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Possible Anthropogenic Sources of Toxic Elements in Snow Bunting Nestlings (*Plectrophenax nivalis*) in Adventdalen, Svalbard.

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Abstract

The Arctic environment is exposed to natural and anthropogenic contamination as a result of local, regional and global pollution. Contaminants accumulating in the Arctic may influence both biotic and abiotic factors. Previous research has quantified elevated concentrations of toxic elements in a confined area between Endalen and Todalen in Adventdalen, Svalbard. The overall aim of the present study was to determine the sources of contamination in this particular area by systematically examining element concentrations in soil, insects, spiders, and feathers of snow bunting nestlings (*Plectrophenax nivalis*). Assumptions regarding elevated concentrations of toxic elements focuses on coal industry related elements (As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, Yb), and elements related to operations at a wartime weather station (Bansø) (Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, Zr).

The study area was divided into a grid pattern consisting of 25 cells. Soil, insect, spider, and nestling feather samples from each cell were collected in June and July 2016, and element composition was quantified using HR-ICP-MS. The principal component analysis (PCA) indicates that most contamination in the investigated area is related to local sources, probably of anthropogenic origin. The results show that cells located close to a coal mine adit (Mine 5) and coal piles have elevated concentrations of coal related elements in soil and feathers, but not in invertebrates. The cell located closest to the coal mine adit is not statistically significant ($p>0.05$). This indicates that coal contamination not necessarily affects the toxic element concentrations in invertebrates.

The soil samples from cell D3 ($p<0.001$), where the Bansø weather station is located, do not show elevated concentrations. Cell D3 show high concentrations in invertebrates, which indicates the importance of this station. Because of lacking data for feathers in the Bansø cell, no conclusion can be drawn regarding the station's impact on birds. However, there are quantified elevated concentrations of Bansø related elements in feathers in Bansø's adjacent cells, which indicate an impact also for birds. The present thesis have not examined parental foraging strategies in snow buntings, and can thus not exclude other possible sources for elevated concentrations, neither for coal related nor Bansø related elements. Based on the results, it can be assumed that the remains of Bansø weather station affects local biota more than coal contamination do. In addition, the results indicate presence of biomagnification of toxic elements, and further studies are therefore recommended.

Sammendrag

Arktis er utsatt for naturlig og antropogen forurensing som en følge av lokale, regionale og globale utslipp. Forurensende stoffer som akkumulerer i Arktis kan påvirke både biotiske og abiotiske faktorer. Tidligere forskning har kvantifisert forhøyede konsentrasjoner av toksiske elementer i et avgrenset område mellom Endalen og Todalen i Adventdalen, Svalbard. Denne avhandlingens overordnede mål var å fastslå kilden til forurensing i dette bestemte området, ved å systematisk undersøke konsentrasjoner av grunnstoffer i jord, insekter, edderkopper og fjær av snøspurvunger (*Plectrophenax nivalis*). Antagelsene knyttet til forhøyede konsentrasjoner av toksiske elementer fokuserer på stoffer knyttet til kullindustrien (As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, Yb) og stoffer knyttet til drift av en værstasjon fra andre verdenskrig (Bansø) (Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, Zr).

Studieområdet ble delt inn i et rutenettmønster bestående av 25 celler. Jord-, insekt-, edderkopp- og fjærprøver ble samlet inn fra hver celle i juni og juli 2016, og grunnstoffsammensetningen i disse ble bestemt ved hjelp av HR-ICP-MS-analyse. Hovedkomponentanalysen (PCA) indikerer at store deler av forurensingen er relatert til lokale kilder, trolig av antropogen opprinnelse. Resultatene viser at celler lokalisert nært kullgruveutgangen (Gruve 5) og kullhauger har forhøyede konsentrasjoner av kullrelaterte elementer i jord og fjær, men ikke invertebrater. Cellen lokalisert nærmest kullgruveutgangen er ikke statistisk signifikant ($p>0.05$). Dette indikerer at kullforurensing ikke nødvendigvis påvirker sammensetningen av toksiske elementer i invertebrater.

Jordprøvene fra celle D3 ($p<0.001$), hvor Bansøstasjonen er lokalisert, viser ikke forhøyede konsentrasjoner. Celle D3 har høye konsentrasjoner i invertebrater, noe som indikerer stasjonens viktighet. På grunn av manglende data for fjær rundt Bansø, kan det ikke trekkes noen konklusjon med tanke på stasjonens innvirkning på fugler. Det er allikevel kvantifisert forhøyede konsentrasjoner av Bansø-relaterte elementer i fjær i celler som grenser mot Bansø, noe som indikerer en påvirkning også for fugler. Denne avhandlingen har ikke undersøkt hvordan snøspurven søker etter og samler inn mat, og andre mulige kilder til disse forhøyede konsentrasjonene kan derfor ikke ekskluderes, hverken for kullrelaterte eller Bansø-relaterte elementer. Basert på resultatene kan det antas at rester etter Bansø værstasjon påvirker lokal biota mer enn kullforurensing gjør. I tillegg indikerer resultatene at biomagnifisering finner sted, og ytterlige studier er derfor anbefalt.

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Table of contents

Abstract ..... **I**

Sammendrag ..... **III**

Acknowledgements ..... **V**

Table of contents ..... **VII**

Abbreviations ..... **IX**

1 Background and theory **1**

1.1 The Svalbard archipelago **2**

1.2 Pollution on Svalbard **3**

1.3 Toxic elements **4**

1.4 Coal **5**

1.4.1 Coal industry on Svalbard 6

1.5 Background and aim of the thesis **7**

2 Study area, materials and methods **9**

2.1 Study area **9**

2.2 Study objects **12**

2.2.1 Soil 12

2.2.2 Insects and spiders 13

2.2.3 Snow bunting feathers 13

2.3 Sampling methods **14**

2.4 Element analyses **15**

2.4.1 UltraClave and HR-ICP-MS 15

2.5 Statistical analyses **16**

2.5.1 Principal Component Analysis 16

3 Results **19**

3.1 Principal Component Analysis **19**

3.2 Spatial distribution of elements in study area **21**

4 Discussion **25**

4.1 Principal Component Analysis **25**

4.1.1 Soil 25

4.1.2 Insects and spiders 25

4.1.3 Snow bunting feathers 26
4.1.4 Summary of PCA results 27
4.2 Contamination related to coal mining 28
4.2.1 Soil 28
4.2.2 Insects and spiders 29
4.2.3 Snow bunting feathers 29
4.2.4 Summary of contamination related to coal mining 30
4.3 Contamination related to Bansø weather station 30
4.3.1 Soil 31
4.3.2 Insects and spiders 31
4.3.3 Snow bunting feathers 32
4.3.4 Summary of contamination related to Bansø weather station 33
4.4 Comparison of results 33
4.5 Limitations 34
4.5.1 Soil 34
4.5.2 Insects and spiders 34
4.5.3 Snow bunting feathers 35
4.6 Recommendations for future studies 37
5 Concluding remarks 39
6 References 41
A. Appendices 51
A.1 Data processing 53
A.2 Principal Component Analysis 57
A.3 Heatmaps 67
A.4 Bar charts 76
A.5 Detailed results 79
A.6 Linear mixed-effect model 91

Abbreviations

Al	-	Aluminium
AMD	-	Acid mine drainage
As	-	Arsenic
B	-	Boron
Ba	-	Barium
Be	-	Beryllium
Ca	-	Calcium
Cd	-	Cadmium
Ce	-	Cerium
CEC	-	Cation exchange capacity
Co	-	Cobalt
Cr	-	Chromium
Cu	-	Copper
DF	-	Detection frequency
dw	-	Dry weight
Fe	-	Iron
H	-	Hydrogen
Hg	-	Mercury
K	-	Potassium
La	-	Lanthanum
Li	-	Lithium
lme	-	Linear mixed-effect model
LOD	-	Limit of detection
Mn	-	Manganese
Mg	-	Magnesium
Mo	-	Molybdenum
MYA	-	Million years ago
N	-	Nitrogen
Na	-	Sodium
Ni	-	Nickel
NTNU	-	Norwegian University of Science and Technology
O	-	Oxygen
P	-	Phosphorus
Pb	-	Lead
PCA	-	Principal component analysis
PAHs	-	Polycyclic aromatic hydrocarbons
PBDEs	-	Polybrominated diphenyl ethers
PCBs	-	Polychlorinated biphenyls
POPs	-	Persistent organic pollutants
Rb	-	Rubidium
S	-	Sulphur
Sb	-	Antimony
Se	-	Selenium
Si	-	Silicon
Sm	-	Samarium
SNSK	-	Store Norske Spitsbergen Kulkompani AS
Th	-	Thorium
Ti	-	Titanium
Tl	-	Thallium
U	-	Uranium
V	-	Vanadium
Y	-	Yttrium
Yb	-	Ytterbium
Zn	-	Zinc
Zr	-	Zirconium

1 Background and theory

The Arctic region is considered a rather pristine area, as it is populated by few people and has low industrial activity. The ecosystem of the Arctic is considered to be sensitive and fragile (Jiang et al., 2011), and can be threatened by both local and long-transported contamination from continental and regional sources. Studies on concentrations of contaminants in the Arctic biota are important for monitoring the quality of the environment. Pollutants introduced to ecosystems may influence competition between organisms before affecting tissues or individuals, and can thus be used as early indicators of environmental impacts of toxic substances (Ellenberg, 1988, Jaworowski, 1989). Hence, the Arctic region is considered predictive of changes in the global environment (Kozak et al., 2013).

Toxic and harmful chemicals and elements accumulating in the Arctic originate from four different sources: local natural pollution, local anthropogenic pollution, remote natural pollution and remote anthropogenic pollution (Jaworowski, 1989). Natural pollution includes weathering of rocks, volcanic eruptions, and forest fires. Industrial processes such as fossil fuel combustion and mining, as well as contamination from human settlements, are examples of anthropogenic pollution. Arctic air pollution is due to long-range transport of pollutants, including the Arctic haze phenomenon; when contaminated air covers large parts of the northern hemisphere during winter (Kozak et al., 2013, Wadleigh, 1996). This is partly a result of anthropogenic emissions at lower latitudes (Akeredolu et al., 1994, Barrie, 1986, Iversen, 1989). Industrial activities in the mid-latitudes emits different pollutants that are transported through the atmosphere and deposited in polar regions (McConnel and Edwards, 2008).

There are several different types of pollutants accumulating in the Arctic. Persistent organic pollutants (POPs) are organic and toxic chemicals. They are resistant to biodegradation and persistent in the environment, and have the potential of bioaccumulation and biomagnification (Zeng, 2015). POPs include polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and various pesticides. Organic pollutants can be derived from both anthropogenic and natural processes (Yunker et al., 2002). Whereas PAHs can be derived from both sources, other organic compounds such as PCBs, PBDEs and various organochlorine pesticides must be synthetically made (Rhind, 2009). Neither organic pollutants nor metal pollutants are easily degraded, and can be accumulated in animal tissues (Rhind, 2008, Rhind, 2009, Zeng, 2015).

Such accumulation may lead to biological responses even with low concentrations of the pollutant, as some can act additively (Kortenkamp, 2007). Accumulation of the contaminant depends partly on the stage of development of the animal, its diet composition, potential for excretion and metabolization, and the pollutant's chemical class and properties (Rhind, 2008).

Certain trace elements are required in small, but critical, quantities for ensuring normal growth of plants and animals. These micronutrients include cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), magnesium (Mg), molybden (Mo), selenium (Se) and zinc (Zn) (Alloway, 2013, Tokar et al., 2013), and are termed essential elements. Essential elements may become toxic if present in excess, as they overwhelm biological systems and bind to unwanted sites (Tokar et al., 2013). Non-essential elements, such as aluminium (Al), arsenic (As), beryllium (Be), cadmium (Cd), chromium (Cr), mercury (Hg), nickel (Ni), and lead (Pb) can mimic essential elements, and thus disrupt key cellular functions (Tokar et al., 2013). Hence, they can lead to toxicity, and are therefore termed non-essential or toxic elements. Certain elements, both essential and non-essential, have the potential to accumulate in tissues and transfer to higher trophic levels. It is important to map concentrations of various toxic trace elements as contaminants entering food webs can influence major parts of the ecosystem (Gulinska et al., 2003). The terrestrial food webs in the Arctic are relatively simple, and introduced contaminants can contribute to potential disturbances of the ecological balance (Kozak et al., 2013). Invertebrates are in close contact with contaminants in the soil compartment (Heikens et al., 2001). As many mammals and birds eat invertebrates, it is important to investigate the toxic element concentration in soil, primary, and secondary consumers.

Svalbard is a region exposed to a greater atmospheric transport of impurities from distant natural and man-made sources than some other Arctic areas (Jaworowski, 1989, Maenhaut and Cornille, 1989). By comparing concentrations of various elements in ice from the pre-industrial period and in contemporary ice, a measure of man's contribution to atmospheric pollution can be achieved.

1.1 The Svalbard archipelago

Svalbard is an archipelago located north of mainland Europe, between 74° and 81° north latitude and 10° and 35° east longitude. The largest island is Spitsbergen, followed by Nordaustlandet and Edgeøya. Longyearbyen, located on Spitsbergen, is Svalbard's main settlement. Other establishments include the Russian mining community of Barentsburg, the research station at Ny-Ålesund and some mining outposts like Svea and the now-abandoned

Pyramiden. The permanent settlements relates mainly to coal industry, research and, in recent time, tourism (Meld. St. 32 (2015-2016)). Svalbard is a free economic and demilitarized zone, managed by the governor of Svalbard, and the Svalbard Treaty of 1920 recognizes Norwegian sovereignty (Svalbardtraktaten, 1920).

The archipelago has an Arctic climate, and several species have adapted to the extreme conditions. The marine food web is extensive, but there are only two indigenous terrestrial mammals: the Arctic fox (*Alopex lagopus*), and the Svalbard reindeer (*Rangifer tarandus platyrhynchus*) (Hisdal, 1998). The bird life is very rich in terms of number of individuals (Kovacs and Lydersen, 2006), especially during summer. The number of breeding species, however, is limited, and the Svalbard rock ptarmigan (*Lagopus muta hyperborea*) is the only terrestrial bird residing in the archipelago throughout the year (Fuglei, 2006). The snow bunting (*Plectrophenax nivalis*) is the only passerine bird regularly breeding in the Arctic (Strøm, 2006). There are recorded more than 250 insect species on Svalbard (Coulson, No year). In addition there are approximately 60 springtail species, various spiders, mites and worms.

The snow buntings in Longyearbyen and Adventdalen, Spitsbergen, have been studied for more than 20 years. The studies have been conducted with regards to hormones and territorial behaviour during breeding (Romero et al., 1998), song complexity related to male quality (Espmark, 1999, Hofstad et al., 2002), the effects of male mating behaviour and food provisioning on breeding success (Hoset et al., 2009), extra-pair paternity in relation to climate (Hoset et al., 2014), and breeding success related to climatic parameters and climate in a 15-year long project (Fossøy et al., 2015). In addition, newer studies have investigated pollution from local anthropogenic sources and its potential effects on the snow bunting. Kristoffersen (2012) examined levels of organohalogenated compounds in eggs, Kvernland (2017) investigated the effects of per- and polyfluorinated alkylated substances in snow bunting eggs, Nordnes (2016) examined whether age and sex affects accumulation of metals and other elements, and Beitveit (2016) investigated whether contamination in the snow bunting reflects emissions from the local coal industry.

