg, Hg, Cd, Se and Pb. This was investigated further to examine if the differences between the areas were significant.

### 3.1.2 Element concentrations in Adventdalen and Longyearbyen compared to Kapp Linné

The essential elements included Ca, Cu, Fe, Mg, S and Zn, while the non-essential elements included Al, As, Cd, Cr, Pb, Hg, Se, Tl and Al. Al is placed together with the essential elements, even though it is a non-essential element, due to its high concentration in the samples.

Due to similar geology, small sample size and no significant difference between faecal concentrations in Adventdalen and Longyearbyen, samples from Longyearbyen were included in the samples from Adventdalen, and reindeer feeding in Adventdalen and Longyearbyen were compared to animals feeding in Kapp Linné.

#### Essential Elements

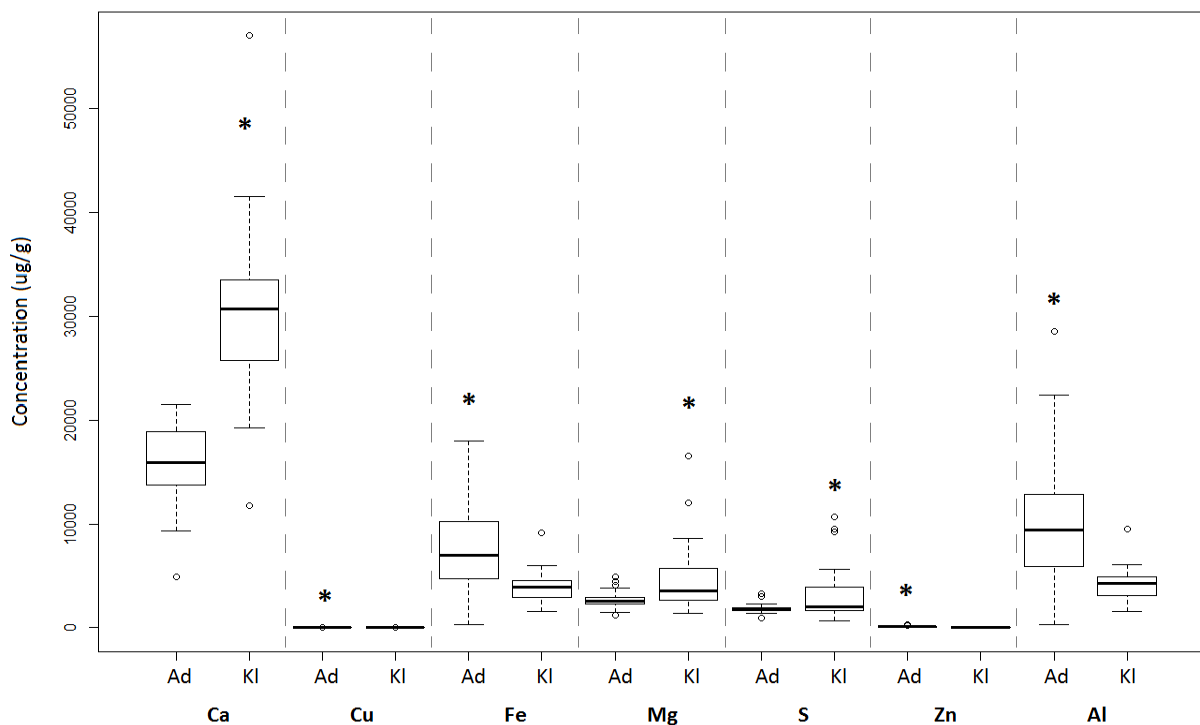


Figure 3.2 Boxplot of essential elements in Adventdalen and Longyearbyen (Ad, n=65) compared to Kapp Linné (Kl, n=32) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces from 2013 to 2017. Asterisks (\*) denote significant difference between the two locations, with level of significance set at  $p < 0.05$ . Copper (Cu), iron (Fe), zinc (Zn) and aluminium (Al) were statistically higher in Adventdalen and Longyearbyen compared to Kapp Linné, whereas calcium (Ca), magnesium (Mg) and sulphur (S) were significantly higher in Kapp Linné.

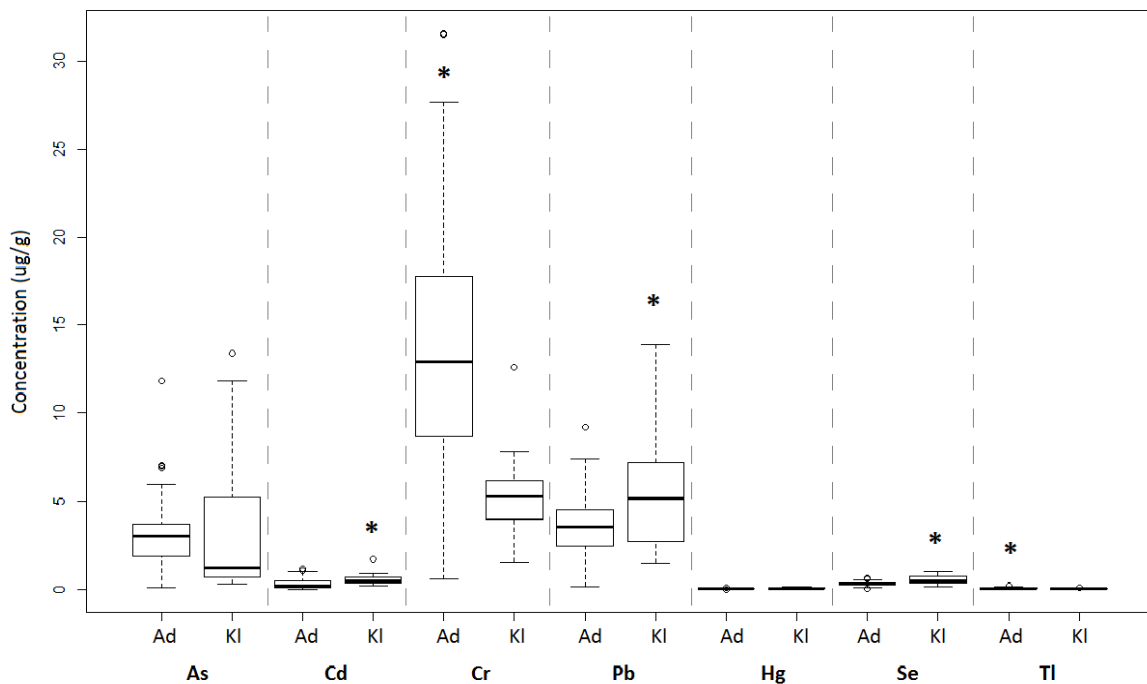
Figure 3.2 shows that Cu, Fe, Zn and Al were statistically higher in Adventdalen and Longyearbyen with p-values of  $p < 0.0001$ . W-value for Cu and Al were  $w = 1776$  and  $1724$ , respectively. T-values and df for Fe and Zn were  $t = 4.42$  (df=88) and  $4.55$  (df=94), respectively.

Cu, Fe, Zn and Al were statistically higher in Adventdalen and Longyearbyen with mean values  $8.53 \mu\text{g/g}$  in Adventdalen and  $5.71 \mu\text{g/g}$  at Kapp Linné for Cu,  $7498.41 \mu\text{g/g}$  in Adventdalen and  $4703.84 \mu\text{g/g}$  at Kapp Linné for Fe,  $82.72$  in Adventdalen and  $57.65 \mu\text{g/g}$  at Kapp Linné for Zn and  $9658.25 \mu\text{g/g}$  in Adventdalen and  $4193.98 \mu\text{g/g}$  at Kapp Linné for Al.

Ca, Mg and S were significantly higher in Kapp Linné with p-values of  $p < 0.0001$ ,  $p = 0.0001$  and  $p = 0.02$ , respectively. T-values and df for Ca and Mg were  $-9.67$  (df=37),  $-3.56$  (df=32), respectively. W-value for S were  $731$ .

Mean concentration for the different elements were  $15952.68 \mu\text{g/g}$  in Adventdalen and  $30290.94 \mu\text{g/g}$  at Kapp Linné for Ca,  $2651.64 \mu\text{g/g}$  in Adventdalen,  $4703.84$  at Kapp Linné for Mg and  $1811.39$  in Adventdalen and  $3082.32 \mu\text{g/g}$  at Kapp Linné. Table for the mean values for essential elements are found in Figure C3 in Appendix C.

## Non-Essential Elements



**Figure 3.3** Boxplot of non-essential elements in Adventdalen and Longyearbyen (Ad, n=65) compared to Kapp Linné (KI, n=32) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces from 2013 to 2017. Asterisks (\*) denote significant difference between the two locations, with level of significance set at  $p < 0.05$ . Chromium (Cr) and thallium (Tl) were significantly higher in Adventdalen and Longyearbyen, while cadmium (Cd), lead (Pb) and selenium (Se) were significantly higher in Kapp Linné.

Figure 3.3 shows that the non-essential elements Cr and Tl were significantly higher in Adventdalen and Longyearbyen compared to Kapp Linné with p-values of  $p < 0.0001$ . T-values and df were  $t = 8.89$  (df=82) and  $4.18$  (df=94).

Mean values for Cr were  $13.2 \mu\text{g/g}$  in Adventdalen and Longyearbyen compared to  $5.3 \mu\text{g/g}$  in Kapp Linné and  $0.08 \mu\text{g/g}$  in Adventdalen/Longyearbyen compared to  $0.05 \mu\text{g/g}$  in Kapp Linné for Tl.

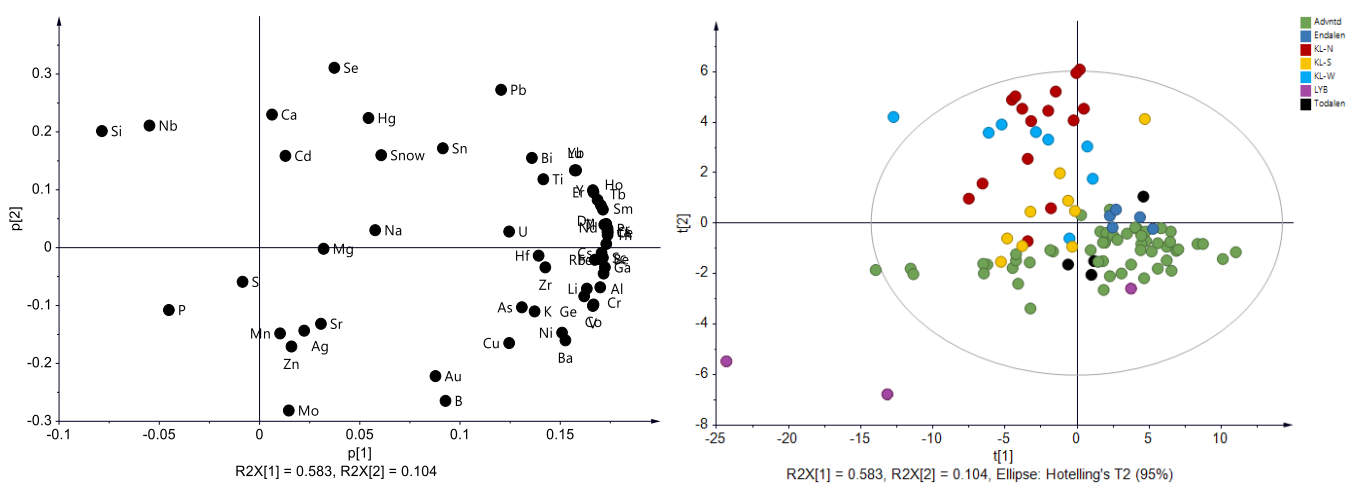
Cd, Pb and Se were significantly higher in Kapp Linné with  $p = 0.0002$ ,  $p = 0.003$  and  $p < 0.0001$ . W-values for Cd were  $w = 0.84$ , while t-values and df for Pb and Se were  $t = -3.18$  (df=41) and  $-4.90$  (df=36), respectively.