1.2 Pollution on Svalbard

Being located halfway between the North Cape of Norway and the North Pole, the archipelago is far from major sources of atmospheric pollution. However, previous studies have shown that remote human activity may change the chemical composition of the polar environment (Arctic Monitoring and Assessment Programme, 2005, Kozak et al., 2013,

Maenhaut and Cornille, 1989, Rose et al., 2004, Simões and Zagorodnov, 2001). Svalbard is particularly exposed to long-range transport due to specific conditions of atmospheric circulation (Barrie, 1986, Jaworowski, 1989).

In addition to long-range transport, local sources of pollution might have contributed to increased amounts of contaminants in the environment (Headley, 1996). Local sources on Svalbard are mainly human settlements, traffic, and coal mining industry, where the latter cover both mine waste (García-Giménez and Jiménez-Ballesta, 2017, Søndergaard et al., 2007), the actual mining operations (Avango et al., 2014) and emissions from coal power plants in Longyearbyen and Barentsburg (Kaczmarska et al., 2016). It is however important to note that this contribution might be localized and difficult to define (Jaworowski, 1989, Rose et al., 2004). Mining processes include coal dust escaping during extraction, storing, loading, and transporting of coal (Simões and Zagorodnov, 2001). During coal extraction, surrounding rocks with low coal content are removed. This material is deposited in piles above ground as mine waste rocks, and contains iron sulphide minerals. These minerals oxidise when in contact with air and water, and produce sulphuric acid (H_2SO_4). This reaction reduces pH in the surrounding soil, and trace elements such as Ni, Zn, and Pb from the sulphide oxidation, and Al and Mn from weathering processes, are leached. The acidic metal-enriched runoff is referred to as acid mine drainage (AMD) (Askaer et al., 2008). Headley (1996) examined the heavy metal concentrations in peat profiles on Svalbard and found increased levels in certain areas, which could not be attributed to pollution from the local mines alone. It is therefore necessary to consider also regional and global sources of contamination on Svalbard.

1.3 Toxic elements

Elements such as metals are naturally occurring pollutant toxicants in all ecosystems, and are virtually ubiquitous in human environments (Arctic Monitoring and Assessment Programme, 2005, Kim et al., 2013, Tokar et al., 2013). Metals are persistent in the human environment, leading to protracted exposures (Gall et al., 2015, Tokar et al., 2013). The use of toxic elements in industry and other anthropogenic activities contributes to increased concentrations in the biosphere at both local, regional and global scales (Arctic Monitoring and Assessment Programme, 2005). Moreover, anthropogenic use may alter the chemical form of different elements, and thereby affect their availability for biotic uptake and toxic potential (Arctic Monitoring and Assessment Programme, 2005, Tokar et al., 2013). The concentration of toxic elements also depends on local geology.

Heavy metals are often introduced through anthropogenic processing such as waste

disposals, agricultural processes, sewage sludging, metal mining, mine tailings and fossil fuel combustion (García-Giménez and Jiménez-Ballesta, 2017, Wuana and Okieimen, 2011). Fossil fuel combustion causes atmospheric releases of elements bound to particles or in complexes (Mandal and Sengupta, 2006, Sabbioni et al., 1984, Wuana and Okieimen, 2011). Tracer measurements have indicated that the main source of certain metals, for instance Hg, in the Arctic after 1860 is a result of fossil fuel combustion and other industrial processes in Asia, Europe, and North America (Kozak et al., 2013, Maenhaut and Cornille, 1989, McConnel and Edwards, 2008), partly because of the ash output resulting from coal combustion. Coal contain traces of almost all elements, and an increase in coal consumption will lead to an increased release of trace metals into the environment (Sabbioni et al., 1984). Coal ash is a potential contaminant of soils, surface water and groundwater, and can be transported by wind and water over vast distances. Ash particles are rather small with a large surface area, and have a tendency to absorb trace elements transferred from coal to waste products during combustion (Güleç et al., 2001, Mandal and Sengupta, 2006).

Several toxic elements, e.g. Hg (Arctic Monitoring and Assessment Programme, 2005, Mahbub et al., 2017), can accumulate and biomagnify in the food chain, and even low levels of atmospheric deposition are therefore a threat to the Arctic ecosystems (Bard, 1999, McConnel and Edwards, 2008). Biomagnification and long-lasting exposure are important with respect to effects on different trophic levels, as bioavailable elements may be biotransferred or bioaccumulated from one trophic level to the next.

1.4 Coal

Coal is an organic, carbonaceous product, formed from decayed plant material altered by various chemical and physical forces (Speight, 1994). Coal formation occurs through several processes. First, biomass turns into peat. Being covered by a layer of overburden, time, pressure and high temperatures the peat will convert to lignite. In time it will develop into subbituminous coal, bituminous coal, and finally anthracite (Higman and van der Burgt, 2008). These four major types of coal have increasing rank on the metamorphic scale (Speight, 1994). In addition to having high carbon content, coal consists of hydrogen (H), oxygen (O), nitrogen (N) and sulphur (S), and several inorganic materials. Higher ranked coal have higher carbon content and energy content, and lower O, H, and moisture content (Higman and van der Burgt, 2008). The carbon content of the world's coal varies from 75-95 %. A variety of minerals and trace elements, incorporating metals, have also been reported to be present in coals (Speight, 1994). The exact composition of coal depends on the geological

time of its formation. The most favourable conditions for coal formation were during the Carboniferous period (approximately 360-290 million years ago (MYA)). Coal from this era is more mature than coal from later periods since it has been exposed to higher pressure and temperatures.

The coal deposits on Svalbard comprises several coals, ranging from Upper Devonian era (383-372 MYA), through the Carboniferous (360-290 MYA), Permian (299-252 MYA), Triassic (252-201 MYA), Jurassic (201-145 MYA), and Cretaceous eras (145-66 MYA), to the early Tertiary era (66-2.6 MYA) (Michelsen and Khorasani, 1991). The coal-bearing strata on Svalbard are mainly of Carboniferous, Triassic, Cretaceous, and Tertiary age (Hoel, 1925, Michelsen and Khorasani, 1991). The coals in Adventdalen, Barentsburg and Svea mines originates mainly from the Jurassic, Cretaceous, and Tertiary eras, and mining in these areas have been based on a Tertiary coal layer (Hjelle, 1993, Hoel, 1925). Longyearbyen coals are in the range of bituminous coals (Cmiel and Fabianska, 2004). Coal from Pyramiden is from the Carboniferous and Permian eras (Hoel, 1925).

1.4.1 Coal industry on Svalbard

The commercial interest for Svalbard has largely been centred around the coal industry. In the late 1800s and early 1900s Svalbard's coal deposits attracted attention from various stakeholders (Hisdal, 1998). At the time, Europe had a huge need of coal for industrial processes, steamboats and an expanding railway network. Many nations established mining companies with prospects of exploiting Svalbard's coal resources. Among these were two Americans, Frederic Ayer and John M. Longyear, who founded the Arctic Coal Company (Hisdal, 1998). In 1916, their mining operations were taken over by Norwegian businessmen, and the company was renamed to Store Norske Spitsbergen Kulkompani AS (SNSK) (Hisdal, 1998, Reymert, 2013).

Today (2017), most of the mines are closed. SNSK is operating in "Gruve 7" (Mine 7) outside Longyearbyen, which produces up to 150 000 tons of coal each year (SNSK, Undated-a). The Svea mines were operating until the end of 2016, but are now provisionally terminated (SNSK, Undated-b). The Russian company Trust Arktikugol have also operated several mines around Svalbard. Today they only operate one mine in Barentsburg, with an annual coal production plan set at 120 000 tons of coal per year (Arktikugol, Undated). Even though most of the mines now are abandoned, many mining constructions and mine adits, particularly surrounding Longyearbyen, are still present as protected cultural remains. This includes the cableway and cable trestles in Adventdalen, which transported coal from the

mines to the harbour until the end of 1987 (Avango et al., 2014, Piepjohn et al., 2012, Reymert, 2013). These remains might have an environmental impact, especially because of coal and coal residues lying beneath the cableways, and must be taken into account in research regarding local Arctic pollution.

In December 2015 a new treatment plant at Longyearbyen Powerplant (Longyear Energiverk) was put into operation (Andersen, 2015). The purification process consists of three steps for reduction of SO_2, NO_x and other particulate matter, before it is emitted through the chimneys (Røkenes, 2017).

1.5 Background and aim of the thesis

Svalbard's Arctic environment is exposed to pollution both from long-range atmospheric transport and local industry, which may lead to increased amounts of toxic elements in some areas. This thesis is a part of the objective of the "Vinterspurv" project (RiS-ID 2272, http://www.researchinsvalbard.no/project/5938) at the Norwegian University of Science and Technology (NTNU) to understand the bioaccumulation of toxic elements in snow buntings. Previous research conducted in the "Vinterspurv" project has shown that the concentrations of several trace elements are particularly high in the confined area between Endalen and Todalen in Adventdalen (see figure 1 in Section 2.1) (Beitveit, 2016). Values for Al, As, Cd, Hg, Mn, Mo, Ni, Pb, Se, Zn, lanthanum (La), antimony (Sb), samarium (Sm), uranium (U), vanadium (V), yttrium (Y), and ytterbium (Yb) in this area were higher in feathers of snow buntings, insects, moss or soil samples, compared to concentrations elsewhere in Adventdalen between Longyearbyen and the abandoned "Gruve 6" (Mine 6) (Beitveit, 2016).

This thesis will investigate concentrations of all the above-mentioned elements, in addition to Be, Cr, Co, Fe, S, barium (Ba), boron (B), cerium (Ce), lithium (Li), rubidium (Rb), thorium (Th), thallium (Tl), and zirconium (Zr), in this confined area in Adventdalen. The assumed reasons for the elevated levels in this area include coal mining, and use and remains of a German wartime weather station (Bansø). Some of the mentioned elements are related to coal industry or are components in coal (As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, Yb). Others are related to components of glass or building materials (Al, Ba, Ce, Cr, La, Li, Rb, Sb, Se, Tl, V, Zn, Zr), or have been used in relevant measuring instruments (as Hg in thermometers and barometers) (Järup, 2003, Kabata-Pendias, 2011, Mahbub et al., 2017, Mogren and Trumble, 2010, Ottesen et al., 2010). These latter elements can be considered a measure of the anthropogenic impact from meteorological activity at Bansø. By examining concentrations in soil, insects, spiders, and feathers from snow bunting

nestlings (which are fed invertebrates), this thesis aims to identify the main sources for elevated concentrations of toxic elements in this confined area, and whether the elements bioaccumulate and biomagnify. As previously mentioned, the snow buntings in Adventdalen might be exposed, through diet or external exposure, to toxic and non-toxic elements originating from coal mining or other anthropogenic processes. This was examined by establishing a grid pattern consisting of 25 cells in a confined area, and collect samples of soil, insects, spiders, and feathers from snow bunting nestlings in every cell. I hypothesized that the concentrations of elements related to coal and mining would be elevated close to the coal mine adit of "Gruve 5" (Mine 5) in the hillside (see Figure 1), partly because of AMD and coal residues stored in piles. The same assumption applies to cells located close to the estuary of the Endal river, as the river passes remains of the main entrance of Mine 5 located in Endalen. In addition, I hypothesized that the concentrations of elements related to the Bansø weather station would be elevated around the station's former location, as remains of the building materials and vast amounts of glass are found in the surrounding soil.

2 Study area, materials and methods

2.1 Study area

In 2015, Beitveit (2016) examined trace element concentrations along a gradient from Longyearbyen to Mine 6. The results showed elevated concentrations in a cabin area between Endalen and Todalen in Adventdalen, approximately 7 kilometres from Longyearbyen. The fieldwork for this thesis was conducted in this specific area. The study area extended over one square kilometre, and was divided into a grid with 25 cells (Figure 1). This area contains several possible sources of contamination. It encompasses cottages and waste piles consisting of old snow mobiles and construction materials. A cableway with cable trestles used for transporting coal from the mines to Longyearbyen stretches throughout the area. In the hillside there is an adit of the abandoned Mine 5 (Figures 1 and 2), with surrounding coal residues and building materials. The Endal river and the Todal river flows in the outskirts of the study area, and both rivers were in spring flood during sampling. According to Norwegian Polar Institute (TopoSvalbard), the main entrance of Mine 5 is found where the Endal river originates. The gravel road from Longyearbyen runs parallel with the grid (with a maximum distance from the grid border of 70 m), with a protruding dirt road into the cottage area. Enclosed within the area is also the historical German weather station Bansø built and used under World War II (WWII) (Capelotti, 2000) with much surrounding glass waste (Albrigtsen, 2013, Hassel, 1994, Reymert, 2013, Riksantikvaren, 2013) (Figures 1 and 3).

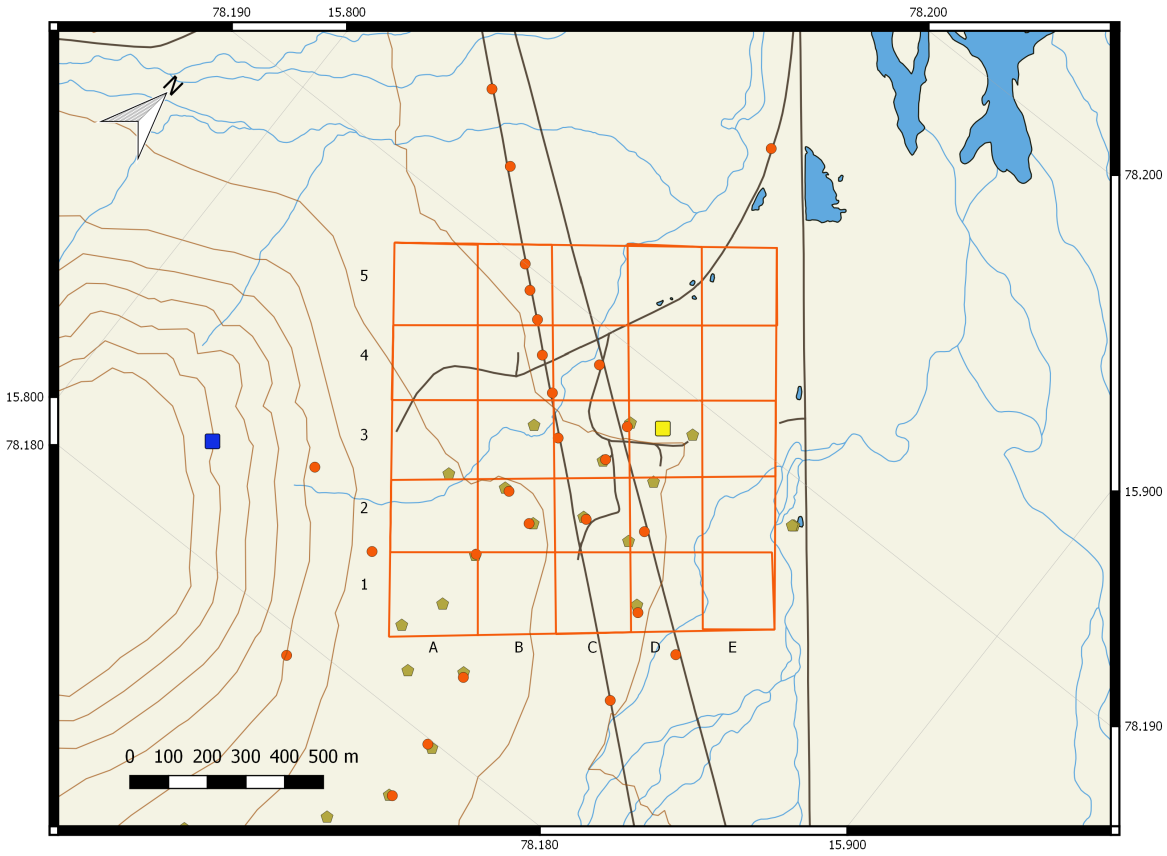

Figure 1. The study area between Endalen and Todalen in Adventdalen. The research area, of 1 km × 1 km is divided into 25 equal cells of 200 m × 200 m each. Columns are named with letters (A-E), and rows with numbers (1-5). The roads in the area are marked as black lines, passing parallel to the grid (right side, maximum distance from the grid border is 70 m), and protruding into the area. All blue markings represent rivers, streams and lakes. The spring flood coincided with the field work. The two long black lines crossing the area marks the cable trestles (left) and electric masts (right). Red dots mark snow bunting nests, which are located both in and outside the grid (maximum distance from the grid border is 400 m), and brown stars mark cabins. The coal mine adit of Mine 5 is marked with a blue square in the hillside left of the grid, and the Bansø weather station is marked with a yellow square in cell D3. Scale is shown at the bottom left of the figure.