Mean values for Cd, Pb and Se were  $0.6 \mu\text{g/g}$  in Kapp Linné compared to  $0.3 \mu\text{g/g}$  in Adventdalen/Longyearbyen for Cd,  $5.4 \mu\text{g/g}$  compared to  $3.6 \mu\text{g/g}$  for Pb and  $0.6 \mu\text{g/g}$  compared to  $0.3 \mu\text{g/g}$  for Se.

Neither As nor Hg concentrations differed significantly between Adventdalen and Kapp Linné, with a mean of 3.13  $\mu\text{g/g}$  in Adventdalen and 3.29  $\mu\text{g/g}$  for Kapp Linné for As and 0.07  $\mu\text{g/g}$  for Adventdalen and 0.08  $\mu\text{g/g}$  at Kapp Linné for Hg. Table for the mean values for non-essential elements are found in Figure C4 in Appendix C.

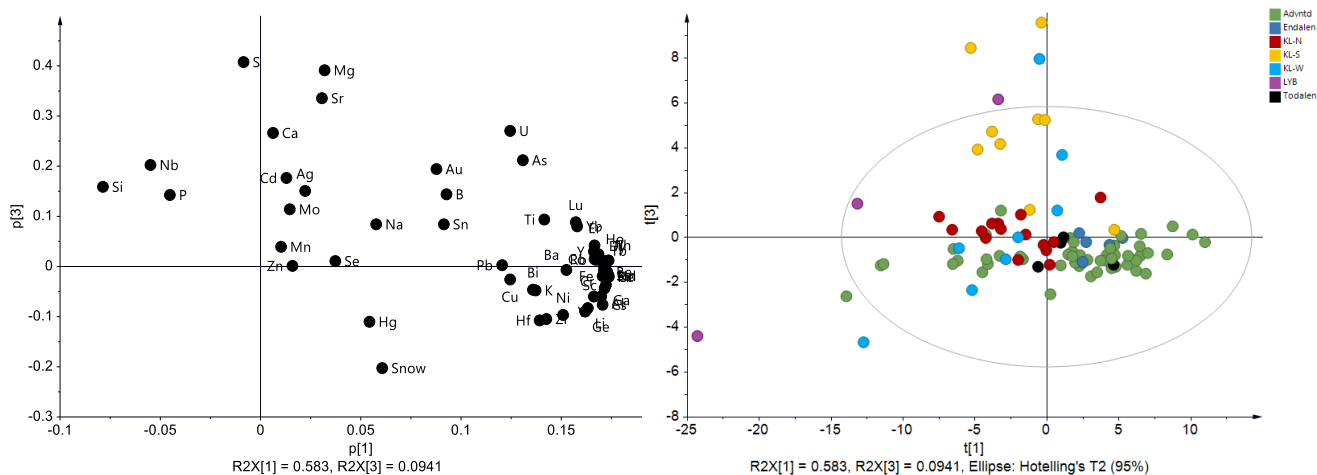
### 3.2 Element concentration at different areas within Kapp Linné

#### 3.2.1 Principal Component analysis



**Figure 3.4** PCA loading and score plot for Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces at different locations. Adventdalen is divided into Adventdalen (green, n=53), Endalen (blue, n=4) and Todalen (black, n=4). Kapp Linné is divided into the north side (KL-N, red, n=15), the south side (KL-S, yellow, n=7) and the west side (KL-W, light blue, n=8). Longyearbyen (LYB, red, n=3) has two outliers and one point positively correlated to the Adventdalen samples. Each sample represents one unique reindeer sample. The figure shows principal component 1 (PC1) and 2 (PC2); PC1 explains 58.2% and PC2 10.4% of the variation. Hotellings T2 ellipse shows four outliers.

The PCA (Figure 3.4) shows PC1 and PC2 and separates Kapp Linné into three areas, north (KL-N), south (KL-S) and west (KL-W), respectively. It also divides Adventdalen into three areas, Adventdalen, Endalen and Todalen. The areas within Adventdalen does not seem to be different. Differences are shown between Kapp Linné, with the north and the west clustering together and the south clustering together with the samples from Adventdalen.



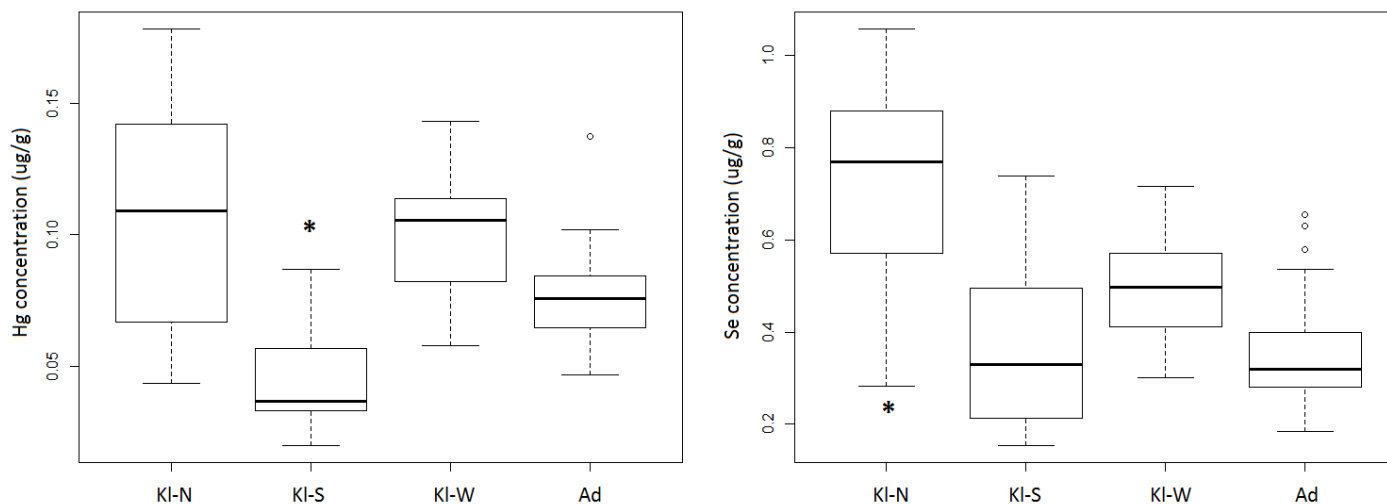
**Figure 3.5** PCA loading and score plot for all elements in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces samples at different locations. For the loading plot, Snow represents the snow depth, with the dot representing where the snow is deepest. The figures show principal component 1 (PC1) and 3 (PC3); PC1 explains 58.3% and PC2 9.41% of the variation. For the score plot, Adventdalen is divided into Adventdalen (green, n=53), Endalen (red, n=4) and Todalen (orange, n=4). Kapp Linné is divided into the north side (KL-N, brown, n=15), the south side (KL-S, yellow, n=7) and the west side (KL-W, blue, n=8). Longyearbyen (LYB, purple, n=3) has two outliers and one point positively correlated to the Adventdalen samples. Hotelling's T2 ellipse shows 7 outliers.

The PCA plot (Figure 3.5) shows PC1 and PC3 and the variation within the samples from Adventdalen and within the samples from Kapp Linné. The samples from Kapp Linné south (KL-S, yellow) are clustered together mostly in the upper part of the plot, along PC1, with some samples located around the origin (0,0). The samples from Kapp Linné north (KL-N, red) are clustered together along PC3, indicating positive correlation within the group. Endalen, Todalen and Adventdalen are clustered together along PC3 as well, indicating low variance within Adventdalen. Regarding the toxic non-essential elements, Se seems to be high in faeces from the north, whereas Cd seems to be high in the south. Cd, however, was investigated, and showed no significant difference between the areas within Kapp Linné. Instead, Hg, which is located opposite to the south, was investigated together with Se to see if these two were negatively correlated (Figure 3.6).

### 3.2.2 Element concentration within Kapp Linné

Hg and Se were investigated within Kapp Linné samples, and Adventdalen samples were included to better understand the difference between Kapp Linné and Adventdalen. Samples

from Longyearbyen was not taken into consideration. As, Cr, Al, Pb, Tl and Cd were also investigated, but none of these showed significant differences between the areas within Kapp Linné and were thus not investigated further.



**Figure 3.6** Boxplots of mercury (Hg) and selenium (Se) concentrations ( $\mu\text{g/g}$ ) in Adventdalen (Ad,  $n=51$ ) and at three different locations within Kapp Linné; north (KI-N,  $n=15$ ), south (KI-S,  $n=9$ ) and west (KI-W,  $n=8$ ), respectively. Asterix's (\*) denote which location is significantly different from the other locations, with a level of significance set at  $p < 0.05$ .

Figure 3.6 shows that Hg was significantly higher in the south area of Kapp Linné compared to the north, the west and Adventdalen with  $p < 0.0001$ ,  $p = 0.003$  and  $p = 0.001$ , respectively. T-values and df were  $t = -4.82$  ( $df=21$ ),  $-4.79$  ( $df=13$ ) and  $-4.29$  ( $df=9$ ), respectively. Regarding Hg, the north and the west of Kapp Linné did not differ from Adventdalen ( $p > 0.05$ ). Mean values for Hg were  $0.11 \mu\text{g/g}$  for north,  $0.045 \mu\text{g/g}$  for south,  $0.10$  for west and  $0.08 \mu\text{g/g}$  for Adventdalen.

Se was significantly higher in the north area of Kapp Linné compared to the south, the west and Adventdalen with  $p$ -values  $< 0.001$ ,  $p = 0.005$  and  $p < 0.0001$ . T-values and df were  $t = 4.05$  ( $df=18$ ),  $3.09$  ( $df=20$ ) and  $6.33$  ( $df=15$ ), respectively.