Figure 2. The coal mine adit in the hillside is circled, and coal residues are apparent below it. Cable trestles and electric masts stretch throughout the area, and are visible along the centre of the photograph. Cabins are apparent to the left. (Photo: Sørhus, H. S. 2016)

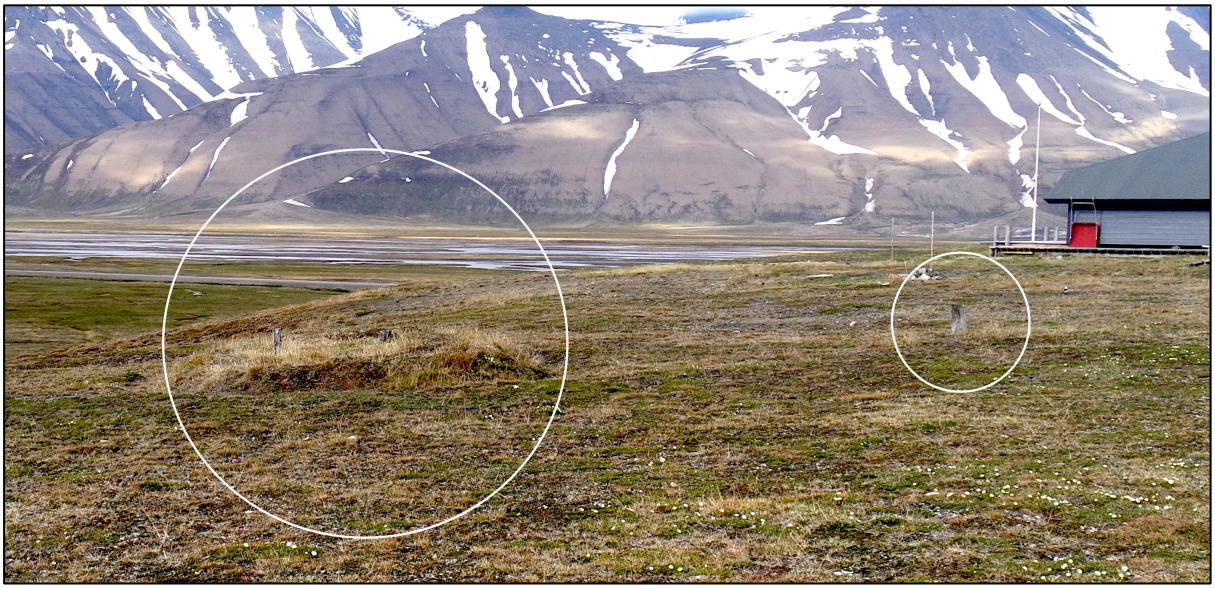

Figure 3. The remains of Bansø weather station is apparent as a round mound with remains of two poles, surrounded by a foot trench (left circle). In addition, there is a broken pole to the right of the trench (right circle). There are vast amounts of glass in the soil. (Photo: Sørhus, H. S. 2016)

2.2 Study objects

2.2.1 Soil

Soil is a complex substance with large variations in chemical and physical composition. It consists of varying components, like water, organic matter, clay, particles and biological entities. Elements in soil may be chelated with or contained in the organic matter or adsorbed on clays, oxide minerals or carbonates (White and Zasoski, 1999). Accumulation of elements in soil depends on the soil components, as well as pH and cation exchange capacity (CEC) (Alloway, 2013). Soil is a buffer controlling the transport of chemical elements and substances between atmosphere, hydrosphere and biota, while also being a geochemical sink for contaminants (Kabata-Pendias, 2011).

All soils contain more or less all heavy metal(loid)s, with varying concentrations (Alloway, 2013). The concentration depends on elements from minerals in the geological parent material on which the soil has developed (the lithogenic source) and possible anthropogenic contamination sources. 99 % of the earth's crust is comprised of ten elements: Al, Fe, Mg, O, calcium (Ca), phosphorus (P), potassium (K), silicon (Si), sodium (Na) and titanium (Ti) (Alloway, 2013). The remainder of the elements is called trace elements, and can become toxic at enhanced levels in the soil (Mandal and Sengupta, 2006). Soils are not considered polluted if the levels of contaminants do not affect biological processes (Kabata-Pendias, 2011). At contaminated sites the most commonly found heavy metals, in order of abundance, are Pb, Cr, As, Zn, Cd, Cu, and Hg (Wuana and Okieimen, 2011). How toxic a contaminant can become depends on its chemical species and bioavailability.

Soils can be contaminated in various ways, since both lithogenic source, local sources and long-range transport can contribute to soil contamination (Thomas, 1986). Gulinska et al. (2003) examined heavy metals in soils of central Spitsbergen, and found that local contamination was significant in soils close to the pollution source, and that the concentrations in Spitsbergen in general were lower than European legal norms. The metal concentrations were mostly related to the local bedrock geology, but elevated in areas where human impact was observed (Gulinska et al., 2003). Natural sources such as glacial erosion and bedrock weathering may also affect the soil composition (Gulinska et al., 2003, Kabata-Pendias, 2011, Kozak et al., 2016). In his report, Jaworowski (1989) states that natural contributions from local weathering processes are more important than long-range transport for soils on Svalbard.

Atmospheric deposition, including both long-range transport and local pollution,

affects mainly topsoil (Mandal and Sengupta, 2006, Sterckeman et al., 2000). However, contaminants can also leach and be transferred to neighbouring soil and affect several soil layers (Mandal and Sengupta, 2006). Heavy metals are more persistent in soil than in other biosphere compartments. They do not undergo microbial or chemical degradation, and will not be depleted (Mandal and Sengupta, 2006, Wuana and Okieimen, 2011).

2.2.2 Insects and spiders

Insects and spiders have been used as bioindicators in ecology for a long time (Azam et al., 2015, Goodyear and McNeill, 1999, Heikens et al., 2001, Jung et al., 2008, Thomé et al., 1987, Yang et al., 2016). Soil-dwelling invertebrates may ingest metals and other toxic elements contained in soil and plants, or absorb them through their exoskeletons (Gall et al., 2015). When accumulated, heavy metals can affect insects and spiders in the form of reducing reproduction, inhibiting growth and development, decreasing their immune response and cause changes in life history traits (Azam et al., 2015, Gall et al., 2015). Insects and spiders have different physiological and biochemical responses, and their exposure pathways varies (Yang et al., 2016). Some taxa are more likely to accumulate metals than others, depending on their exposure to metalliferous soils through habitat or diet, and physiological condition (Gall et al., 2015). Certain species have developed metal tolerance or various elimination and detoxification mechanisms, which may reduce transfer to higher trophic levels. Spiders are more resistant to metal uptake than insects (Migula et al., 2013, Yang et al., 2016), and therefore considered to be better bioindicators regarding toxic element pollution. Spiders are polyphagous, and have, because of their carnivorous nature and ability to store metals in their tissues, been recognized as potent biological indicators (Migula et al., 2013, Sanders, 2013, Wilczek et al., 2005).

2.2.3 Snow bunting feathers

The snow bunting is widely distributed in the high Arctic environments of Svalbard (Hoset et al., 2014). The bird nests in cavities between or under rocks and boulders, but also in buildings and other manmade voids (Bangjord et al., 1999, Cramp, 1994, Hindrum, 1994). Despite this apparent protection, nest predation by e.g. Arctic foxes, glaucous gulls (*Larus hyperboreus*), and Arctic skuas (*Stercorarius parasiticus*) (Hoset et al., 2009), as well as examination by eager researchers, occurs. The snow bunting are first level predators (Kozak et al., 2013). Adult birds feed mainly on seeds, but their nestlings are fed invertebrates during the breeding season (Cramp, 1994, Falconer et al., 2008). For comparisons of concentrations

in prey and predator, the present study examines concentrations in both invertebrates and feathers from snow bunting nestlings.

Birds can eliminate heavy metals and other pollutants by depositing them in their feathers (Burger and Gochfeld, 1991, Goede and Debruin, 1984). Internal tissues, blood, eggs, and feathers can all be used for monitoring contamination (Kim et al., 2013). The element concentrations in feathers are consistent with other body tissue levels, and the feathers can thus be used as an indicator tissue of metal exposure in birds (Goede and Debruin, 1984, Kim et al., 2013). Experimental use of nestling feathers are reasonable because they quickly regrow and are easy to store (Burger and Gochfeld, 1991). When conducting research, it is advantageous to sample feathers since this can be done while the bird is still alive.

When studying bioaccumulation in birds, using nestlings is a good analytical method. The element concentrations in nestling feathers are representative for contaminants accumulated from food brought to them by their parents, and are therefore a measure of the local concentrations of metals (Burger and Gochfeld, 1991). Since birds migrate, the heavy metal concentrations in adult birds are not limited to local contamination, but reflect accumulations from both past and present exposures. For nestlings, which are fed locally acquired invertebrates, bioaccumulation can be used as an indicator for the contamination in the area (Burger and Gochfeld, 1991). Adult snow buntings feed almost entirely on the ground in close vicinity to their nests (Cramp, 1994).

2.3 Sampling methods

The insects and spiders were sampled using pitfall traps in the centre of each cell (Figure 1), from June 28 to July 2, 2016. The pitfall traps consisted of a plastic cup containing approximately 50 mL of 70 % v/v ethanol solution, and were buried in the soil with the upper rim of the cup flush with the soil surface. The traps were left in the field for 48 hours, before the contents (insects and spiders) were collected in plastic tubes and frozen at -20 °C. The samples were stored without alcohol to minimize the diluting effect of alcohol on the element concentrations (Braun et al., 2009). The invertebrate sampling were conducted twice with a two days interval, and the two different samples were pooled for further analyses.

The soil samples were taken from the same positions as the pitfall traps, from June 28 to June 30, 2016. Topsoil (humus layer) was collected, as this is the most significant sink for trace elements and the assumed source of accumulation into plants (Kabata-Pendias, 2011, Mandal and Sengupta, 2006, Sterckeman et al., 2000). The soil samples were frozen at -20 °C within a few hours.

Feather samples were taken from the abdomen of snow bunting nestlings 8-9 days after hatching, approximately 3-5 days prior to fledging, during the period of June 18 to July 6, 2016. The nests were located in or just outside of the grid area, with a maximum distance of 400 m outside the grid border (Figure 1). In those cases where the nests were located outside of the grid, they were assigned the number of the nearest cell. Feathers from all nestlings in the same nest were pooled. Hence, the concentrations of heavy metals from feather samples represent the average concentration from one nest. In addition, feathers from one dead nestling was sampled and analysed separately. Its concentrations did not differ particularly from the living nestlings in the same nest, and these samples were therefore also pooled for statistical analyses. All feathers were stored in small, labelled paper envelopes.

To reduce the risk of contamination of samples, all sampling was conducted using non-metallic equipment, such as Teflon or plastic tweezers, plastic spoons and cups.

2.4 Element analyses

The feather samples were washed with acetone alternating with double deionized water (Milli-Q), and air-dried at room temperature for approximately 72 hours. They were then dried in a Termaks drying oven (series TS8000) on 50°C for 16 hours. The insects and spiders were classified and sorted into taxonomic groups (Cyclorrhapha, Acari, Nematocera, Collembola, Hymenoptera, and Araneae) (Table A.1, Appendix A.1). The samples were left to dry overnight at room temperature. The following day insects and spiders were dried in the same Termaks drying oven as feathers, on 60°C for 18 hours. After thawing the soil samples, they were dried on 50°C for approximately 22 hours in the same oven. To ensure that all soil samples were completely dry, they were dried again on 60°C for another 21 hours. The soil samples were homogenized using an agate mortar, and subsequently dry-sieved through 2 mm non-metallic polyester sieves (Gilson company inc., Ohio, USA, SV-126). All samples were stored in room temperature pending further processing. Placing clean filter papers on top of all samples prevented contamination during the drying processes. All laboratory equipment used in the element analysis was non-metallic, prewashed with nitric acid (HNO_3, 1M), and rinsed with Milli-Q water.

2.4.1 UltraClave and HR-ICP-MS

All samples (n_{total}=166, of which $n_{insects}$=89, $n_{spiders}$=23, $n_{feathers}$=29, and n_{soil}=25) were digested with nitric acid (HNO_3, 50 %, 2 mL, 4.5 mL or 9 mL) in a high-pressure microwave system (Milestone UltraClave, EMLS, Leutkirch, Germany) using 18 mL teflon vials. After digestion

the samples were diluted with Milli-Q water in a PFA flask. Amount depended on the type of sample: Feather, insect and spider samples were diluted to approximately 20 mL, while soil samples were diluted to approximately 108 mL. The diluted samples were transferred to 15 mL PP-vials. Final determination of 58 elements was carried out using a Thermo Finnigan model Element 2 instrument (Bremen, Germany) at the Department of Chemistry, NTNU (HR-ICP-MS). Blanks containing HNO_3 were analysed for feather, insect/spider and soil samples.

Limits of detection (LODs) were calculated for all analytical matrices (Table A.2, Appendix A.1) based on different weight classes (Table A.3, Appendix A.1). Elements where 50 % or more of the analysed concentrations of each class were below LOD, were excluded from the dataset used for statistics (Helsel, 2005). For elements not excluded, single values below LOD were replaced with a value of LOD multiplied with the detection frequency of the specific element (LOD×DF). The analytical procedure was verified with the use of certified GBW-0740 reference material.