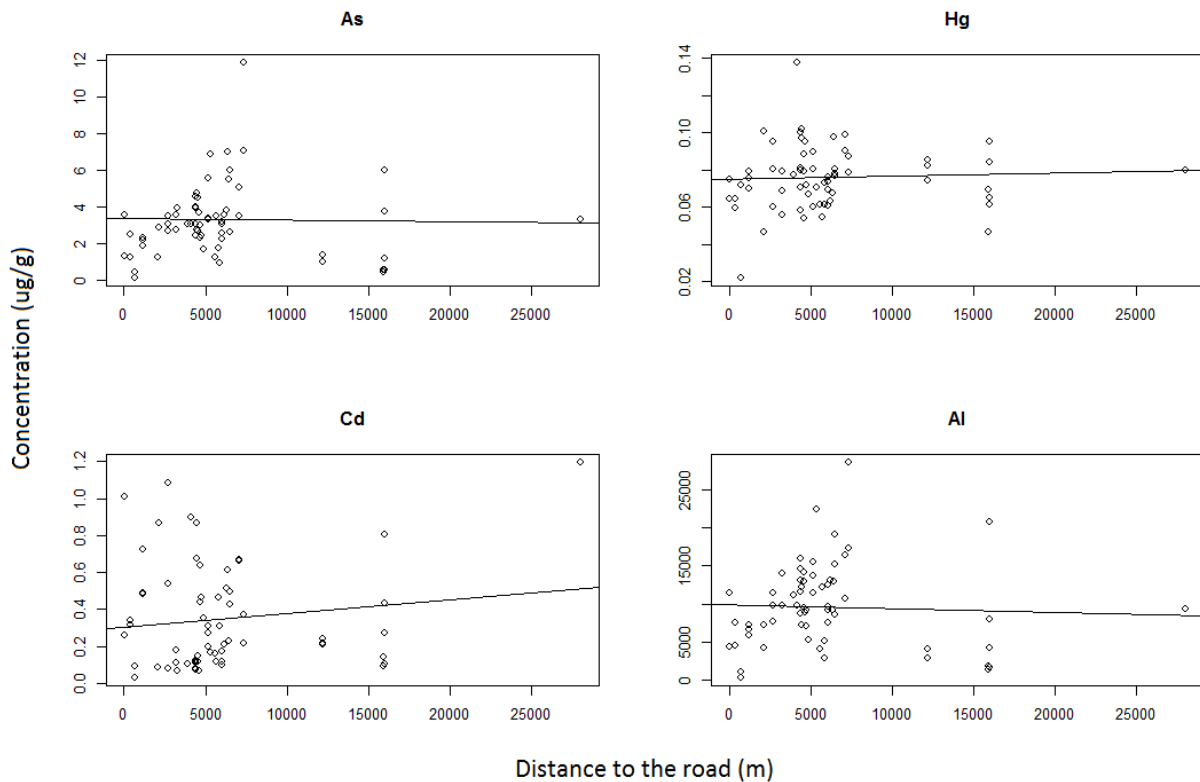
Regarding Se, the south and west of Kapp Linné did not differ from Adventdalen ( $p > 0.05$ ). Mean values of Se were  $0.72 \mu\text{g/g}$  for north,  $0.37 \mu\text{g/g}$  for south,  $0.49 \mu\text{g/g}$  for west and  $0.35 \mu\text{g/g}$  for Adventdalen.



### 3.3 Toxic elements in relation to distance to road, pipe and ocean

This study also investigated if Svalbard reindeer feeding close to the road and to the coal power plant in Longyearbyen and Adventdalen could be affected by toxic non-essential elements such as As, Hg, Cd and Al that had been taken up by nearby soil and plants. Reindeer feeding close to the ocean in Kapp Linné were suspected to contain higher levels of toxic elements of oceanic origin, such as As and Cd, as well as elements stored in kelp such as Ca, Fe, Mg and Cu (Daniels, nd, Hansen et al., 2012). The possible correlation between the distance from the samples to these possible sources of contamination and element concentration was investigated using correlation analysis (Pearson Spearman depending on normal distribution of data).

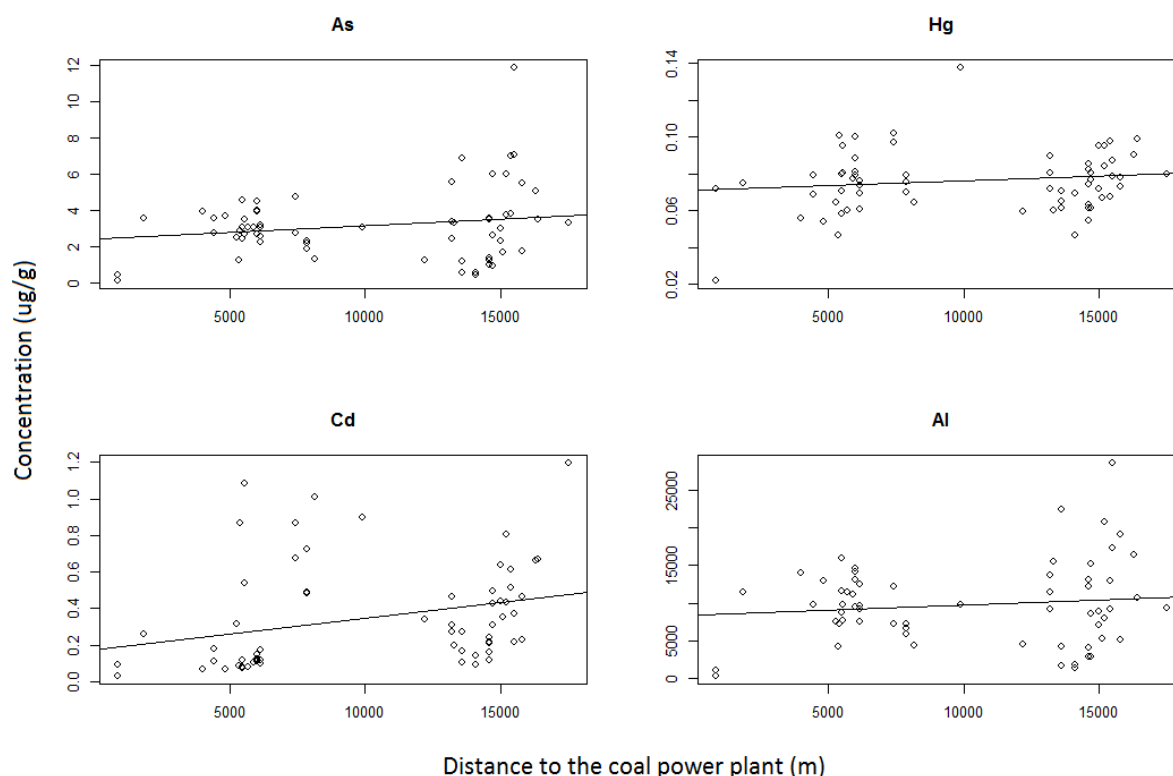
#### 3.3.1 Influence of the road on element concentration



**Figure 3.7** Scatter plot of arsenic (As), mercury (Hg), cadmium (Cd) and aluminium (Al) concentration ( $\mu\text{g/g}$ ) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces in Adventdalen and Longyearbyen ( $n=65$ ) in relation to the distance from the road (m) where the samples were collected. The  $R^2$  values for the trendlines were calculated to be 0.0001, 0.0035, 0.0148 and 0.0032 for As, Hg, Cd and Al, respectively, and  $p$ -values were  $p>0.05$  for all. Equations of the trendlines were calculated to be  $y = 0.0004x + 3.16$ ,  $y = 0.0000002x + 0.07$ ,  $0.000006x + 0.03$  and  $-0.06x + 10035$  for As, Hg, Cd and Al respectively.  $Df = 64$  for all elements.

None of the toxic elements tested were significantly correlated with distance to the road ( $p > 0.05$ ) in Adventdalen and Longyearbyen (Figure 3.7). As, Hg, Cd and Al all had one peak at 5 km, with most of the animals located before or at 5 km and only five or six animals at or further away from 15 km.

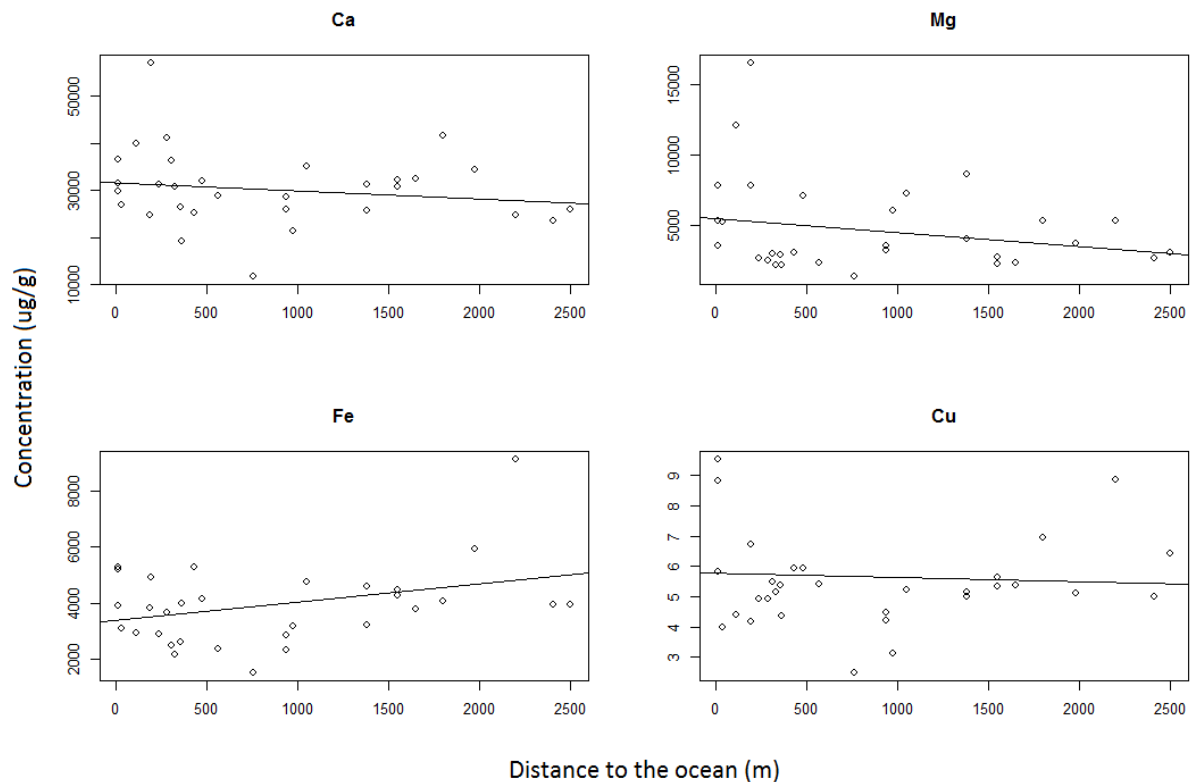
### 3.3.2 Influence of the coal power plant on element concentration



**Figure 3.8** Arsenic (As), mercury (Hg), cadmium (Cd) and aluminium (Al) concentration ( $\mu\text{g/g}$ ) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces in Adventdalen and Longyearbyen ( $n=65$ ) in relation to the distance to the coal power plant in Longyearbyen (m). The  $R^2$  values for the trendlines were calculated to be 0.0405, 0.0242, 0.0703 and 0.0139 for As, Hg, Cd and Al, respectively. P-values were  $p > 0.05$  for As, Hg and Al, and  $p = 0.03$  for Cd. Equations of the trendlines were calculated to be  $y = 0.00008x + 2.28$ ,  $0.0000005x + 0.07$ ,  $0.00002x + 0.19$  and  $0.13x + 8289$  for As, Hg, Cd and Al respectively. Df = 64 for all elements.

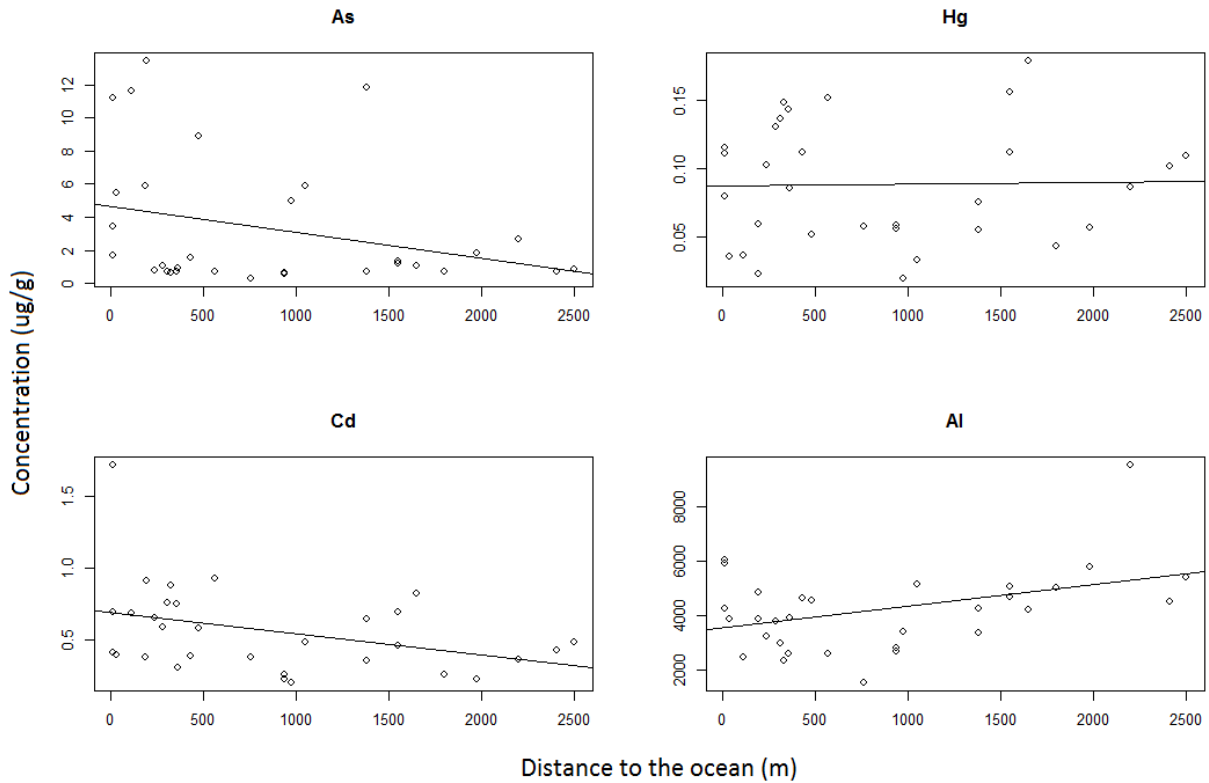
Cd was significantly correlated with the distance to the coal power plant, with less Cd closer to the power plant and higher concentration further away (Figure 3.8). As, Hg and Al were not significantly correlated. As, Hg, Cd and Al all have an increasing trendline, indicating more of the toxic elements further away from the coal power plant. All of the elements also have two peaks, at 5 km and at 15 km, with most of the animals being located at these sites.

### 3.3.3 Influence of the ocean on element concentration



**Figure 3.9** Calcium (Ca), magnesium (Mg), iron (Fe) and copper (Cu) concentration ( $\mu\text{g/g}$ ) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces in Kapp Linné ( $n=32$ ) in relation to distance to the ocean (m). The  $R^2$  values for the trendlines were calculated to be 0.00253, 0.0537, 0.1094 and 0.0015 for Ca, Mg, Fe and Cu respectively, and  $p>0.05$  for all. Equations for the trendlines were calculated to be  $y = -1.66x + 31709$ ,  $-0.90x + 5541$ ,  $0.62x + 3398$  and  $-0.0001x + 5.79$  for Ca, Mg, Fe and Cu respectively. Df = 31 for all elements.

No significant correlation with distance to the ocean was shown for Ca, Mg, Fe or Cu in samples from Kapp Linné (Figure 3.9). Ca and Cu have a trendline that is almost horizontal, indicating no relationship. Mg has a decreasing trendline and Fe has an increasing trendline, indicating a small correlation with the ocean in Mg and no correlation in Fe. The animals were sampled in a gradient between 0 and 25 km from the ocean.

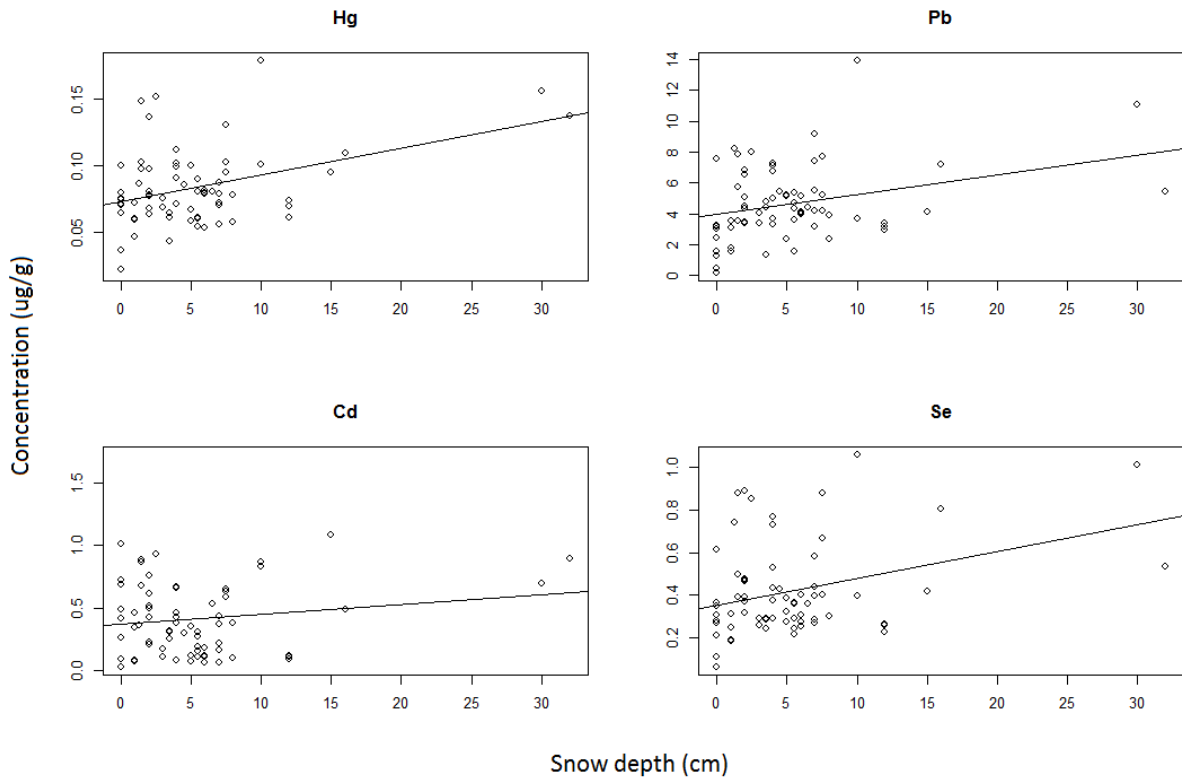


**Figure 3.10** Arsenic (As), mercury (Hg), cadmium (Cd) and aluminium (Al) concentration ( $\mu\text{g/g}$ ) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces in Kapp Linné ( $n=32$ ) in relation to the distance to the ocean. The  $R^2$  values for the trendlines were calculated to be 0.0836, 0.0013, 0.1237 and 0.1678 for As, Hg, Cd and Al respectively. P-values were  $p>0.05$  for As, Hg and Al, and  $p=0.04$  for Cd. Equations for the trendlines were calculated to be  $-0.0015x + 4.55$ ,  $0.000002x + 0.08$ ,  $-0.0001x + 0.67$  and  $0.80x + 3508$  for As, Hg, Cd and Al respectively.  $Df = 31$  for all elements.

Cd was significantly correlated with distance to the ocean, indicating more Cd in samples collected closer to the ocean near Kapp Linné (Figure 3.10). As, Hg and Al were not significantly correlated with a horizontal trendline for Hg and an increasing trendline for Al. As and Cd are the only two that show signs of a downward trend, which could indicate that these two elements originate from marine sources of pollution, although only Cd were significantly correlated. The animals were sampled in a gradient between 0 and 25 km from the ocean.

### 3.4 Element concentration in snow

According to the PCA plot (Figure 3.1 and 3.6), concentrations of Hg and Cd seem to be high in deep snow. These elements were investigated together with Pb and Se, which also have been shown to travel via long-range transport.



**Figure 3.11 Mercury (Hg), lead (Pb), cadmium (Cd) and selenium (Se) concentrations ( $\mu\text{g/g}$ ) in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces sampled at different snow depths (cm). The  $R^2$  values for the trendlines were calculated to be 0.1613, 0.1152, 0.0304 and 0.0936 for Hg, Pb, Cd and Se respectively. P-values were  $p=0.055$  for Hg,  $p=0.011$  for Pb and  $p>0.05$  for Cd and Se. Equations for the trendlines were calculated to be  $y = 0.002x + 0.07$ ,  $0.14x + 3.86$ ,  $0.008x + 0.035$  and  $0.01x + 0.035$  for Hg, Pb, Cd and Se respectively.  $Df = 68$  for all elements.**

Figure 3.11 suggests higher concentrations of Hg, Pb and Se as the snow deepens, which can suggest input of these elements from long-range transport to Svalbard. Pb was significantly correlated with deep snow, while Hg was close to significantly correlated. Se and Cd did not have a clear indication of higher concentration in deeper snow, with a low  $R^2$  value and a  $p>0.05$ . There seem to be more samples where the snow depth was between 0 and 5 cm, and only two samples were taken where the snow was above 20 cm, making it hard to conclude firmly on anything without further investigations.

## **4. Discussion**

### **4.1 Element composition in Adventdalen/Longyearbyen and Kapp Linné**

Faecal sampling and investigation can be used to describe the element concentration in the surrounding environment. Both essential and toxic non-essential elements were detected in faeces from Svalbard reindeer feeding in Longyearbyen, Adventdalen and Kapp Linne, Svalbard. The PCA plot for Adventdalen, Longyearbyen and Kapp Linné (Figure 3.1) indicates variation between the two main locations, with both Adventdalen and Kapp Linné clustered together separately. Only three samples were taken from Longyearbyen, making it difficult to conclude on any correlation between these samples. The three samples were instead analysed together with the samples from Adventdalen.

#### **4.1.1 Essential trace elements**

This study shows significantly higher concentration of Cu, Fe, K and Zn in faeces from Adventdalen and Longyearbyen, and significantly higher concentration of Ca, Mg and S in faeces from Kapp Linné.

Soil acts as a geochemical sink for contaminants, as well as a natural buffer that controls the transport of contaminants and other elements from the atmosphere or biota (Iimura et.al., 1977). Trace elements from atmospheric fallout may reach the surface soil, but their further fate depends on the soil chemical and the physical properties, as well as the speciation of the element. Once in the soil, elements are persistent and are only depleted slowly by leaching, plant uptake, erosion or deflation. Half-life of some non-essential trace elements in soil was measured by Iimura et.al (1977) and showed great variance. For Zn, the half-time was 70 to 510 years, for Cd, 13 to 1100 years, for Cu, 310 to 1500 years, for Pb, 740 to 5900 years (Iimura et.al., 1977). This happens at a much slower rate in cold climates, such as in Svalbard, compared to soils in a more tropical climate, which is what was measured (Kapata-Pendias, 2011, Iimura et.al., 1977). The different soil and rock types in Adventdalen and Longyearbyen compared to Kapp Linné may explain some of the difference between element composition in the reindeer faeces.