2.5 Statistical analyses

Heatmaps and bar charts for all metals (Al, As, B, Ba, Be, Cd, Ce, Co, Cr, Fe, Hg, La, Li, Mn, Mo, Ni, Pb, Rb, S, Sb, Se, Sm, Th, Tl, U, V, Y, Yb, Zn, Zr) were created by using the software R (R Development Core Team, 2008) with the package ggplot2 (Wickham, 2009). For the creation of heatmaps, all concentrations below LOD were replaced with LOD×DF, and all values were standardized on a scale from 0 to 1. For heatmaps showing elements related to coal (As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, Yb) or Bansø weather station (Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, Zr), the packages dplyr (Wickham, 2011) and vegan (Oksanen et al., 2017) were used. A linear mixed-effect model (lmer) were created using R with the package lmerTest (Kuznetsova et al., 2016), to get p-values and determine statistical significance. For comparative purposes log-scale was used. This model aimed to demonstrate the relationship between element concentrations in the different cells in the grid, and examine whether these relationships were statistically significant compared to a reference cell (cell A1). Statistical significance was accepted at $p \leq 0.05$.

2.5.1 Principal Component Analysis

SIMCA (Version 13, Umetrics, Umeå, Sweden) was used to perform a principal component analysis (PCA) of all samples. PCA is a projection method that extracts and displays

systematic variation in a data matrix, making it easier to interpret (Dabiri et al., 2017, Eriksson et al., 2013). It represents a multivariate data table as a low-dimensional plane, which gives an overview of the data. This estimates the internal relations in the data set, and can uncover different trends and observations otherwise unnoticed (Eriksson et al., 2013, Kozak et al., 2016). A PCA computes several principal components, which represents the eigenvector with the highest eigenvalue. The first principal component, PC1, represents the direction in the data and contains the largest variation. PC2, orthogonal to PC1, represents the largest residual variation around PC1, and improves the approximation of the data (Eriksson et al., 2013, Kozak et al., 2016). A third principal component, PC3, will be orthogonal to both PC1 and PC2, and represents the direction of the highest residual variation (Kozak et al., 2016). By plotting the results, a PCA makes it possible to visualize the structure of the dataset in question, and distinguish groups and outliers (Eriksson et al., 2013, Kozak et al., 2016). Several PCA plots were created, both for comparing all sample materials, and for each separate material.

When interpreting a PCA, the significance of principal components is important. Significance of principal components depends on sample number and Q^2-value. Q^2 represents the fraction of the total variation of X or Y that can be predicted by a principal component (Eriksson et al., 2013). If the sample number exceeds 100, a principal component is significant when Q^2 is higher than 0. If the sample number is equal to or less than 100, Q^2 must be higher than 0.05 in order for the principal component to be significant (Eriksson et al., 2013).

3 Results

For this thesis, 25 soil samples were analysed: one from each cell. In addition, it was collected 89 insects samples from 25 cells. When several insect samples were analysed from the same cell their concentrations were averaged, resulting in one mean concentration for every investigated element per cell. The same was done for the 23 spider samples collected in 22 cells. Nestling feathers were collected from 101 individuals in 28 different nests, sampled from 14 cells, in addition to the aforementioned dead nestling. Concentrations in feathers for samples collected from the same cells were averaged in the same way as for insects and spiders, giving in total 14 mean concentrations reflecting respective cell concentrations of each element.

3.1 Principal Component Analysis

The PCA of all sample materials resulted in a model with three significant principal components explaining 85.05 % of the variation (PC1-3, $R^2X(cum)=0.85$, $Q^2(cum)=0.783$) (Table 1). PC1 explained 73.9 % of the variation, while PC2 explained 7.01 % and PC3 4.05 %. Results from PCA analysis for all sample materials with PC1 and PC2 are presented in Figures 4 and 5. These figures indicate which elements that are responsible for the clustering of the different analytical matrices, as well as how the elements correlate with each other. Variables located close to each other tend to be positively correlated, while variables located on opposite sides of the plot origin are negatively correlated (Eriksson et al., 2013). The score plot (Figure 4) shows a clear separation of soil, insects/spiders, and feather samples. The loading plot (Figure 5) reveals which elements that are related to the different matrices. Results for each separate sample material are found in Appendix A.2.

Table 1. Percentage contribution of the principal components in the PCA of all sample materials (soil, insects/spiders, and feathers). This model incorporates 166 samples, and the principal components are significant if $Q^2>0$ (Eriksson et al., 2013). Thus, the principal components 1-3 are significant. R^2X represents the fraction of the total variation of X or Y that can be predicted by a principal component.

Component	R^2X	Eigenvalue	Q^2	$Q^2(cum)$
1	0.739	37	0.731	0.731
2	0.0701	3.5	0.154	0.772
3	0.0405	2.02	0.047	0.783

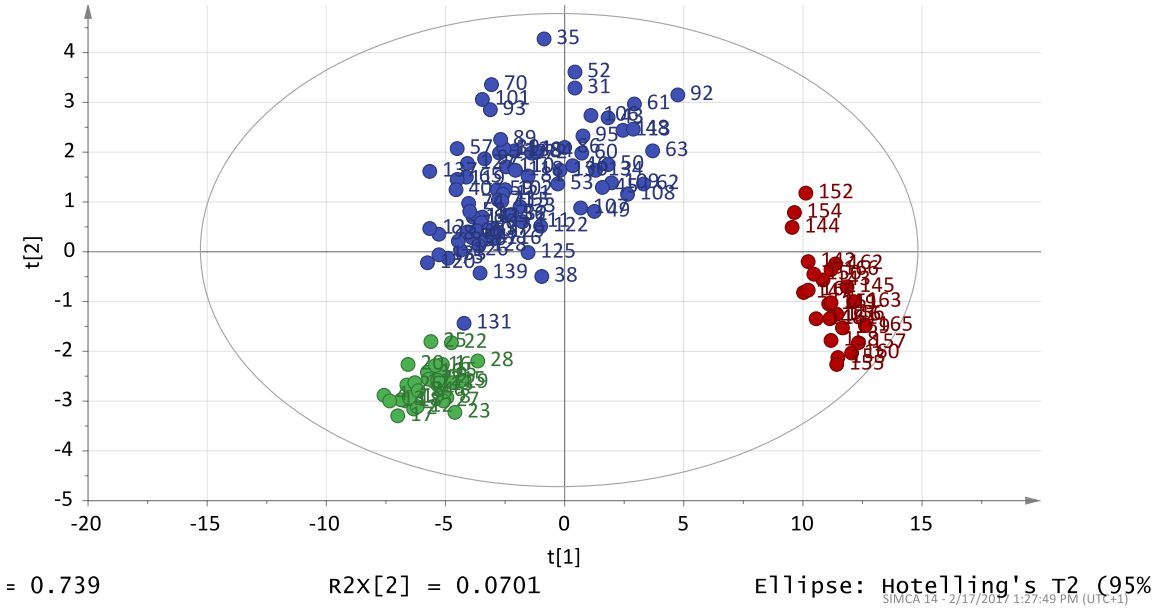

Figure 4. PCA score plot for all sample materials grouped into soil (red, n=25), insects/spiders (blue, n=78), and feathers (green, n=29). Numbers represent individual sample numbers. The figure is showing principal components 1 and 2, where PC1 explains 73.9 % and PC2 7.01 % of the variation. Numbers represent individual sample numbers. R2X=0.0701 show how the concentrations in feathers better reflect the concentrations in insects/spiders than in soil along the PC1 axis.

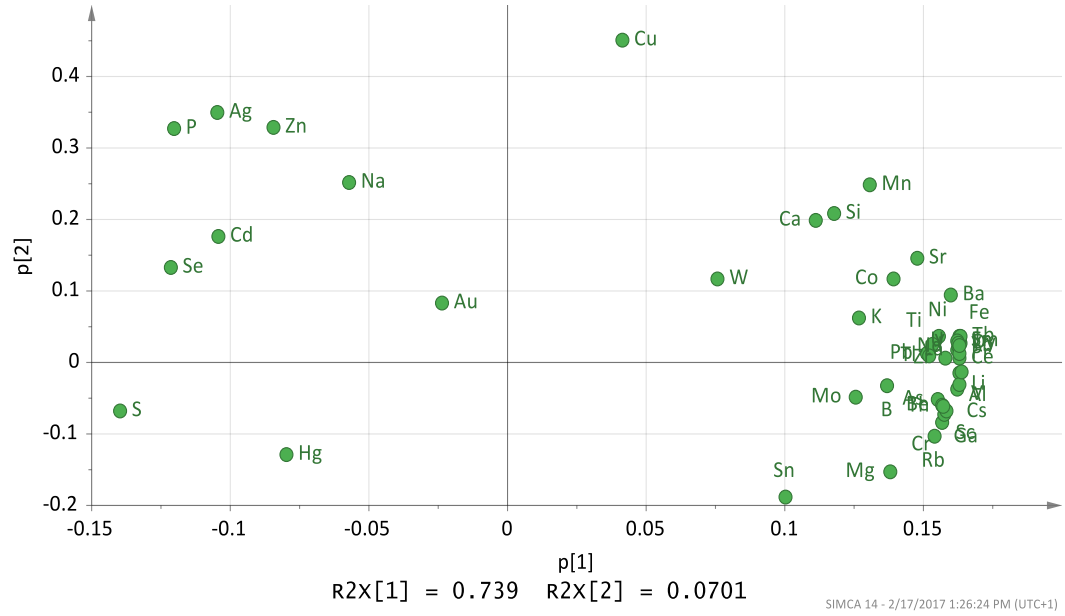

Figure 5. PCA loading plot for all sample materials. All elements where 50 % of the data was above LOD are presented. When comparing with Figure 4, this figure represents which elements that are present in the different analytical matrices. The figure is showing principal components 1 and 2, where PC1 explains 73.9 % and PC2 7.01 % of the variation. $R^2X[1]=0.739$ show the fraction of the variation of the variables explained by the model.

Figures 4 and 5 show how the element compositions in the different analytical matrices are clearly differing. The score and loading plot explains that soil samples contain most of Al, As, B, Ba, Be, Ce, Co, Cr, Fe, Li, Mn, Mo, Ni, Pb, Rb, Sb, Th, Tl, V, and Zr; insect and spider samples contain most of Cd, Se and Zn; whereas feather samples contain most of Hg and S.

The separate PCA results for soil, insects/spiders, and feathers are all shown in Appendix A.2. The PCA for soil resulted in a model with three principal components, explaining 59.8 %, 9.98 %, and 7.75 %, respectively (PC1-3, $R^2X(cum)=0.773$, $Q^2(cum)=0.582$) (Table A.4). Only PC1 and PC3 are significant, as the model incorporates 25 separate samples, and the Q^2 value for PC2 is below 0.05 (Eriksson et al., 2013). The score values are presented in Table A.5, and the loading values in Table A.6. Loading values close to zero indicate little influence on the variation. The PCA for insects/spiders resulted in a model with three significant principal components, explaining 55 %, 12.2 %, and 6.9 %, respectively (PC1-3, $R^2X(cum)=0.74$, $Q^2(cum)=0.65$) (Table A.7). The score plot for PC1 versus PC2 is shown in Figure A.1, and the corresponding loading plot in Figure A.2. The score plot in Figure A.1 shows how the second principal component, PC2, which represent the largest residual variation around PC1, accounts for the differentiation of spiders from insects. PC2 is responsible for 12.2 % of the variation. According to the corresponding loading plot in Figure A.2 spiders accumulate more of S, Zn, Hg, Cu, Ag, and P than insects. Score and loading plot for insects with PC1 plotted against PC3 are shown in Figure A.3 and A.4, respectively. These figures show that PC3 separates Cyclorrhapha from both Araneae and the rest of the insects, and that Cyclorrhapha accumulate more of B, Cd, and Se. The PCA for feathers resulted in a model with three principal components, where PC1 explained 33.8 %, PC2 explained 15.6 % and PC3 explained 11.4 % (PC1-3, $R^2X(cum)=0.689$, $Q^2(cum)=0.327$) (Table A.8). PC2 is not significant, however, as the model incorporates 29 separate samples, and the Q^2 value for PC2 is below 0.05 (Eriksson et al., 2013). The score and loading values for feathers are shown in Tables A.9 and A.10.

3.2 Spatial distribution of elements in study area

Heatmaps for all materials based on mean standardized concentrations of selected metals related to coal (As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, Yb) are shown in Figure 6. The soil heatmap in Figure 6 show elevated mean concentrations of these elements in cells C3, C5, and D4, as well as close to the mine adit. Insects and spiders have their highest concentrations in cell D3, where Bansø was located. Spiders also have very high concentrations in cell E1, located by the estuary of the Todal river. The feather heatmap in

Figure 6 show high mean concentration in cell A2, close to the mine adit, and in cells neighbouring D3 (Bansø weather station).

Heatmaps showing elements related to meteorology and Bansø weather station (Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, Zr) are shown in Figure 7. The soil heatmap in Figure 7 show elevated mean concentrations of these elements in several cells. Insects and spiders both have high mean concentrations in cell D3, where Bansø was located. Insects show high mean concentrations also in cell E3, close to the Bansø station, while the spiders have elevated levels in cell E1, which as mentioned is located by the Todal river estuary. There are no data for feather samples in cell D3, but the neighbouring cells C4 and C3 show high mean concentrations. Cell A2 and A3, both located close to the mine adit, also show elevated mean concentrations in feathers.

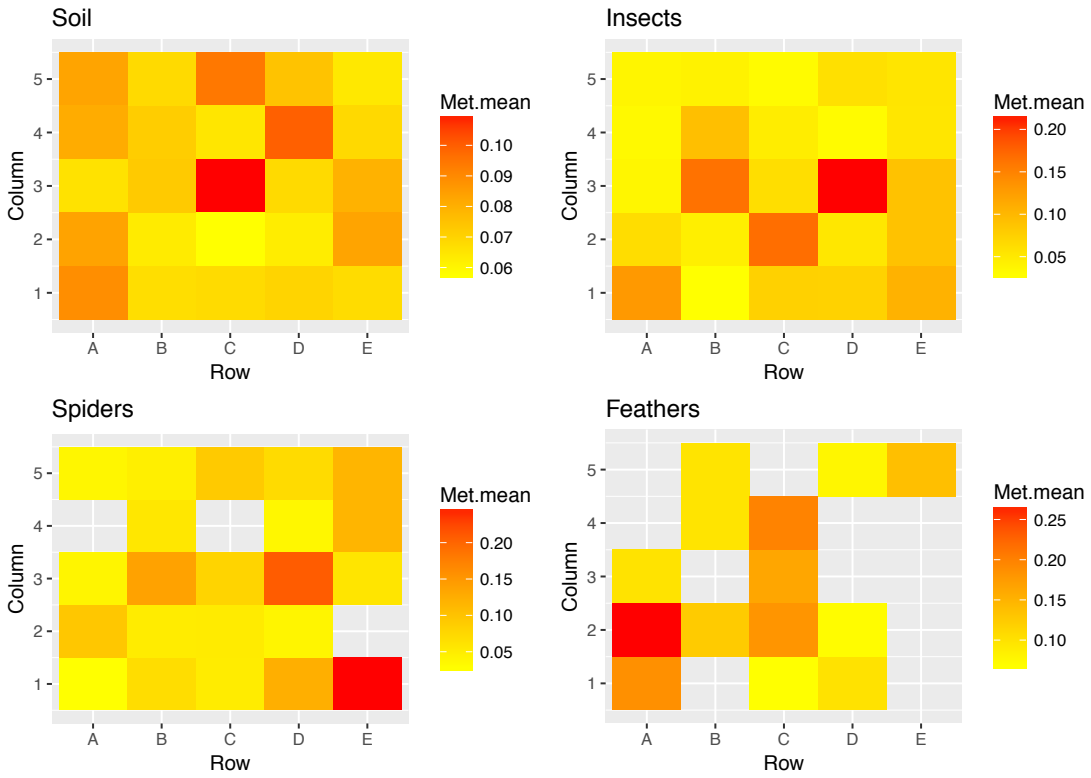

Figure 6. Mean concentrations (µg/g) in soil, insects, spiders, and feathers based on standardized concentrations of coal related elements As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, and Yb. Rows and columns are consistent with the cells in Figure 1. Data below LOD is replaced with LOD×DF.