A map from the Norwegian Polar Institute (2015) (Appendix A, Figure A.1) shows that Longyearbyen and Adventdalen consist of mostly sedimentary rocks, with sandstone, siltstone, shale and bituminous shale. Kapp Linné has different geology and consists of both sedimentary and metamorphic rocks, with mostly phyllite, quartzite and limestone, with some marble,

sandstone, siltstone, shale and tilloid rocks in alternation with other rocks (Dallmann, 2015). Adventdalen and Longyearbyen are mostly covered in sandstone, which is high in Fe (Marszalek et.al., 2014). This correlates with the findings that Fe is statistically higher in faeces from Adventdalen and Longyearbyen compared to faeces from Kapp Linné and indicates that reindeer foraging in these areas may eat sandstone gravel when consuming plants. Kapp Linné has different geology and consists of limestone, which is a sedimentary rock composed primarily of calcium carbonate ( $\text{CaCO}_3$ ) (Thomas, 1986). In the reindeer faeces samples Ca was significantly higher in Kapp Linné compared to in Adventdalen and Longyearbyen, which could be explained by the presence of limestone in Kapp Linné and its absence in Adventdalen or Longyearbyen. This may result in higher concentration of Ca in the plants in Kapp Linné and thus higher concentration in the faeces. However, it may also be due to consumption of gravel containing Ca, as reindeer faeces have been shown to contain gravel and rocks (Hansen, B.B, pers. comm.) This therefore indicates that uptake and transfer of elements into the diet of the reindeer are an important source for concentration in their faeces. The majority of essential elements are absorbed in the gut by a passive paracellular mechanism and stored (Jahnen-Dechent et.al., 2012). The excess concentrations of essential elements are then excreted by the kidneys or in the faeces (Casarett and Doull, 2013). This makes it difficult to conclude on the essential elements in the faeces. Any animals with low levels of any essential elements will absorb more, while animals with excess levels will excrete more of essential elements.

#### **4.1.2 Non-essential trace elements**

Concerning non-essential toxic elements, this study shows significantly higher levels of Cr and Al in faeces from Adventdalen and Longyearbyen compared to Kapp Linné, and significantly higher levels of Cd, Pb, Se and Tl in Kapp Linné.

It was hypothesized that there would be higher concentrations of toxic elements in Longyearbyen and Adventdalen compared to in Kapp Linné due to more human activity and release of toxic elements from mine drainage, the coal power plant, the airport and exhaust from cars, snow mobiles and boats (Granberg et.al., 2017). However, Cd, Pb and Se were significantly higher in animal faeces from Kapp Linné. A possible explanation can be due to sandstone in Adventdalen and Longyearbyen, which is a fine-grained rock (Appendix A, Figure A.1). Sandstone has been shown to be better at filtering out pollutants from the surface compared to rocks with cracks and crevices such as limestone, which was one of the main rock types in Kapp Linné (Khattak, 2015). This could be one cause of why there are more toxic elements in Kapp Linné compared to in Adventdalen and Longyearbyen.

Another explanation can be that Kapp Linné is more affected by toxic elements such as Hg, Cd, Pb and Se entering the Arctic due to long-range transport. Figure 3.11, as well as different studies (Poikalainen et.al., 2004, AMAP, 2011) suggest that the main source of Hg, Cd and Pb pollution in the Arctic is due to anthropogenic sources via long-range transport. Figure 3.11 showed that Pb was significantly correlated with snow depth ( $p=0.011$ ), while Hg was close to significantly correlated in deep snow ( $p=0.055$ ). This indicates that Hg and Pb may originate from long-range transport entering the Arctic as snow. Hg levels have been seen to rise in some parts of the Arctic, despite reduction in emission from human activity over the past 30 years in some parts of the world (AMAP, 2011). Toxic elements can thus have been transported in elemental form as aerosol particles to this area due to more precipitation in Kapp Linné, which has a mean annual precipitation of 480 mm at the Isfjord Radio compared to 190 mm in Longyearbyen and Adventdalen (Poikalainen et.al., 2004, Watanabe et.al., 2013, Førland et.al., 2011). Even though the snow depth was measured in both Kapp Linné and Adventdalen, more snow will fall in Kapp Linné due to higher precipitation rates, enabling more of the toxic elements to be taken up in soil and plants. Snow depth at sampling locations is also a result of snowdrift and accumulation due to wind, making it difficult to conclude on the significance of these data, although they are sensible due to proven long-range transport of these elements.

Hg and As were not significantly different between Adventdalen, Longyearbyen and Kapp Linné, which can be explained by different sources of pollution between the two areas. Coal burning is the main source for human emission of Hg (AMAP, 2011), while studies have shown As in drainage from mines (Granberg et.al., 2017). There is still one operating mine in Adventdalen (Gruve 7) and levels of Hg and As in Longyearbyen and Adventdalen may therefore originate from coal burning in the coal power plant and the resultant releases into air. This would not be the source of Hg or As in Kapp Linné, as there is no coal mine located there. Instead, high levels of Hg were found in faeces samples collected in areas with larger depths of snow, which could indicate input of Hg from long-range transport. As has been shown as an element that can travel to the Arctic via long-range transport (Pacyna et.al., 1985), but concentrations of As were not significantly correlated with large snow depth (Figure 3.11, Appendix C, Figure C.3).

As has also been shown to be higher in marine animals and plants compared to in terrestrial animals and plants (Lunde, 1977). Kapp Linné is a coastal area with high precipitation rates and reindeer feeding close to the ocean could be exposed to As originating from the marine



environment. This was also shown with a negative relationship between the faecal As concentration and the distance from the ocean in Kapp Linné (Figure 3.10), which would indicate higher concentrations of the element in reindeer faeces closer to the ocean and less in faeces from further away. As was, however, not significantly correlated with distance from the ocean ( $p=0.17$ ). Hg and As may have different sources in Kapp Linné and Adventdalen/Longyearbyen, with animals feeding at Kapp Linné more exposed to toxic elements originating from oceanic and atmospheric input, whereas animals feeding in Adventdalen/Longyearbyen could be more exposed to toxic elements originating from human activity.

Halbach et.al. (2017) conducted a study where they investigated the concentration of different metals in the surface soils in Svalbard to examine the source of pollution. They collected soil in Adventdalen and Ny-Ålesund in August 2014 and 2015. Their results show that the investigated trace elements can be divided according to their sources. Al, As, Cr, Cu, Fe, Ni and S most likely reflect the influence of the underlying mineral soils and bedrock. Cd, Pb and Zn might reflect a deposition from long-range atmospheric transport, and Hg and S might result from atmospheric deposition of marine aerosols (Halbach et.al., 2017). In the current study, Al, Cr, Cu and Fe were significantly higher in Adventdalen and Longyearbyen than at Kapp Linné, which could be explained by both human activity and underlying geology. Cd and Pb were significantly higher in Kapp Linné, which can thus be explained by long-range transport, and is in accordance with the findings of Halbach et.al (2017).

The Svalbard reindeer have developed different strategies for surviving the harsh environment on Svalbard, including migrating as little as possible. They are considered a sedentary species that may have genetic differences at different locations (Pacyna et.al., 2018). This could thus also explain some of the differences between the populations at Kapp Linné and in Adventdalen in relation to uptake and excretion, and should be investigated further.

#### **4.1.3 Rare earth elements**

The PCA plot (Figure 3.1) also shows that the REEs clustered together, most likely because they occur together in the same ore deposits (Obhođaš et.al., 2018). Figure 3.1 also indicates that the rare earth elements were higher in the samples from Adventdalen. This was tested and proved (Appendix C, Figure (C.1), and further indicating that they occur together in nature as well. The rare earth elements were investigated in order to see if they did indeed cluster together and will not be discussed further.

## **4.2 Element concentration within Kapp Linné**

The PCA loading and score plots (Figure 3.4 and 3.5) that divide Adventdalen and Kapp Linné into sub-areas indicated different element composition between faeces collected in the north, west and south of Kapp Linné. Se was shown to be significantly higher in the samples from the north, while Hg was significantly lower in the south (Figure 3.6). No difference was seen in Hg between Adventdalen and Longyearbyen compared to Kapp Linné, while Se was significantly higher in Kapp Linné (Figure 3.3). Figure 3.8 therefore shows that Se was only higher in Kapp Linné because of the samples from the north. No difference was shown between Adventdalen, south of Kapp Linné and west of Kapp Linné.

Kapp Linné is a coastal area with the ocean on one side and steep mountains on the other, and the samples were collected from reindeer between the ocean and the mountains. The area is not vast, but has different geology nevertheless. The northern part of Kapp Linné consists of mainly tilloid rock and limestone, while the western part consists of mainly marble and phyllite and the southern part consists of phyllite and sandstone (Appendix A, Figure A.1). These differences could contribute to different element consumption and excretion among the different herds of reindeer. As previously discussed, the sandstone, which is present in the south part of Kapp Linné, is fine-grained and have been shown to filter out contaminants from the surface.

Although the small sample size for the different areas within Kapp Linné makes it difficult to conclude, there seems to be an indication of different element composition and concentration in the north and the south on Kapp Linné that should be investigated further.

## **4.3 Element concentration in reindeer feeding close to contaminated sources**

### **4.3.1 Influence of the road on element concentration**

Coal is transported in trucks driving on the road in Longyearbyen and through Adventdalen, and the effect on the animals feeding close to the road was investigated. The elements investigated were As, Hg and Cd, which are all found in coal, and Al due to its high concentration in the soil.

This study hypothesized that reindeer feeding closer to the road would be more exposed to toxic non-essential elements. This does not, however, seem to be the case. None of the elements investigated were significantly correlated with distance to the road (Figure 3.7). All of the elements have a peaking point at around 5 km from the road, indicating higher levels of As, Hg,

Cd and Al there. However, most of the samples were collected between 0 and 5 km from the road (Figure 3.7). This makes it difficult to conclude on the effect of the dust from the road on surrounding environment. The levels of Al in the faeces would most likely originate from the soil and not reflect pollution from the road. Pb is more likely to originate from road pollution. When tested, Pb had almost the exact same pattern as Al, As and Hg (Appendix C, Figure C.2). The results could therefore also indicate that some of the metals are highly correlated, with more or less the exact same pattern across the distance.

#### **4.3.2 Influence of the coal power plant on element concentration**

Figure 3.8 shows that Cd was significantly correlated with distance to the coal power plant, with less Cd close to the power plant and more further away. None of the other elements (As, Hg and Al) were significantly correlated and all of them had an increasing trendline suggesting more of the toxic elements further away from the power plant. This could either be explained by no effect of the coal power plant on reindeer faeces or that the smoke travel far away and more likely effect the reindeer feeding further away. This makes it difficult to say for certain that concentrations of toxic elements found in Adventdalen or Longyearbyen were related to the pipe or came from other sources of pollution, such as the road or the soil. In either case, further studies should be conducted to test if the coal power plant affect the environment in Longyearbyen and Adventdalen.

It does seem to be a geological correlation between the elements, with two peaks, at 5 km and at 15 km, for As, Hg, Cd and Al. This indicates that the elements are correlated to each other and found in the same place. The two peaks are also a result of sampling, with all of the animals sampled at 5 km and 15 km. There was therefore no clear gradient of sampling from the pipe, which would have helped get a better understanding of pollution in the surrounding area of the pipe.

#### **4.3.3 Influence of the ocean on element concentration**

None of the essential elements tested (Ca, Mg, Fe and Cu) had a significant correlation with distance to the ocean (Figure 3.9). The concentration of essential elements in reindeer faeces most likely reflects the excess levels of elements their body does not need, making it difficult to conclude on the source of the essential elements in the faeces.

Regarding the non-essential elements, some correlation could be seen in As and Cd, but not for the other elements. Only Cd had a significant correlation with distance to the ocean ( $p=0.043$ ). Figure 3.9 shows that Al is the only element that has an increasing trendline, indicating higher

Al concentration the further the animals fed from the ocean. Since Al is high in soil and not in the marine environment, higher concentration in animals further away from the ocean would make sense. The non-essential element that was the highest in concentration in all reindeer faeces was Al. Al was significantly higher in Adventdalen and Longyearbyen compared to in Kapp Linné, indicating differences in soil composition. The high levels of Al in the samples might be due to the high levels of Al in soil, where it is the fourth most common element (Frankowski, 2016).

Both As and Cd have been shown to be high in the marine environment (Duncan et.al., 2015, Xu et.al., 2013), giving reason to believe that reindeer feeding close to the ocean would be exposed to higher concentrations of these two elements. As and Cd showed a trend of decreasing concentrations in faeces collected further away from the ocean, indicating As and Cd originate from the marine environment (Figure 3.10). As has been shown to have high concentrations in drinking water and sea food. According to Neff (1996) the total natural emission of As to the atmosphere is 1.5 times higher than emission from human activity (45000 metric tons/year compared to 28000 metric tons/year) and the total As concentration in clean ocean and coastal water is 1-3 µg/L (0.003 µg/g) in the US. Therefore, the major sources of As to surface waters of the oceans are riverine inputs and upwelling of deep ocean water rich in As (Neff, 1996). Cd is an element that has been widely studied in seawater. Shi et.al. (2016) showed in their studies that bivalves accumulated significantly higher Cd in more acidic ocean, indicating changes in speciation, toxicity and rates of redox processes in metals in seawater with changes in ocean pH (Shi et.al., 2016). Cd also has the ability to accumulate in plants, as was shown in Japan in 1960, when Cd released by a Zn mine in the upper reaches of the Jinzu River contaminated rice paddies and fields and caused the itai-itai disease. Itai-itai disease caused renal dysfunction, malabsorption, anemia, osteoporosis and osteomalacia (Nordberg, 2003). Both As and Cd in the marine environment could therefore accumulate in kelp or other marine plants and be taken up by reindeers and cause toxicity.

## **4.4 Associated investigations**

### **4.4.1 Comparative studies**

Several studies have investigated element concentrations in soil and moss at Svalbard (Beitveit, 2016, Halbach et.al., 2017). Beitveit (2016) conducted a study where element concentrations in moss samples from Adventdalen were investigated, while Halbach et.al (2017) conducted a study where element concentrations in surface soil from Adventdalen were investigated. These

concentrations can be used to compare with reindeer faeces to examine if some of the concentrations in the faeces may originate from the soil or moss. The concentrations of Al, As, Cd, Cr, Pb, Hg, Zn and Cu are presented in table 4.1.

**Table 4.1 Mean concentrations ( $\mu\text{g/g}$ ) of aluminium (Al), arsenic (As), cadmium (Cd), lead (Pb), mercury (Hg) and zinc (Zn) in samples from reindeer *Rangifer tarandus platyrchus* faeces (n=62), moss (n=18) and surface soil (n=57) from Adventdalen, Svalbard. Moss data were received from Beitveit (2016), while soil data were received from Halbach et.al. (2017).**

	<b>Sample type</b>	<b>N</b>	<b>Mean (<math>\mu\text{g/g}</math>)</b>	<b>Median (<math>\mu\text{g/g}</math>)</b>	<b>Minimum (<math>\mu\text{g/g}</math>)</b>	<b>Maximum (<math>\mu\text{g/g}</math>)</b>
<b>Al</b>	<b>Faeces</b>	62	9658	6554	320	28561
	<b>Moss</b>	18	6883	5101	2458	20266
	<b>Soil</b>	58	14400	13.6	1500	36500
<b>As</b>	<b>Faeces</b>	62	3.13	2.69	0.13	11.86
	<b>Moss</b>	18	2.71	2.21	1.01	6.51
	<b>Soil</b>	58	3.47	2.96	0.93	9.99
<b>Cd</b>	<b>Faeces</b>	62	0.30	0.036	0.03	1.19
	<b>Moss</b>	18	0.25	0.21	0.10	0.55
	<b>Soil</b>	58	0.44	0.38	0.13	1.00
<b>Hg</b>	<b>Faeces</b>	62	0.07	0.07	0.02	0.14
	<b>Moss</b>	18	0.04	0.04	0.03	0.05
	<b>Soil</b>	58	0.11	0.12	0.04	0.03
<b>Pb</b>	<b>Faeces</b>	62	3.60	3.89	0.17	9.21
	<b>Moss</b>	18	2.73	2.06	1.09	7.38
	<b>Soil</b>	58	11.90	10.70	5.60	25.1
<b>Zn</b>	<b>Faeces</b>	62	82.72	69.66	39.19	290.80
	<b>Moss</b>	18	41.77	33.84	19.33	109.21
	<b>Soil</b>	58	66.00	64.00	16.00	106.00

Table 4.1 shows that Al, As, Cd, Pb, Hg and Zn all can be found in soil and moss samples from Svalbard. All of the elements were similar in concentration, with some higher concentration in soil for Al, As, Cd, Pb and Hg. The similarities of element concentrations in moss and faeces can indicate that some of the elements taken up and/or excreted by reindeer originate from the

moss. It could also indicate that there is an equilibrium of the elements due to the constant eating of moss and other plants that can accumulate toxic elements. These findings could provide further knowledge about the element composition in the Arctic environment and sources of pollution for herbivores at Svalbard. It also shows that toxic elements in the environment are present in the soil and taken up in plants such as moss. Faeces samples can therefore be a good indicator of the elements in moss of the contamination load in the environment. Both soil and moss therefore seem to be possible sources of element contamination for Svalbard reindeer in Longyearbyen, Adventdalen and Kapp Linnè.

#### **4.4.2 Further investigations**

The Arctic is experiencing a change in the environment including higher temperatures, more extreme events and altered precipitation patterns (Hansen et.al., 2011). A warmer climate can result in less permafrost and less sea ice. With thawing of the permafrost, carbon and other possible toxic substances, such as toxic non-essential elements, could be released into the atmosphere and provoke a positive feedback to climate warming and pollution in the Arctic (Knoblauch et.al., 2013). Recent studies (Sun et.al., 2017, Schuster et.al., 2018) suggest that the Arctic permafrost holds large amounts of Hg that could be released into the atmosphere if the permafrost melts further. Permafrost is soil at or below 0° C for at least two consecutive years. It consists of an active layer of surface soil that thaws in the summer time and can accumulate and lock the Hg and other toxic elements in the permafrost once it freezes again (Schuster et.al., 2018). This indicates that the large areas of permafrost regions may become an important source of global Hg emission as a result of ongoing widespread permafrost degradation (Sun et.al., 2017).

Guerreiro et.al. (2014) conducted a study where they measured in air quality of As, Hg, Pb and Cd in Europe. All of the elements were shown to decrease in Europe in the period 2002 - 2011, with As being reduced by 3%, Hg by 26%, Pb by 20% and Cd by 27% (Guerreiro et.al., 2014). Although some toxic elements are released at a lower rate in the world, the Arctic is still an accumulation region that receives toxic elements from long-range transport. Toxic non-essential elements released into the atmosphere can be deposited on terrestrial or water surfaces in the Arctic and subsequently build-up in soils or sediments, causing toxicity to both plants and animals. Further studies are hence required to help monitor toxic elements in the Arctic environment to better understand sources of contamination.

## 5. Conclusion

A clear difference in element concentrations of both essential and non-essential elements were detected in the investigated areas. Concerning the essential elements, this study shows significantly higher concentration of Cu, Fe, K and Zn in faeces from Adventdalen and Longyearbyen, and significantly higher concentration of Ca, Mg and S in faeces from Kapp Linné. Concerning non-essential toxic elements, this study shows significantly higher levels of Cr and Al in faeces from Adventdalen and Longyearbyen, and significantly higher levels of Cd, Pb, Se and Tl in Kapp Linné.

These differences could be explained by either different geology or different sources of anthropogenic contamination. Adventdalen and Longyearbyen consist mostly of sandstone, while Kapp Linné has high geological diversity with some limestone. High concentration of Fe is found in sandstone, while high concentration of Ca is found in limestone, which is in accordance with the finding of the present study. In addition, sandstone has been shown to filter out contaminants from surface soil, which could be a possible explanation of higher concentrations of Cd, Pb, Se and Tl in Kapp Linné, as well as filter out Hg in the south part of Kapp Linné.

Differences in element concentrations of the toxic non-essential elements could be due to different sources of contamination. Adventdalen and Longyearbyen are areas with have high human activity and were expected to have higher concentrations of toxic elements compared to Kapp Linné. Nevertheless, toxic elements were found in higher concentrations in Kapp Linné, indicating higher contamination originating from long-range transport than from human activity. The results showed significant correlations between snow depth and levels of Pb and Hg. This, together with the fact that there is more precipitation in Kapp Linné compared to Adventdalen and Longyearbyen, could further indicate input from anthropogenic sources through long-range transport in Kapp Linné.

No significant correlation was shown in animals feeding close to the road in Adventdalen, while Cd was higher in faeces collected further away from the coal power plant in Longyearbyen and higher in faeces collected closer to the ocean near Kapp Linné. Based on these findings, it cannot be concluded that animals feeding closer to the road or the coal power plant are more contaminated by toxic non-essential elements than animals feeding further away. However, samples collected in Kapp Linné had higher levels of Cd and As, indicating higher contamination from oceanic input.

For further studies, samples should be sampled in a more widespread range to establish if the road, the coal power plant or oceanic input are contributing to contamination in the Arctic environment.