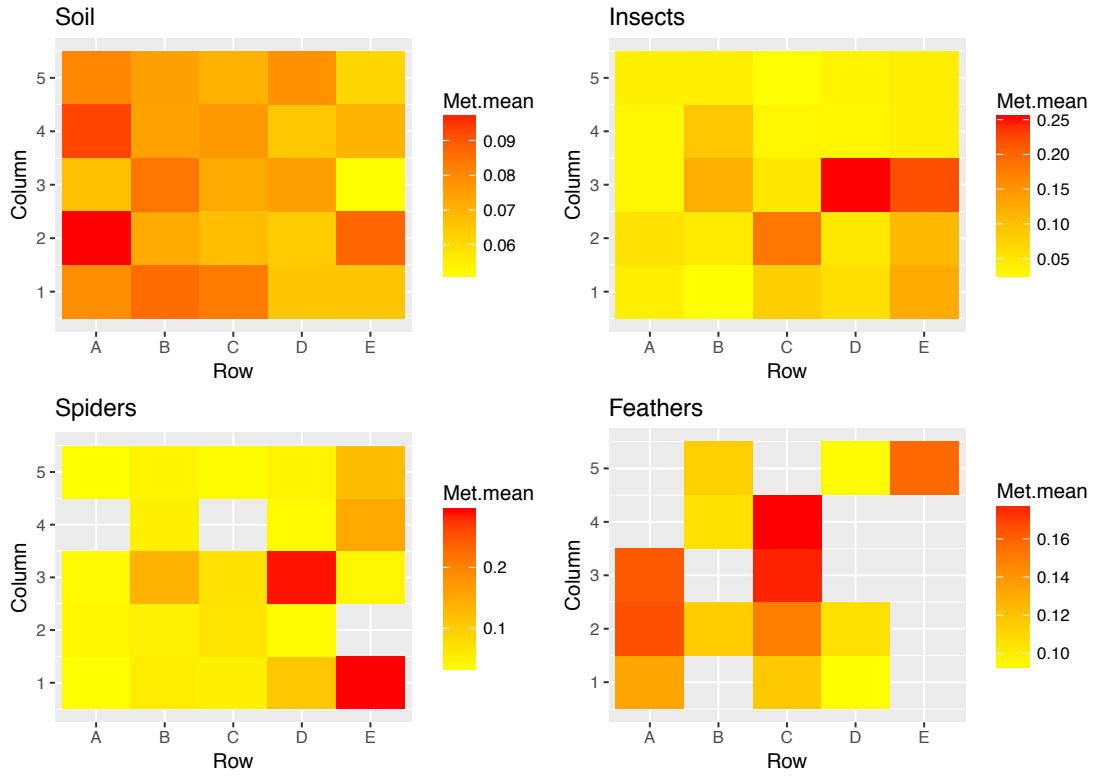

Figure 7. Mean concentrations in soil, insects, spiders and feathers based on standardized concentrations of Bansø related elements Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, and Zr. All numbers are given in µg/g. Rows and columns are consistent with the cells in Figure 1. Data below LOD is replaced with LOD×DF.

Heatmaps based on mean concentrations of all investigated elements (i.e. both coal related and Bansø related elements) are found in Figure A.5 (Appendix A.3). Heatmaps presenting the distribution of individual element concentrations in all analytical matrices, separately, are shown in Figures A.6-A.9 (all in Appendix A.3).

Bar charts showing accumulation of all investigated metals in soil, feathers, Araneae, Collembola, Cyclorrhapha, Hymenoptera, and Nematocera are shown in Figure A.10 (Appendix A.4). Complete tables with results from the HR-ICP-MS analysis for all analytical matrices are shown in Tables A.11-A.14 (Appendix A.5). A compressed table with mean, median, standard deviation, minimum and maximum concentrations is shown in Table A.15 (Appendix A.5). Tables A.11-A.15 present concentrations in µg/g, dry weight (dw). Table A.16 (Appendix A.6) shows the results from the linear mixed-effect model, including p-values. The p-values are used to determine whether the cells are statistically significant, with cell A1 as a reference cell.

4 Discussion

4.1 Principal Component Analysis

4.1.1 Soil

According to Figures 4 and 5, soil contain most of Al, As, B, Ba, Be, Ce, Co, Cr, Fe, Li, Mn, Mo, Ni, Pb, Rb, Tl, V, and Zr. As, Be, Cr, Ni, and Pb are major toxic elements, and according to the bar charts in Figure A.10 (Appendix A.4), soil as analytical matrix contains higher levels of these compared to the other matrices. High levels of these non-essential and toxic elements can be indicators of pollution. The quantified concentrations expressed in Figure A.10 show that the concentration of As and Ni are higher than the average concentrations in the Earth's crust, whereas the quantified concentrations of Be are lower than the average and much lower than studied concentrations in coal (Kabata-Pendias, 2011). Cr is also found in lower concentrations than the general distribution in the Earth's upper crust, and Pb is present at approximate average concentrations (Kabata-Pendias, 2011). Some of the other elements mainly related to soil, Co, Fe, Mn, and Mo, are essential elements and thus expected found in soil. Minor toxic elements, e.g. Ba and V, might cause toxicity in higher concentrations.

The PCA plots in Figures 4 and 5 do not show any apparent contamination pattern in soil in the study area. Figure A.6 (Appendix A.3) presents soil concentrations of all elements in every cell, and shows that various cells differ with regards to element concentrations in soil. This missing overall pattern indicates that element concentrations in soil are affected by local sources. Since the possible contamination sources vary between cells, it can be assumed that these local sources are of anthropogenic origin.

4.1.2 Insects and spiders

The PCA results for insects and spiders (Figures A.1 and A.2, Appendix A.2) show high relatedness to Ag, Cd, Cu, Hg, P, S, Se, and Zn. Figure A.1 also show a clear separation between spiders and insects, which can be explained by their different needs for elements. A possible explanation for the spiders' higher levels of Cu is based on the gas exchange in insects and spiders. This is most often facilitated through tracheae and tracheoles, which enables oxygen to diffuse to the metabolically active tissues (Brusca and Brusca, 2003, Pick et al., 2009). Oxygen-transporting proteins have therefore been regarded as unnecessary. Other arthropod subphyla, among them chelicerata (including spiders), use hemocyanins in their respiration. Hemocyanins are Cu-containing respiratory proteins enabling specific O_2-transport (Pick et al., 2009, Rehm et al., 2012). The oxygen-binding site on a hemocyanin

molecule is two Cu ions linked to an amino acid side chain (Brusca and Brusca, 2003, Burmester, 2004, Rehm et al., 2012, van Holde et al., 2001). Not all chelicerates have hemocyanins, and others have both trachea and hemocyanins. For the hemocyanin-lacking Acari (mites and ticks), diffusion through tracheas is sufficient for supporting the aerobic metabolism, because of their small body size (Rehm et al., 2012). Cu naturally associates with Ag and Zn (Ottesen et al., 2010), which might explain the elevated concentrations of these elements in spiders. However, little is known about specific Zn receptors in insects (Stolpe and Mueller, 2016). S and P are essential elements, and their elevated concentrations in spiders can be explained by a higher need for these elements. Hg, however, is a non-essential and bioaccumulative element. The bar charts (Figure A.10, Appendix A.4) show that spiders have high accumulation levels of Hg, as compared to insects. A possible explanation of why spiders accumulate more Hg than insects, is their carnivorous nature and coping mechanisms regarding heavy metal contamination (Migula et al., 2013, Wilczek et al., 2005).

The division between Cyclorrhapha, Araneae and the rest of the insects is shown in Figures A.3 and A.4 (both in Appendix A.2). According to the bar charts (Figure A.10, Appendix A.4), Cyclorrhapha accumulate more of B, Cd, and Se than the other insect types. These elements are not associated with each other (Ottesen et al., 2010). Both B and Se are essential elements, and can be regulated to a certain degree by several species (Gentz and Grace, 2006, White and Zasoski, 1999). Regulation of these elements can lead to rather constant body concentrations over a range of soil concentrations (Heikens et al., 2001), and it is difficult to deduce whether these essential elements are present in toxic concentrations or not. Cd is considered one of the most ecotoxic metals, and the US Environmental Protection Agency recognizes it as a priority toxic pollutant (Kabata-Pendias, 2011, Mogren and Trumble, 2010). Cd exhibits adverse effects on various biological processes as it can be transferred up the food chain and possibly biomagnify (Kabata-Pendias, 2011, Sanita di Toppi and Gabbrielli, 1999, Stolpe and Mueller, 2016). Cd has previously been found bioaccumulating in flies (Diener et al., 2015, van der Fels-Klerx et al., 2016), so the division between the fly taxa Cyclorrhapha and other insects and spiders is not a surprising result. There is probably little or no regulation of Cd in flies (Heikens et al., 2001).

4.1.3 Snow bunting feathers

The PCA results in Figures 4 and 5 show that the elements accumulating in feathers are mainly Hg and S. The bar charts (Figure A.10, Appendix A.4) confirm that feathers contain more of these elements than the other analytical matrices do. Feather proteins consist of the

sulphur-containing amino acid cysteine, which is important for feather growth (Almquist, 1959, Murphy et al., 1990). Hg is a volatile and bioaccumulating element (Arctic Monitoring and Assessment Programme, 2005). Thompson and Furness (1989) showed that essentially 100 % of Hg in feathers is methylmercury, a highly toxic form capable of causing central nervous system effects, loss of coordination, starvation, reproductive failure and reduction in growth and development (Arctic Monitoring and Assessment Programme, 2005). Birds contaminated with toxic elements such as heavy metals have been found to have lower growth rates (Eeva and Lehikoinen, 1996) and display behavioural changes (Janssens et al., 2003). As birds do not need Hg and excrete it, elevated levels in feathers indicate contamination. The highest concentrations of Hg in snow bunting feathers are found in cell A2, near coal piles, and C2, near Bansø weather station (Figure A.9, Appendix A.3). Both coal contamination and remains of the Bansø weather station are thus possible sources of Hg contamination. Since the levels of Hg are lower in soil, spiders, and insects than in feathers of snow bunting nestlings, biomagnification can be assumed. However, there are only feather data for 14 cells, and it is therefore difficult to draw conclusions regarding this sample material.

4.1.4 Summary of PCA results

The PCA results show a division between the three analytical matrices soil, insects/spiders, and feathers. This is expected, as toxic elements accumulate differently in the various matrices.

By comparing Figures 4 and 5 with the soil pattern in Figure A.6 (Appendix A.3), it is apparent that the different cells vary in their accumulation patterns. This makes it possible to conclude that local sources are the main cause of contamination in every cell. Because the study area is relatively small, sources of natural origin will probably be less important than anthropogenic sources. Ground-dwelling invertebrates will be affected by their surrounding soil, and concentrations in insects and spiders can thus be assumed to reflect the values in soil. According to Figures A.7 and A.8 (Appendix A.5) showing concentrations for insects and spiders in every cell, this is not the case. These figures show different patterns from Figure A.6, and bioaccumulation from soil to insects cannot be assumed for all elements. However, the results imply that biomagnification of some elements, e.g. Hg, is present, as levels increase from spiders and insects to feathers.

4.2 Contamination related to coal mining

Figure 6 is based on standardized mean concentrations of 16 elements known to be related to coal composition (As, B, Be, Cd, Co, Mo, Th, U, Y) or otherwise related to mining processes (Fe, Mn, Ni, Pb, S, Sm, Yb) (Kabata-Pendias, 2011, Ottesen et al., 2010) in all investigated matrices. The assumptions related to this model predict elevated concentrations in the cells located near and below the coal mine adit (cells A1-A4), as well as in cells bordering the estuary of the Endal river, compared to the reference cell A1. The elevated concentrations may be caused by leaching or AMD, and might vary in different sample materials.

4.2.1 Soil

The soil heatmap in Figure 6 show that soil has higher mean concentrations in cells C3, C5, and D4 compared to the other cells. However, none of these cells are statistically significant ($p>0.05$). They are not located close to the mine adit or coal piles, but cells C3 and C5 encompass cable trestles. During mining operations these trestles transported coal from the mines to the harbour, and a lot of coal might have been lost during transport. These coal remnants might be a source of contamination. Cell C5 is also in close proximity to the Endal river, which might carry contaminants from passing the main entrance of Mine 5 in Endalen. In addition, soil samples have slightly elevated mean concentrations of coal related elements in cells A1, A2, and A4, all located close to the mine adit. The coal piles and remains of the mine adit might be the main source of contamination in these cells, and both cell A1 and A2 are statistically significant ($p<0.05$).

The bar charts (Figure A.10, Appendix A.4) show that soil have high concentrations of all elements except Co, Hg, S, Sb, Se, and Zn. Soil is a sink for contaminants, and high concentrations are expected. As Wuana and Okieimen (2011) found, the most commonly found heavy metals in soil are As, Cd, Cr, Cu, Hg, Pb, and Zn. In the present study, As, Cd, and Pb are selected as coal related elements. As and Pb show elevated concentrations near the coal mine adit, whereas Cd concentrations are elevated in cell E3, neighbouring the Bansø weather station. However, none of these elements are found in high concentrations in cell A3, which is the cell located closest to the coal mine adit. This finding, in addition to the low concentrations of the aforementioned elements, indicates that coal does not have a particularly great impact on soil concentrations.

4.2.2 Insects and spiders

The insect heatmap in Figure 6 show elevated concentrations in cell B3 ($p<0.01$), located near a stream passing coal piles and cabins, cell C2 ($p<0.01$), located near Bansø weather station, and cell D3 ($p<0.01$), where Bansø was located. The spider heatmap in Figure 6 show elevated concentration in cells D3 and E1 ($p<0.01$), the latter located by the Todal river estuary. None of these cells are bordering the coal mine adit. These findings can be interpreted as meaning that the coal mine not necessarily affects the toxic element concentrations in invertebrates. This is consistent with the results for coal related elements in soil.

The bar charts (Figure A.10, Appendix A.4) show that elements accumulate differently in spiders and insects. The most important results are that Araneae have high concentrations of Cd and Hg, and Cyclorrhapha have high concentrations of Cd. These results are important because of the elements' toxic properties. As previously mentioned, both elements are highly ecotoxic with potential of bioaccumulation and biomagnification. Hg might be related to coal use and combustion, but are not a major component in it (Drevnick et al., 2012, Higman and van der Burgt, 2008). Cd have previously been associated with mine tailings (García-Giménez and Jiménez-Ballesta, 2017). These indistinct associations leads to the assumption that other sources than mine tailings or coal piles must be the main causes of contamination.