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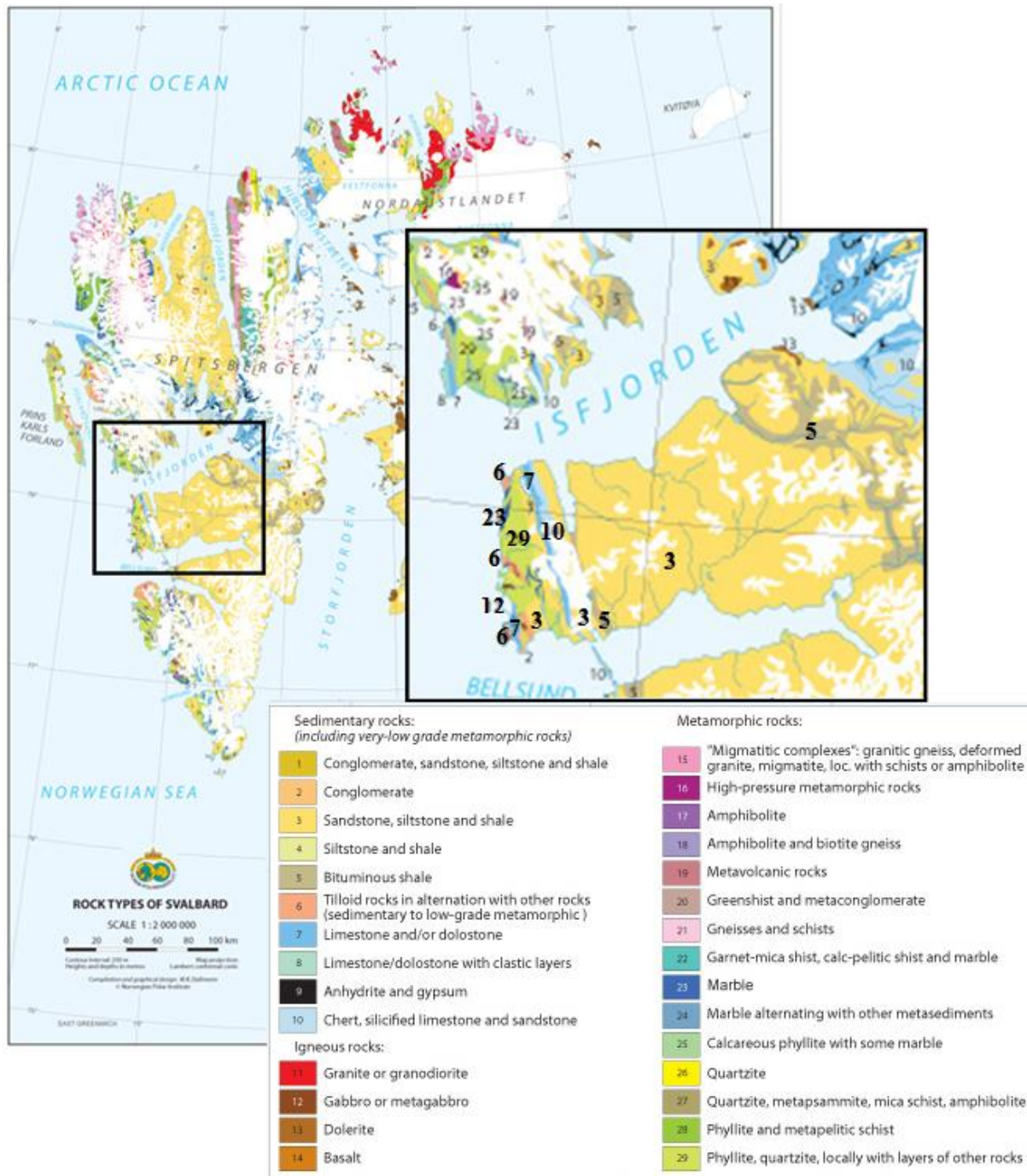
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## **Appendices**

## Appendix A: Geological Map



**Figure A.1 Geological map of rock types on Svalbard. Longyearbyen, Adventdalen and Kapp Linné are included in the square, which has been made larger for viewing purposes. Sedimentary, igneous and metamorphic rocks have been divided further into rock types and the map has been colored accordingly (Dallmann, Norwegian Polar Institute, 2015).**



## Appendix B: Principal Component Analysis

Table B.1 Percentage contribution of the principal components in the PCA of all samples of Svalbard reindeer faeces. The principal components are significant if  $Q2 > 0$ . R2X represent the fraction of the total variation of X and Y that can be predicted by a principal component.

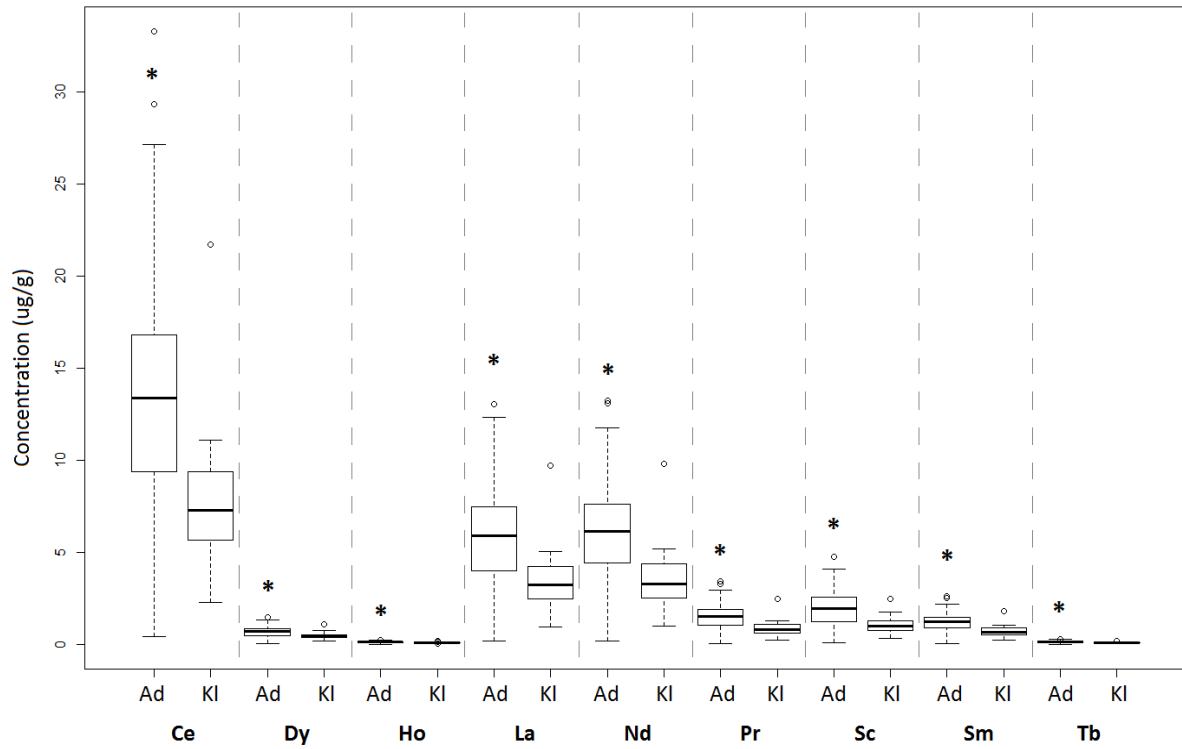
Component	R2X	Eigenvalue	Q2	Limit	Q2(cum)	Significance	Iterations
1	0.546	33.3	0.523	0.0263	0.523	R1	10
2	0.126	7.66	0.152	0.0266	0.595	R1	30
3	0.0956	5.83	0.204	0.027	0.678	R1	24
4	0.0589	3.59	0.162	0.0274	0.73	R1	19
5	0.0323	1.97	0.0257	0.0278	0.737	R2	137



59	Advntd	M	A	2017	5.5	12000	5700	14600	0.24	0.11	0.05	0.09	3.62	12120.12	3.48	17.77	2675.74	1280.57	1715.58	6283.84	21037.32	9753.70	8.65	52.26	683.42
60	Advntd	F	A	2017	3.5	12000	5860	14700	0.24	0.31	0.06	0.03	1.32	2826.00	0.92	3.72	2156.19	1804.55	2017.66	3217.54	15568.21	2059.87	5.33	89.80	2989.44
61	Advntd	F	A	2017	2.0	11600	6160	14600	0.37	0.21	0.06	0.11	4.34	13139.36	3.57	18.64	2813.14	1813.06	1884.75	6449.27	21411.62	9531.79	9.54	68.67	461.52
62	Advntd	F	A	2017	2.0	12000	6500	14700	0.32	0.50	0.08	0.07	3.36	8569.35	2.62	11.53	2234.92	1314.92	1625.77	4766.78	20658.80	6251.49	7.85	79.78	729.74
63	Advntd	C	C	2017	2.0	12000	6500	14700	0.46	0.43	0.08	0.13	5.09	15149.06	6.01	19.38	3085.64	1522.06	2059.56	6674.72	17683.15	11052.08	10.76	87.22	1614.00
64	KL-W	M	A	2014		12			0.39	1.72	0.08	0.06	5.18	4278.59	11.21	5.51	7836.73	2978.39	9493.43	4793.26	36471.99	3887.61	9.52	87.13	5631.25
65	KL-W	M	A	2014		12			0.52	0.41	0.12	0.09	6.98	6039.73	1.71	7.22	3568.66	1338.51	2207.91	5518.08	31611.84	5204.78	5.84	49.05	1116.31
66	KL-W	M	A	2014		12			0.48	0.69	0.11	0.08	7.76	5902.25	3.42	7.24	5357.02	1515.77	3921.39	6257.64	29853.30	5272.62	8.83	78.26	1200.52
67	KL-N	F	A	2015		937			0.50	0.25	0.06	0.04	2.51	2809.50	0.64	3.44	3226.25	1830.89	2651.98	4005.11	25848.91	2864.18	4.46	36.26	1277.03
68	KL-N	F	C	2015		937			0.46	0.23	0.06	0.03	2.17	2702.70	0.55	3.02	3552.93	1729.54	2666.12	4309.21	28686.52	2337.05	4.22	31.41	1857.08
69	KL-N	M	A	2015		1380			0.44	0.64	0.06	0.04	2.88	3373.74	11.82	4.38	8623.81	2394.87	5669.72	3567.26	31195.88	3217.05	5.16	62.08	591.58
70	KL-N	F	A	2015		1380			0.64	0.35	0.08	0.05	3.39	4263.64	0.74	5.30	4013.59	1945.29	2670.23	3688.51	25709.52	4583.89	5.02	49.61	3428.10
71	KL-S	F	A	2015		1980			0.58	0.22	0.06	0.06	3.91	5791.86	1.87	6.04	3688.69	1522.28	2410.24	4660.81	34500.47	5949.01	5.10	40.58	1297.51
72	KL-S	M	A	2015		477			0.49	0.58	0.05	0.05	4.35	4542.72	8.87	5.90	7063.67	1937.76	4826.66	5454.46	32048.43	4148.60	5.93	60.65	605.33
73	KL-S	M	A	2015		34			0.33	0.40	0.04	0.05	2.65	3870.14	5.44	4.26	5224.13	1397.93	3908.23	4764.92	26896.78	3093.54	3.99	36.16	678.61
74	KL-W	M	A	2017		356			0.72	0.75	0.14	0.03	5.27	2604.84	0.71	2.87	2937.02	1586.43	1583.07	3260.78	26420.69	2606.16	5.38	75.80	616.88
75	KL-N	F	A	2017	1.5	328			0.88	0.88	0.15	0.04	7.82	2340.75	0.65	3.37	2207.71	1810.81	1955.76	3064.15	30834.96	2150.98	5.17	81.67	529.50
76	KL-N	M	A	2017	7.5	284			0.88	0.59	0.13	0.05	7.68	3793.72	1.06	5.06	2503.56	1926.04	1754.82	2509.69	41109.25	3671.00	4.91	63.64	257.20
77	KL-N	C	C	2017	7.5	236			0.67	0.65	0.10	0.04	5.19	3224.68	0.80	4.64	2682.30	1749.32	1999.23	3740.19	31270.50	2886.47	4.92	74.55	1035.74
78	KL-N	M	A	2017	2.5	565			0.85	0.93	0.15	0.04	8.01	2603.22	0.73	3.69	2323.85	1718.86	1885.90	2775.87	28817.34	2385.08	5.43	86.10	535.41
79	KL-N	F	A	2017	2.0	310			0.89	0.76	0.14	0.04	6.54	2973.43	0.74	4.32	2976.48	2094.43	2035.49	2540.51	36279.38	2486.72	5.48	76.25	338.89
80	KL-N	M	A	2017	30.0	1650			1.01	0.70	0.16	0.07	11.09	4702.26	1.33	6.38	2246.06	1575.63	1366.75	2384.29	32288.09	4285.08	5.32	63.63	201.61
81	KL-N	F	A	2017	10.0	1650			1.06	0.83	0.18	0.07	13.92	4233.78	1.04	5.96	2320.19	1937.42	1498.78	2703.28	32543.71	3789.95	5.37	73.00	249.66
82	KL-N	F	A	2017	4.0	1550			0.77	0.46	0.11	0.07	7.28	5080.43	1.24	6.34	2764.74	1451.85	1783.06	3817.67	30745.36	4472.36	5.64	52.67	364.22
83	KL-N	M	A	2017	3.5	1800			0.28	0.26	0.04	0.05	4.43	5034.77	0.71	7.81	5292.79	1376.76	1778.23	4378.18	41522.70	4063.35	6.96	71.41	464.07
84	KL-N	C	C	2017	4.0	2410			0.73	0.43	0.10	0.06	6.73	4496.52	0.72	5.90	2627.61	1136.55	1426.27	4150.14	23449.17	3931.72	5.01	53.13	736.23
85	KL-N	C	C	2017	16.0	2500			0.81	0.49	0.11	0.07	7.20	5423.97	0.87	6.71	3083.72	1663.60	1945.43	4905.12	26037.80	3938.67	6.42	56.65	537.83
86	KL-W	M	A	2017	4.5	362			0.43	0.30	0.09	0.04	5.40	3923.29	0.91	4.14	2176.52	934.45	1089.82	2034.65	19225.43	3975.41	4.35	39.75	246.30
87	KL-W	M	A	2017	4.0	431			0.53	0.38	0.11	0.05	7.14	4629.11	1.56	5.58	3036.01	1270.21	1410.15	2782.48	25227.22	5282.46	5.94	49.97	361.59
88	KL-W	F	A	2017	8.0	759			0.30	0.38	0.06	0.02	2.33	1545.47	0.32	1.58	1392.29	620.88	678.76	1272.25	11736.56	1525.17	2.50	33.30	228.26
89	KL-S	M	A	2017	1.3	2200			0.74	0.36	0.09	0.11	8.26	9520.51	2.71	12.61	5362.90	1110.52	1366.32	6009.69	24824.23	9124.15	8.87	55.32	663.30
90	KL-S	M	A	2017		1050			0.31	0.48	0.03	0.06	3.13	5142.97	5.92	6.44	7295.81	1246.51	4369.00	3997.88	35174.77	4771.67	5.21	48.14	658.87
91	KL-S	F	A	2017		974			0.16	0.20	0.02	0.04	1.71	3412.22	4.97	3.83	6009.28	809.57	3082.09	1939.87	21420.93	3164.07	3.14	28.89	186.91
92	KL-S	M	A	2017		192			0.33	0.92	0.06	0.06	2.21	4840.36	13.42	5.44	16506.88	2167.15	10692.63	2739.11	57031.71	4900.21	6.71	75.71	465.45
93	KL-S	M	A	2017		190			0.15	0.38	0.02	0.04	1.62	3879.86	5.87	4.48	7793.97	938.40	3853.20	2204.15	24701.16	3824.03	4.18	39.68	223.09
94	KL-S	F	A	2017	0.0	112			0.21	0.69	0.04	0.03	1.51	2483.60	11.61	3.15	12076.72	2132.49	9241.63	1372.29	39842.02	2915.73	4.38	59.69	276.30
95	KL-W	F	C	2017	0.0	330			0.61	0.41	0.10	0.06	7.54	4742.87	0.97	5.57	2751.10	1080.10	1411.98	3387.55	25983.44	4886.76	13.34	54.81	667.55
96	LYB	M	A	2013	0.0	791	695	865	0.11	0.10	0.07	0.01	0.48	994.24	0.42	1.74	4463.42	7457.51	3045.10	5238.22	20371.24	1095.11	13.22	290.80	160.99
97	LYB	M	A	2014	0.0	787	687	870	0.06	0.03	0.02	0.00	0.17	320.59	0.13	0.60	1226.40	1374.75	897.87	980.62	4894.61	319.73	3.29	72.46	88.27
98	LYB	M	A	2014	0.0	24	26	1840	0.37	0.26	0.07	0.08	3.26	11495.62	3.57	17.11	4880.94	3120.31	3318.42	4098.31	13940.44	10103.25	17.40	137.59	611.41







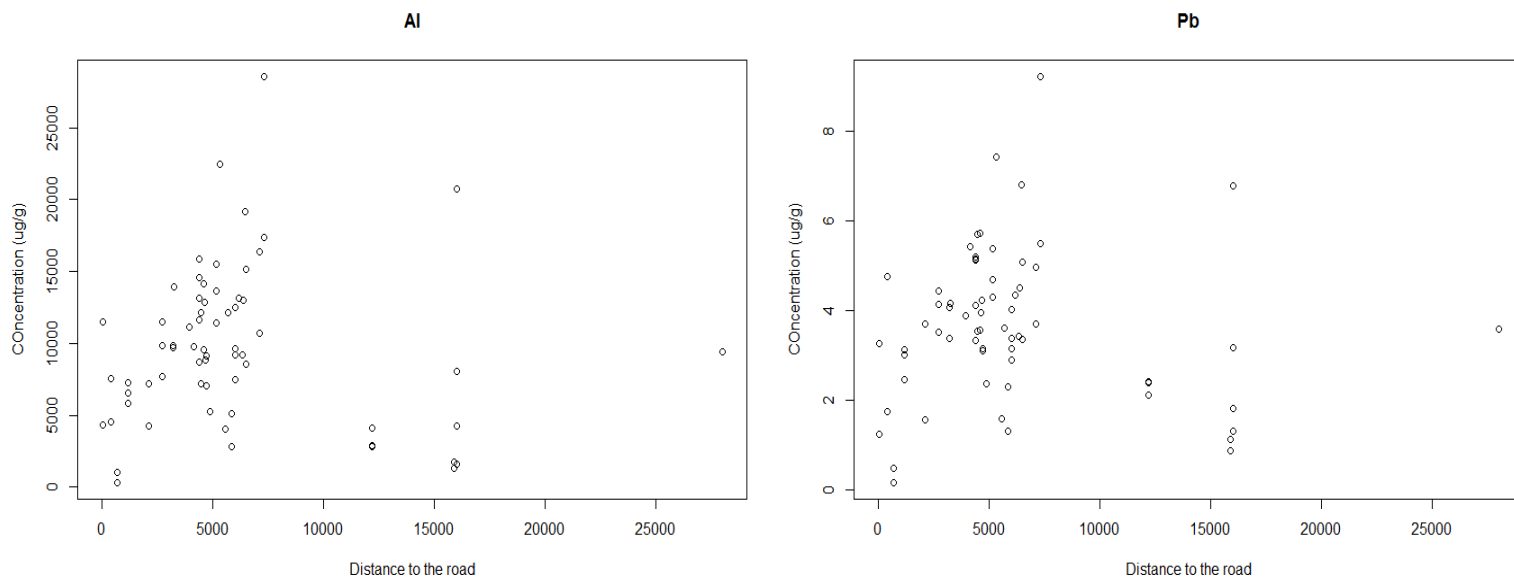
**Figure C.1 Rare earth elements in Adventdalen and Longyearbyen (n=65) compared to Kapp Linné (n=32). Asterisks (\*) denote significant difference between the two locations, with level of significance set at  $p < 0.05$ . Cerium (Ce), dysprosium (Dy), holmium (Ho), lanthanum (La), neodymium (Nd), praseodymium (Pr), scandium (Sc), samarium (Sm) and terbium (Tb) were significantly higher in Adventdalen and Longyearbyen compared to Kapp Linné**

**Table C.3 Mean concentrations ( $\mu\text{g/g}$ ) of essential elements in Adventdalen and Longyearbyen (Ad, n=65) and Kapp Linne (Kl, n=32) in Svalbard reindeer faeces from 2013 to 2017.**

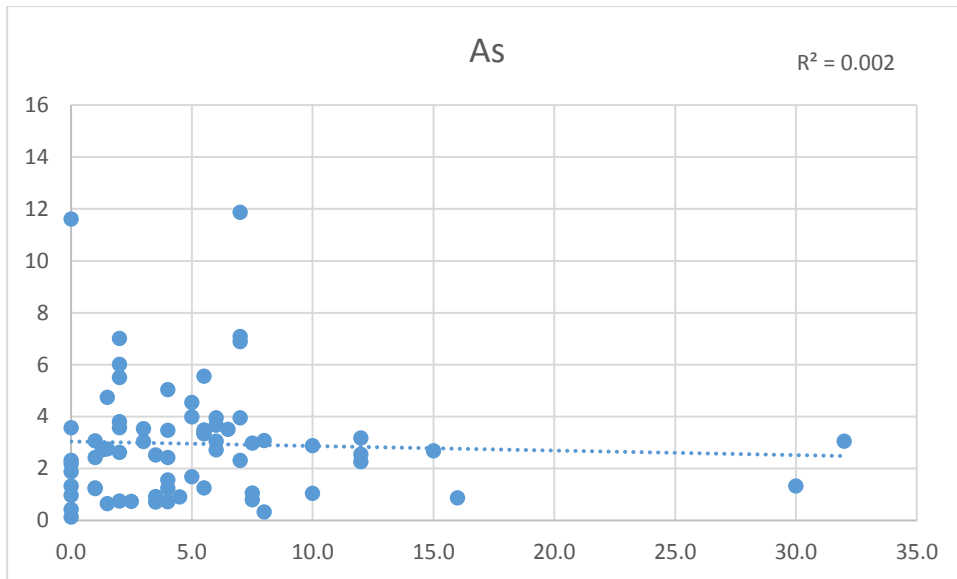
	Ca	Cu	Fe	Mg	S	Zn	Al
Ad	15952.6807	8.53509423	7498.40628	2651.63632	1811.3945	82.7224079	9658.253
Kl	30290.9389	5.70883608	4703.84322	4703.84322	3082.32127	57.6549366	4193.984

**Table C.4 Mean concentrations ( $\mu\text{g/g}$ ) of toxic non-essential elements in Adventdalen and Longyearbyen (Ad, n=65) and Kapp Linne (Kl, n=32) in Svalbard reindeer faeces from 2013 to 2017.**

	As	Hg	Tl	Pb	Cd	Se	Cr
Ad	3.13284029	0.075354	0.079175	3.652569	0.34862953	0.34249803	13.19684
Kl	3.28620097	0.088209	0.053795	5.430327	0.55384053	0.56739528	5.255697



**Figure C.2 Comparison between aluminium (Al) and lead (Pb) in Svalbard reindeer faeces close to the road in Adventdalen.**



**Figure C.3 Aluminium (Al) concentration ( $\mu\text{g/g}$ ) in Svalbard reindeer faeces sampled at different snow depths (cm). The  $R^2$  of the trendline is shown to be 0.002 and  $p=0.44$ .**