4.2.3 Snow bunting feathers

According to the feather heatmap in Figure 6, feathers have particularly high mean levels in cells A1 and A2 (both with $p<0.05$). As mentioned, these cells are located close to both the coal mine adit and large coal heaps. However, since adult birds feeding their chicks find food in their vicinity and not exclusively in certain cells, other reasons for contamination cannot be excluded. The feather heatmap also show elevated concentrations in cells C2 ($p<0.01$), C3 ($p>0.05$), and C4 ($p<0.05$), all neighbouring cell D3, the location of the Bansø station. However, since there is no data for cell D3, other sources than Bansø weather station must be considered. The electric masts and cable trestles, with its related coal piles, stretches throughout cells C2, C3 and C4, and may be possible sources of contamination of coal related elements.

The bar charts (Figure A.10, Appendix A.4) show high concentrations of As, Hg, S, Se, and Zn in feathers. The heatmaps for feathers (Figure A.9, Appendix A.3) show that both As, Hg, S, and Zn have especially high concentrations in cell A2. This cell is located close to

coal heaps and the mine adit. Spiders show a similar pattern, whereas insects do not. This study has not examined the food intake of snow bunting nestlings, and can therefore not confirm that the high concentrations in cell A2 are due to diet. As is a known component of coal (Kabata-Pendias, 2011), whereas Hg, as previously mentioned, is not. Agusa et al. (2005) studied body distribution and transfer of trace elements to eggs and feathers in black-tailed gulls (*Larus crassirostris*), and found that feathers were the significant excretory pathway for Hg, and that Se was easily transferred to eggs. Dauwe et al. (2003) suggested that Hg levels in feathers are actual reflections of levels in the blood during formation. The elevated concentrations of these major toxic elements are signs of bioaccumulation in snow bunting feathers.

4.2.4 Summary of contamination related to coal mining

Figure 6 shows that the assumption of higher element concentrations close to the coal mine adit or the Endal river is true for feathers and to some extent soil, but not for insects or spiders. The elevated element concentrations in soil in cells close to cable trestles might be due to the trestles themselves or the coal piles around them, as well as other related sources such as cables and building materials. Invertebrates show high concentrations of Cd and Hg, but the source of contamination is uncertain. Therefore, it cannot be concluded that the coal mine necessarily affects invertebrates. Birds might be affected, since analysed feathers show much higher concentrations of As and Hg close to the mine adit and large coal piles. However, the sources for feather concentrations are also uncertain, because of the unknown foraging behaviour of the birds.

4.3 Contamination related to Bansø weather station

Beitveit (2016) examined concentrations of 24 elements, whereas this study has focused on 30 elements. In addition to focusing on coal and mining processes, the present study includes an assumption regarding Bansø weather station. I assumed that Bansø weather station would have higher concentrations of Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, and Zr (Kabata-Pendias, 2011, Ottesen et al., 2010) in cell D3 where the station was located, as compared to the reference cell A1. Remnants of Bansø weather station include vast amounts of glass, and the six additional elements in this study are all related to glass or meteorological operations. Beitveit (2016) provides no basis for comparison for these elements. According to the linear mixed effect model, cell D3 is statistically significant ($p<0.001$).

4.3.1 Soil

The soil heatmap in Figure 7 show elevated mean concentrations in several cells, the highest concentrations neighbouring the mine adit. All Bansø related elements originate partly from geogenic sources like local bedrock, and elevated concentrations are therefore expected. However, considering the assumptions regarding Bansø related elements, there is no apparent pattern of where the highest concentrations are found. The soil samples show only slightly elevated concentrations in cells in close proximity to cell D3, which encompasses Bansø. As previously mentioned, the bar charts (Figure A.10, Appendix A.4) show that soil contain high concentrations of most elements throughout the study area. The quantified concentrations of Al, Ba, Ce, Cr, La, Li, Rb, Tl, V, and Zr are higher in soil than in other analytical matrices. Of these, only Cr is a major toxic element. Cr was presumably used in building materials in Bansø, and is usually accumulated at the thin top layer of soils (Kabata-Pendias, 2011, Ottesen et al., 2010). Surprisingly, the concentrations are low for the hypothesized Bansø related elements Hg, Sb, Se, and Zn. Se and Zn are both essential elements, and high concentrations were thus expected. Hg is, as mentioned, a major toxic metal that might cause adverse effects even in low concentrations. Hg is a component in thermometers, barometers and possibly other measuring instruments used at Bansø weather station (Järup, 2003, Kabata-Pendias, 2011, Ottesen et al., 2010). Sb is used in various glass and paints, where the former is found in great amounts around Bansø. Because of the applications of Hg and Sb, high concentrations in cell D3, encompassing Bansø, would be expected. In contrast to these expectations, the concentrations were low. This might be due to the soil properties in cell D3, which are not measured. It is therefore difficult to conclude whether the levels of elements in soil are affected by the meteorological activity at Bansø weather station.

4.3.2 Insects and spiders

Figure 7 show that insects have elevated mean concentrations of Bansø related elements in cells C2 ($p<0.001$), D3 ($p<0.001$), and E3 ($p>0.05$), where D3 have the highest quantified mean concentration. D3 is the location of Bansø weather station, and E3 a neighbouring cell. C2 is located close to cabins and encompasses cable trestles. Spiders show high mean concentrations of Bansø related elements in cell D3 and E1 ($p<0.001$), the latter located in the estuary of the Endal river. The other cells in the insect and spider heatmaps in Figure 7 show low concentrations of these elements, and Bansø can thus be assumed to be the main source of contamination.

The heatmaps in Figures A.7 and A.8 (Appendix A.3) show high concentrations of almost all Bansø related elements in cell D3 or in neighbouring cells. The insect and spider heatmaps in Figure 7 substantiates this finding. This is in contrast to the soil concentrations, so bioaccumulation cannot be the only source. However, these findings indicate that contamination in the area affects invertebrates.

The bar charts in Figure A.10 (Appendix A.4) show that insects and spiders accumulate more of Ba, Hg, Sb, Se, and Zn, compared to the other sample materials. While Se and Zn are essential elements, Ba and Sb are minor toxic elements, and Hg a major toxic element. The quantified concentrations of Ba, Hg and Sb are thus an indication of contamination resulting from Bansø, suggesting that invertebrates are affected by elevated concentrations of Bansø related elements as opposed to the soil samples.

4.3.3 Snow bunting feathers

This study have no data for feathers in cell D3, which is the cell encompassing Bansø. However, the feather heatmap in Figure 7 show high mean concentrations of Bansø related elements in cells C3 ($p>0.05$) and C4 ($p<0.05$), which are neighbouring cells to cell D3. In addition, the feather samples have higher concentrations of these elements in cells A2 ($p<0.05$) and A3 ($p>0.05$), close to the coal mine adit and coal piles.

Figure A.9, showing accumulated concentrations of all selected elements in feathers, show higher concentrations of Al, Be, Cr, Fe, Hg, Mn, Rb, Sb, Th, Tl, V, and Zr in cells C2, C3 and/or C4, all bordering to Bansø's original location. As Be, Fe, Mn, and Th are selected as mostly related to coal or mining processes (Ottesen et al., 2010), the main source of contamination in these neighbouring cells is not necessarily the remains of Bansø weather station. Even though the selected Bansø related elements are components of glass, building materials, and measuring instruments, some are also related to coal combustion (Cr, Hg, Sb, Se, V) and traffic (V, Zn). Elevated concentrations of certain elements can thus be a result of just some or all of these contamination sources.

The bar charts in Figure A.10 (Appendix A.4) show that the quantified concentrations of As, Hg, S, Se, and Zn are higher for feathers than for other sample materials. The lack of overlap in elements between Figure A.9 and A.10 show that the accumulation of toxic elements in feathers is not restricted to the cells neighbouring cell D3. Hg is the only exception, as it is found in elevated concentrations in cells C2, C3, and to some extent C4 (all bordering cell D3), and highly accumulates in feathers. Pollution of Hg may result from use

of thermometers and barometers, and the elevated concentrations might be an indication of contamination from Bansø weather station.

4.3.4 Summary of contamination related to Bansø weather station

The assumption that the remains of Bansø weather station would increase concentrations of glass and meteorology related elements has been examined. Figure 7, presenting heatmaps with mean concentrations of selected elements for all analytical matrices, show that there is no apparent pattern of element concentrations in soil associated with this assumption. The elevated mean concentrations in insects, spiders, and feathers, however, make it possible to indicate that the remains of the installation Bansø affect biota in the station's vicinity. This is also substantiated by heatmaps presenting element concentrations for each separate analytical matrix, in every cell. However, other sources of contamination might affect these results. The Bansø weather station is located close to a road, electric masts, and cabins, and might thus be affected by e.g. traffic, building materials, waste, and cables stored in piles. Still, Bansø's indicated effect on toxic element concentrations in biota increases the importance of investigating similar installations elsewhere in the Arctic.

4.4 Comparison of results

The results suggest that Bansø weather station is more important for biota than the coal mine and coal piles are. Previous research conducted in Kongsfjorden, Spitsbergen, have shown that trace element concentrations are lower in local coal than in the examined uppermost centimetre of peat (Headley, 1996). Even though the impact of the coal piles in Adventdalen cannot be neglected, it is unlikely that they are the main source of toxic element concentrations in soil, invertebrates, and snow bunting feathers. Other sources of contamination must therefore be considered.

Analysed element concentrations in the different matrices are probably affected by multiple contamination sources. Since the present study focused on the impact of coal piles, mining processes, and remnants of Bansø weather station, other contamination sources might be overlooked. This includes cabins supplying the environment with excessive amounts of toxic elements through wastewater (Cd, Se), building and construction (Al, Fe, Zn), wood preservatives (As, Cr, Hg, Zn), batteries (As, Cd, Hg, Li, Mn, Ni, Pb, Sb, Zn), paints (As, Hg, Pb, Sb, Zn), glassware (Ba, Ce, La, Li, Rb, Sb, Se, V, Zn, Zr), or elements related to instruments used in Bansø (Hg) (Järup, 2003, Kabata-Pendias, 2011, Mogren and Trumble, 2010, Ottesen et al., 2010). The confined area is located close to trafficked roads, which can

lead to increased concentrations of elements related to such activities (Cd, Mn, Ni, Pb, V, Zn) (Kabata-Pendias, 2011, Ottesen et al., 2010). The topographic variations in the area must also be taken into account, as it includes a hillside, two rivers and both dry and moist areas. The Endal and Todal river pass through the area before draining into Advent river or lake Isdammen. The water discharge from these rivers includes natural products from glacial erosion and weathering of bedrock, as well as anthropogenic pollutants originating from mine adits and cabins along the river courses.

4.5 Limitations

4.5.1 Soil

Despite the small sized study area, physical properties and chemical composition of soil varied between the different cells. Stones and dryness dominated some cells (e.g. cell D3 and A3), whereas others consisted of mosses and plants anchored in wet or semi-wet soil (e.g. cell C3). In addition to varying water content, organic matter, clay content and biological entities may have differed between the cells. The amount of clay, soil pH, CEC, and redox conditions have an effect on the bioavailability of metals and adsorption of elements to soil components (Alloway, 2013). Hence, low clay content might result in less binding of metals in the soil. Since this study have not analysed any soil components except element composition, they must be considered as possible causes for differing concentrations. Moreover, external contamination during sampling and processing can have occurred.

4.5.2 Insects and spiders

Element concentrations in ground-living insects and spiders might be affected by soil properties in different cells. Because of the varying water content in the study area, some cells have better conditions for growth of food supply than others, which again can lead to increased numbers of invertebrates. The bar charts (Figure A.10, Appendix A.4) show how elements accumulate differently in different taxa. Collembola accumulate large quantities of B, Mo, and Sb, whereas Araneae accumulate more of Cd, Hg, S, and Zn. Zn is an essential heavy metal, but can lead to toxicity in higher concentrations (Mogren and Trumble, 2010). Hg is not required for normal biological functions, and can become toxic even in low concentrations (Gall et al., 2015). It is considered a global pollutant with potential of biomagnification, and is therefore important to monitor (Kabata-Pendias, 2011). However, the differences in bioaccumulation between taxa in this study might be caused by different sample composition as a consequence of limited sampling possibilities. Because the HR-ICP-

MS results include possible external contamination on insects, conclusions regarding their bioaccumulative properties cannot be drawn. In addition, the origin of some species is unknown because of flying abilities, and these individuals might therefore reflect exposure from other cells (Nummelin et al., 2007). Because of the varying species composition, it is difficult to compare the results in various cells.

In addition to varying absorption of toxic elements depending on taxa, contamination of samples might have occurred during sampling, transportation or processing. Insects and spiders were sampled using pitfall traps with ethanol. To ensure reliable analytical results, the samples were quickly removed from ethanol and freezed. By freezing the insects soon after capture, the risk of leakage of elements from the body fluids and tissues into the ethanol solution was reduced. Still, some leakage might have occurred during the time the insects were in the pitfall traps. Different species have different cuticle properties. Species with soft exoskeletons might experience rapid leaching out of insect tissues, or enriching of elements from the trapping fluid to its tissues (Braun et al., 2009). Conservation in ethanol can also lead to loss of invertebrate body mass, which again may cause significant differences in the concentrations of certain elements (Braun et al., 2009). The ideal method in this study would have been to live trap insects with non-metallic nets and freeze them immediately after catching. This could have given a more precise measurement of environmental pollutants in the insect and spider samples, as most elements in insects and external contamination would be conserved. Therefore, live trapping of invertebrates without alcohol before freezing should be considered for further studies where the aim is precise measurement of environmental pollutants. Due to limited time available for sampling it was difficult to collect enough individuals to ensure a reliable chemical analysis. All samples were sorted after weight, and all samples weighing below 0.6 mg was excluded for further analyses (Table A.3, Appendix A.1).

4.5.3 Snow bunting feathers

To interpret the accuracy of the concentrations in feathers, an understanding of the deposition is needed. As the feathers represent the circulating concentrations in the nestling's blood during feather formation (Burger and Gochfeld, 1991, Dauwe et al., 2003, Goede and Debruin, 1984), their diet is important. The nestlings are fed invertebrates, and in the present study concentrations in collected invertebrates are quantified. The nestlings are exposed to both the invertebrates' internal and external contamination. If the element concentrations in insects and spiders are affected by the ethanol sampling technique, the concentrations

measured in the snow bunting feathers will not give a realistic insight into the bird's exposure. The amount of food and feeding rate is also important, and the flight and time spent away from the nest is energetically costly. The feeding rate decreases with the distance the parents have to travel to acquire food (Falconer et al., 2008). It is therefore assumed that they fetch food in close proximity to their nest, but detailed diet data is inaccessible. In field, the parents were seen close to their nest in almost all cases, which confirms this assumption. It is possible that parents from different nests overlap in their feeding grounds. Still, using feathers as an indicator tissue for element concentration analyses is a useful non-invasive method.

Although not quantified, the nestlings body conditions appeared to differ both between and within nests, possibly resulting from varying clutch size, body size, feeding rates and amounts, individual differences in absorption and disposition, elimination rates, pulmonary ventilation, and multiple routs of exposure. Varying food provisioning may have lead to differing gastrointestinal absorption, which again may have caused varying excretion to feathers. However, the chemical analyses treated all nests as single samples, and therefore the differences in diet between nests in different cells is the most important factor. Because of unknown foraging behaviour by the parents, however, this cannot be discussed in this study.

Internal tissues are considered more reliable sources of information regarding recent exposure in birds (Ansara-Ross et al., 2013). Dauwe et al. (2000) examined nestling feathers of great tits (*Parus major*) and blue tits (*Parus caeruleus*), and found that they appeared to be suitable only for monitoring lead pollution. Future research should be conducted considering whether toxic element concentrations in feathers reflect internal tissue levels and food chain exposure. Feather sampling can give a misleading indication of incorporated contaminant exposure in nestlings at the time of feather growth, as it is influenced not only by diet, but also moult and external deposition. The latter include contamination from the nests, which are composed of materials like reindeer hairs, feathers from other birds, and grass. Contamination can also occur from the deposition of contaminants during preening (Goede and Debruin, 1984). The four-step washing procedure used in this study is quite common (Ansara-Ross et al., 2013, Burger et al., 2007a, Burger et al., 2007b, Costa et al., 2013, Dauwe et al., 2000), but Cardiel et al. (2011) demonstrated that no washing procedure ensures complete removal of all external contamination from feathers. Nordnes (2016) implies that feathers of nestlings are less affected by external contamination than adults, and hence is a better representation of internal concentrations.

4.6 Recommendations for future studies

The study area is chosen based on Beitveit's (2016) results, but the study could possibly benefit from expanding the study area. It may then be easier to isolate the sources of contamination. This is especially true with regards to the feather samples, as the snow buntings feed in their vicinity and not only in certain cells. By expanding the study area the territorial factor could be eliminated.

In future studies, sampling methods should be extended to include more samples from each sampling point. In this study, only one soil sample and one insect sample were sampled for all cells, and feather samples are only found in some cells. The differences between sample materials mean that correlation analyses cannot be computed, as possible correlations may be caused by distinctive physical and chemical properties in the material.

The availability of elements in soil depends on various factors, including pH, redox potential, available water content, and temperature (Alloway, 2013, Heikens et al., 2001, Kabata-Pendias, 2011). A decrease in pH might facilitate the movement of elements (Sterckeman et al., 2000), and thus, this should be examined in future studies. Other soil factors, such as CEC, granulometric fraction, organic matter and clay content, can also be important in the behaviour of trace elements (Heikens et al., 2001, Kabata-Pendias, 2011), and should also be considered investigated.

As discussed, it is difficult to ensure complete removal of external contamination from feathers. To check whether feather concentrations are an accurate measure of the snow bunting's internal concentrations, one should examine levels in other tissues of the bird for comparison. Comparing different tissue levels with concentrations in invertebrates can be helpful in investigating the possible bioaccumulation and biomagnification in the birds. However, this raises the need for examining the nestling's diets, which implies more extensive invertebrate sampling.

The present study has focused mainly on local contamination sources. However, being located close to the Endal and Todal river, atmospheric deposition resulting from snow and glacier melting may affect the results. Regarding atmospheric pollution, the purification system implemented in Longyearbyen's coal treatment plant in 2015 might have had an impact. Beitveit's (2016) and Nordnes' (2016) results reflect element concentrations in snow buntings prior to the installation of this emission control technology. When comparing the present thesis' quantified element concentrations in snow buntings with the corresponding results reported by Beitveit (2016) and Nordnes (2016), there is no clear evidence that the

purification system have had an effect on biota in Adventdalen. However, this has not been properly examined, and could be subject for further analyses. Future studies examining this area should also include examination of mosses, which are considered good terrestrial bioindicators for airborne pollution (Jaworowski, 1989). Mosses trap atmospheric particles in their leaves, and can be used for isolation of local contamination. They are also well studied, and data for comparison is available (Alloway, 2013). By sampling mosses, this study could also better have been compared to Beitveit's (2016) study.

5 Concluding remarks

This thesis has monitored concentrations of 30 elements in soil, insects, spiders, and feathers in 25 cells in Adventdalen (figure 1). Based on Beitveit's (2016) findings, elements related to coal mining and mining processes (As, B, Be, Cd, Co, Fe, Mn, Mo, Ni, Pb, S, Sm, Th, U, Y, Yb) were investigated. In addition, this thesis have examined elements linked to materials and operations related to Bansø weather station (Al, Ba, Ce, Cr, Hg, La, Li, Rb, Sb, Se, Tl, V, Zn, Zr). The PCA indicates that all examined cells in the investigated area are mainly affected by local sources of contamination, possibly of anthropogenic origin.

The results show elevated concentrations of coal related elements in soil sampled from cells close to the coal mine adit and coal piles. Invertebrates do not show a similar pattern regarding coal related elements, neither close to the coal mine adit nor in cells close to the Endal river estuary. Therefore, it can be alleged that coal contamination not necessarily affects the toxic element concentration in invertebrates, even though its impact cannot be neglected. The feather samples show elevated concentrations of the coal related elements in cells neighbouring the coal mine adit and coal piles, but since the results are based on data in only 14 of 25 cells, conclusions regarding this analytical matrix cannot be drawn. However, the results indicate that coal contamination might lead to elevated concentrations also in birds.

Soil does not show elevated concentrations of elements related to activities at the Bansø weather station, which was located in cell D3. Insects and spiders, however, show elevated concentrations of these elements at the Bansø location. There are no data for feathers in cell D3, but neighbouring cells show high concentrations of Bansø related elements also for feathers. Since the elevated concentrations are only found in biota, this indicates bioaccumulation and biomagnification, as exemplified by Hg. These results indicates the importance of the remains of Bansø weather station, even decades after its demolition. Future research should examine the impact of similar installations present elsewhere in the Arctic.

Several cells located close to rivers or melting streams in the confined area have elevated concentrations of various elements, both coal related and Bansø related elements. This is expected, as the river transfer contaminants from both natural and anthropogenic sources located along its course. Other sources such as trafficked roads, waste piles and cabin operations might affect the results, but there are no clear pattern for this.

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A. Appendices

A.1 Data processing

Table A.1. Division of insects and spiders from all cells into taxonomic groups. Number of individuals found in each cell are included.

Cell	Taxonomic group and number of individuals	Total number of individuals
1	1 Acari, 22 Araneae, 31 Cyclorrhapha, 22 Nematocera	76
2	13 Araneae, 8 Cyclorrhapha, 29 Nematocera	50
3	6 Araneae, 3 Collembola, 10 Cyclorrhapha, 2 Hymenoptera, 8 Nematocera	29
4	12 Collembola, 3 Cyclorrhapha, 24 Nematocera	39
5	17 Araneae, 6 Collembola, 10 Cyclorrhapha, 40 Nematocera	73
6	8 Araneae, 1 Collembola, 11 Cyclorrhapha, 1 Nematocera	21
7	6 Araneae, 18 Collembola, 14 Cyclorrhapha, 28 Nematocera	66
8	1 Araneae, 5 Collembola, 22 Cyclorrhapha, 40 Nematocera	68
9	2 Acari, 6 Araneae, 3 Collembola, 24 Cyclorrhapha, 1 Hymenoptera, 6 Nematocera	42
10	16 Araneae, 38 Cyclorrhapha, 17 Nematocera	71
11	7 Araneae, 5 Collembola, 25 Cyclorrhapha, 7 Hymenoptera, 3 Nematocera	47
12	5 Araneae, 6 Collembola, 21 Cyclorrhapha, 2 Hymenoptera, 5 Nematocera	39
13	5 Acari, 8 Araneae, 8 Cyclorrhapha, 5 Nematocera	26
14	17 Araneae, 10 Collembola, 11 Cyclorrhapha, 1 Nematocera	39
15	26 Araneae, 6 Collembola, 3 Cyclorrhapha, 6 Nematocera	41
16	6 Araneae, 7 Collembola, 7 Cyclorrhapha, 2 Nematocera	22
17	1 Acari, 19 Araneae, 7 Collembola, 2 Cyclorrhapha, 3 Nematocera	32
18	10 Araneae, 18 Collembola, 6 Cyclorrhapha, 12 Nematocera	46
19	5 Araneae, 23 Collembola, 18 Cyclorrhapha, 8 Nematocera	54
20	1 Acari, 16 Araneae, 8 Collembola, 16 Cyclorrhapha, 4 Hymenoptera, 1 Nematocera	46
21	12 Araneae, 15 Collembola, 3 Cyclorrhapha, 6 Nematocera,	36
22	46 Collembola, 4 Cyclorrhapha, 1 Hymenoptera, 33 Nematocera	84
23	8 Araneae, 25 Collembola, 6 Cyclorrhapha, 2 Hymenoptera, 54 Nematocera	95
24	3 Acari, 10 Araneae, 11 Collembola, 114 Cyclorrhapha, 10 Nematocera	148
25	9 Araneae, 5 Collembola, 1 Hymenoptera, 24 Nematocera	39

Table A.2. Limits of detection (LODs) for elements in different sample weight classes. Each element is reported with monitored isotope and instrument resolution (RES). LODs are given in µg/g.

Element	Isotope	RES	Maximum LOD				
			Class 1	Class 2	Class 3	Class 4	Class 5
Al	27	MR	68.05	8.71	1.95	0.34	0.08
As	75	HR	2.66	0.34	0.08	0.01	0.01
B	11	LR	18.90	2.42	0.54	0.09	0.02
Ba	137	MR	1.38	0.18	0.04	0.01	0.01
Be	9	LR	0.21	0.03	0.01	0.00	0.00
Cd	114	LR	0.21	0.03	0.01	0.00	0.00
Ce	140	LR	0.02	0.00	0.00	0.00	0.00
Co	59	MR	0.43	0.06	0.01	0.00	0.00
Cr	53	MR	3.67	0.47	0.10	0.02	0.00
Fe	56	MR	16.75	2.14	0.48	0.08	0.01
Hg	202	LR	0.11	0.01	0.00	0.00	0.00
La	139	MR	0.21	0.03	0.01	0.00	0.00
Li	7	LR	0.48	0.06	0.01	0.00	0.00
Mn	55	MR	2.50	0.32	0.07	0.01	0.00
Mo	98	MR	2.13	0.27	0.06	0.01	0.01
Ni	60	MR	1.60	0.20	0.05	0.01	0.01
Pb	208	LR	0.26	0.03	0.01	0.00	0.00
Rb	85	MR	1.28	0.16	0.04	0.01	0.00
S	34	MR	2129.93	272.59	60.92	10.63	8.33
Sb	121	MR	0.54	0.07	0.02	0.00	0.00
Se	82	LR	5.32	0.68	0.15	0.03	0.02
Sm	147	LR	0.06	0.01	0.00	0.00	0.00
Th	232	LR	0.18	0.02	0.01	0.00	0.00
Tl	205	LR	0.03	0.00	0.00	0.00	0.00
U	238	LR	0.03	0.00	0.00	0.00	0.00
V	51	MR	0.32	0.04	0.01	0.00	0.00
Y	89	LR	0.04	0.01	0.00	0.00	0.00
Yb	172	LR	0.04	0.01	0.00	0.00	0.00
Zn	66	MR	4.10	0.52	0.12	0.02	0.01
Zr	90	LR	0.12	0.02	0.00	0.00	0.00

Table A.3. Division of samples based on sample weight.

Class	Sample weight interval (mg)	Material	Average sample weight (mg)
1	0.1-0.5	Insects, spiders	0.188
2	0.6-3.0	Insects, spiders, feathers	1.467
3	3.3-15.1	Insects, spiders, feathers	6.566
4	18.2-102.1	Insects	37.644
5	205.9-293.7	Soil	259.381

A.2 Principal Component Analysis

Table A.4. Percentage contribution of the principal components in the PCA of soil. This model incorporates 25 samples, and the principal components are significant if $Q^2 > 0.05$. Thus, principal components 1 and 3 are significant. R^2X represents the fraction of the total variation of X or Y that can be predicted by a principal component.

Component	R^2X	Eigenvalue	Q^2	Q^2(cum)
1	0.598	14.9	0.536	0.536
2	0.0998	2.49	0.019	0.544
3	0.0755	1.89	0.083	0.582

Table A.5. Score values resulting from PCA of soil.

Cell	M2.t[1] (PC1)	M2.t[2] (PC2)	M2.t[3] (PC3)
1	5.1085	-1.4190	-0.1205
2	0.1443	-0.9315	1.7867
3	9.4736	-3.2759	-0.4125
4	-7.2746	-1.5514	4.8001
5	-2.6474	-5.5601	2.1276
6	5.7493	-1.7635	0.1913
7	2.6402	-1.8048	-1.6279
8	-2.6797	-2.1776	0.5513
9	4.3382	-0.7988	3.2941
10	-1.6076	0.6290	0.6682
11	10.265	3.0758	0.1679
12	-3.3522	-0.4481	-1.9710
13	12.8595	2.3196	2.0627
14	-2.1037	-3.4450	-2.8047
15	-2.3698	1.3524	-1.4257
16	-6.0971	-0.5825	-0.8093
17	-1.3201	0.0827	-2.0942
18	-4.5741	1.3930	-1.4097
19	-4.2065	-0.6287	-2.0942
20	-1.8455	1.6980	-2.9799
21	-1.0785	3.7202	1.8868
22	-6.8495	2.6230	1.2388
23	6.9141	1.5847	-3.1342
24	-9.5097	2.7996	0.4378
25	0.0233	3.1089	1.6705

Table A.6. Loading values resulting from PCA of soil.

Metal	M2.p[1] (PC1)	M2.p[2] (PC2)	M2.p[3] (PC3)
Ag	0.0943	0.0570	0.1493
Al	-0.1698	-0.0213	0.0159
As	-0.1609	-0.0224	-0.0424
Au	0.0513	0.0394	0.0422
B	-0.0876	0.2798	-0.0708
Ba	-0.0548	0.1744	0.3402
Be	-0.1224	0.1085	0.1906
Bi	-0.1604	0.0494	0.0740
Ca	0.1321	0.1722	0.0759
Cd	0.1514	0.0394	0.0621
Ce	-0.1692	-0.0568	-0.0111
Co	0.0727	0.1497	0.2976
Cr	-0.1704	-0.0087	-0.0367
Cs	-0.1644	0.0803	-0.0248
Cu	-0.0706	0.0665	0.3240
Dy	-0.1632	0.0169	0.1064
Er	-0.1635	0.0337	0.0974
Fe	-0.1618	-0.0445	0.0697
Ga	-0.1698	-0.0112	0.0037
Hf	-0.1212	0.2498	-0.1578
Hg	0.1080	0.1934	-0.0310
K	-0.1693	-0.0262	-0.0013
La	-0.1691	-0.0634	-0.0136
Li	-0.1695	-0.0367	-0.0086
Lu	-0.1576	0.0929	0.0875
Mg	-0.1424	0.0756	0.0222
Mn	0.1321	0.0790	0.2615
Mo	-0.0219	0.1844	-0.1660
Na	-0.0461	0.0565	0.0494
Nb	-0.0086	0.2456	-0.1250
Nd	-0.1693	-0.0596	0.0228
Ni	0.0902	0.1251	0.3241
P	0.1232	0.1125	0.2444
Pb	-0.1300	0.0345	0.1196
Pr	-0.1700	-0.0545	-0.0015
Pt	-0.0834	-0.0205	0.0698
Rb	-0.1692	0.0097	0.0220
S	0.0860	0.2258	0.0350
Sb	0.0510	0.2831	-0.1310
Sc	-0.1698	0.0005	0.0033
Se	-0.0220	-0.0226	0.1909

Metal	M2.p[1] (PC1)	M2.p[2] (PC2)	M2.p[3] (PC3)
Si	-0.0093	0.2808	-0.2295
Sm	-0.1672	-0.0562	0.0650
Sn	-0.0323	0.3272	-0.1683
Sr	0.0705	0.2835	0.0756
Tb	-0.1639	-0.0160	0.1044
Th	-0.1643	-0.1071	0.0429
Ti	-0.1037	0.2262	-0.0772
Tl	-0.1637	0.0625	-0.0033
Tm	-0.1602	0.0805	0.0875
U	-0.1568	0.0020	0.0310
V	-0.1672	0.0130	-0.0500
Y	-0.1533	0.0686	0.1397
Yb	-0.1606	0.0736	0.0864
Zn	0.1345	0.0668	0.1672
Zr	-0.1318	0.2244	-0.1289

Table A.7. Percentage contribution of the principal components in the PCA of insects and spiders. This model incorporates 112 samples, and the principal components are significant if $Q^2 > 0$. Thus, the principal components 1-3 are significant. R^2X represents the fraction of the total variation of X or Y than can be predicted by a principal component.

Component	R^2X	Eigenvalue	Q^2	Q^2(cum)
1	0.55	27.5	0.527	0.527
2	0.122	6.08	0.176	0.61
3	0.0692	3.46	0.102	0.65

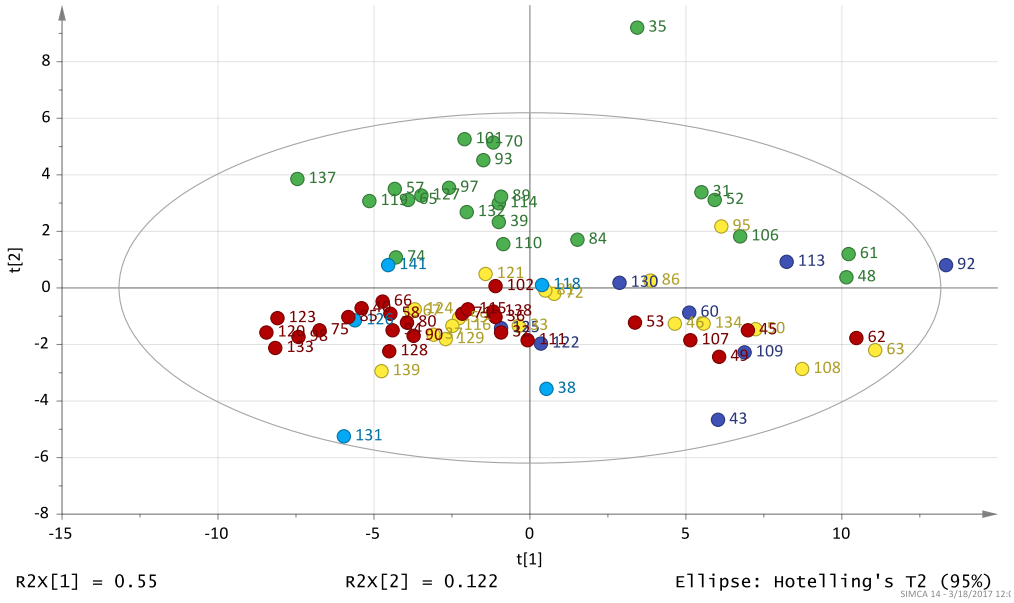


Figure A.1. PCA score plot for insects and spiders grouped by taxonomic division into Araneae (green, n=22), Collembola (dark blue, n=8), Cyclorrhapha (red, n=25), Nematocera (yellow, n=18), and Hymenoptera (light blue, n=5). The figure is showing principal components 1 and 2, where PC1 explains 55 % and PC2 12.2 % of the variation. Numbers on the figure represent individual sample numbers. R2X=0.122 show the fraction of the variation of the variables explained by the model.

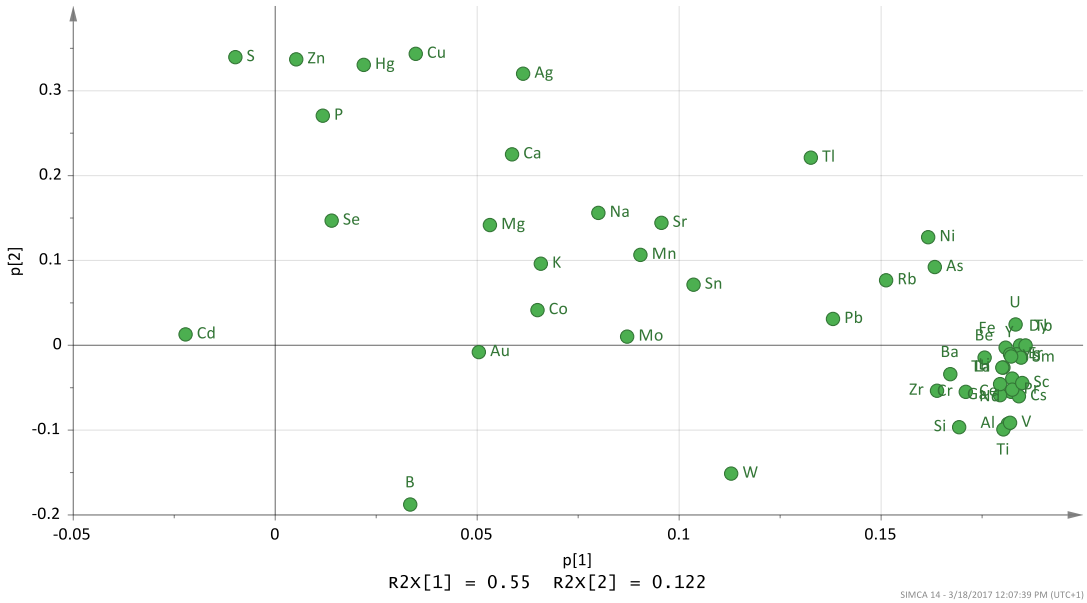


Figure A.2. PCA loading plot for insects and spiders. All elements where 50 % of the data was above LOD are presented. When comparing with Figure A.1, this figure represents which elements that are present in which insect and spider samples. The figure is showing principal components 1 and 2, where PC1 explains 55 % and PC2 12.2 % of the variation. $R^2X[1]=0.55$ show the fraction of the variation of the variables explained by the model.

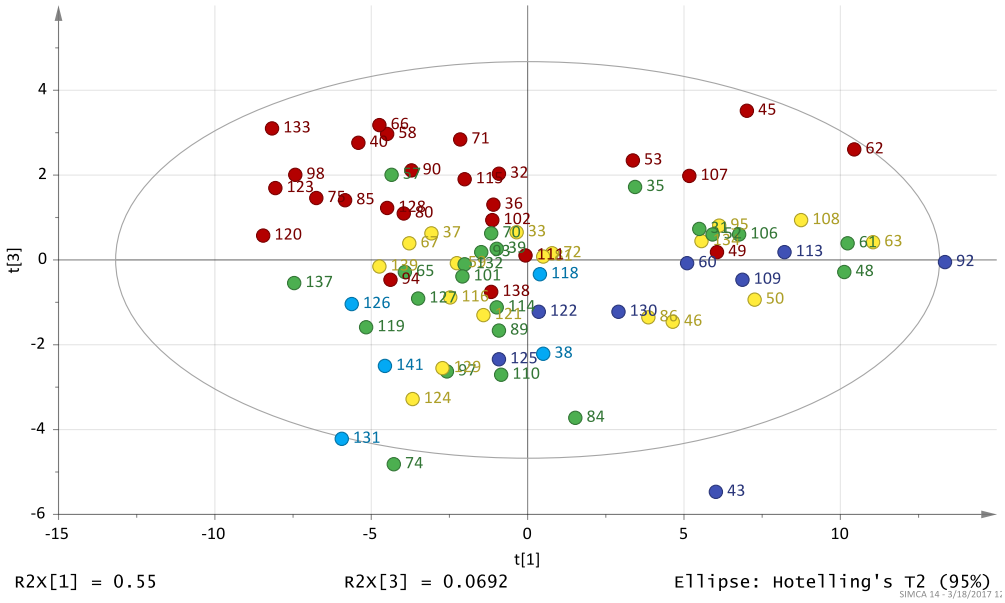


Figure A.3. PCA score plot for insects and spiders grouped by taxonomic division into Araneae (green, n=22), Collembola (dark blue, n=8), Cyclorrhapha (red, n=25), Nematocera (yellow, n=18), and Hymenoptera (light blue, n=5). The figure is showing principal components 1 and 3, where PC1 explains 55 % and PC3 6.9 % of the variation. Numbers on the figure represent individual sample numbers. R2X=0.0692 show the fraction of the variation of the variables explained by the model.

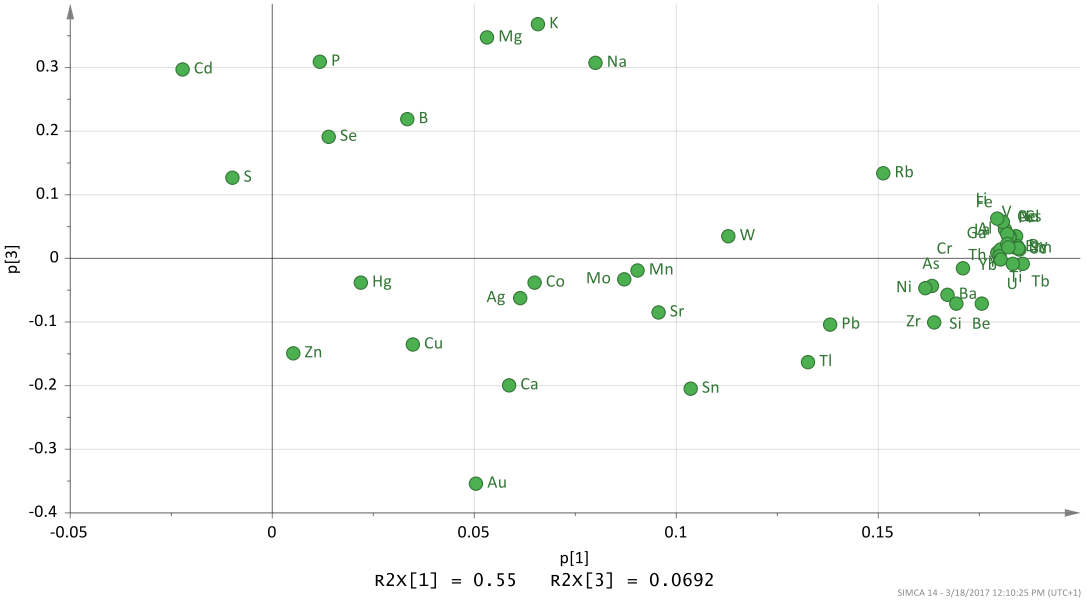


Figure A.4. PCA loading plot for insects and spiders. All elements where 50 % of the data was above LOD are presented. When comparing with Figure A.3, this figure represents which elements that are present in which insect and spider samples. The figure is showing principal components 1 and 3, where PC1 explains 55 % and PC3 6.9 % of the variation. $R^2X[1]=0.55$ show the fraction of the variation of the variables explained by the model.

Table A.8. Percentage contribution of the principal components in the PCA of feathers. This model incorporates 29 samples, and the principal components are significant if Q^2>0.05. Thus, the principal components 1 and 3 are significant. R^2X represents the fraction of the total variation of X or Y that can be predicted by a principal component.

Component	R^2X	Eigenvalue	Q^2	Q^2(cum)
1	0.338	9.81	0.264	0.264
2	0.156	4.53	0.0016	0.265
3	0.114	3.32	0.084	0.327

Table A.9. Score values resulting from PCA of feathers.

Cell	M2.t[1] (PC1)	M2.t[2] (PC2)	M2.t[3] (PC3)
15	0.4729	-0.9184	-0.4646
20	-3.4006	-2.8629	-1.0924
20	-2.2641	-2.4027	-1.0617
24	5.5886	-5.3484	-2.2413
25	1.7685	-6.8779	-4.0484
25	-2.9463	-1.1508	-1.3399
1	3.4136	1.2906	0.8712
6	-2.4806	0.8290	1.2423
6	-4.9761	1.9745	1.2477
7	-7.1015	0.0108	-0.2438
12	5.3093	2.0154	1.1570
13	-0.1009	2.6624	0.3338
13	1.5163	-1.4775	3.2894
13	2.6733	1.5318	1.3482
14	3.0031	0.5680	1.6407
17	-4.0504	1.3240	-0.2988
17	-0.7324	-1.5511	2.3484
19	2.1465	0.9238	1.8217
19	-6.4699	-1.3402	-0.4593
20	1.7909	1.4660	1.1272
20	4.5154	-3.0542	4.2355
20	-4.7562	-1.7907	1.5645
10	-3.0145	0.3367	0.2155
23	1.6202	2.1548	0.6429
25	0.1876	5.0897	-6.2073
25	-3.2147	1.7894	-0.9245
25	2.1424	1.0997	-0.3001
25	6.7561	0.9675	-3.7013
25	2.6036	2.7406	-0.7027

Table A.10. Loading values resulting from PCA of feathers.

Metal	M2.p[1]	M2.p[2]	M2.p[3]
Al	0.2486	-0.0622	0.0338
Au	-0.04010	-0.1845	-0.2214
B	0.0763	0.2991	-0.0740
Ba	0.1891	-0.0936	-0.1179
Bi	0.0806	-0.1759	-0.3309
Ca	0.0881	0.2496	-0.2857
Ce	0.2381	-0.0291	0.1161
Co	0.1565	0.0616	-0.1331
Cs	0.1320	0.0788	0.0167
Cu	0.1342	0.1332	-0.0497
Dy	0.2382	0.0339	0.0011
Er	0.1590	0.2062	0.1518
Fe	0.2267	-0.0104	0.1682
Hg	0.0642	-0.0775	0.1188
K	-0.0026	0.1719	-0.0356
La	0.2173	0.0049	0.1537
Li	0.1460	-0.1916	-0.1585
Mg	0.0409	0.2926	-0.1866
Mn	0.1669	-0.0556	0.2124
Na	0.0103	0.0016	0.0449
Nd	0.2352	-0.0118	0.1381
Ni	0.1252	-0.0782	-0.3399
P	0.0254	-0.2793	0.1118
Pb	0.0430	-0.1252	-0.1227
Pr	0.2378	-0.0248	0.1471
Rb	0.0863	0.2066	-0.0550
S	0.0536	-0.2204	-0.1748
Se	0.0168	0.0463	-0.1405
Si	0.2247	-0.1626	-0.0157
Sm	0.2161	-0.1621	0.0825
Sn	0.0978	-0.2072	-0.1962

Metal	M2.p[1] (PC1)	M2.p[2] (PC2)	M2.p[3] (PC3)
Sr	0.1105	0.2404	-0.2525
Tb	0.1878	0.1362	0.2002
Ti	0.1852	-0.0721	-0.2467
Tl	0.1167	0.2194	-0.1378
U	0.1726	0.0514	0.0100
V	0.2383	-0.0014	0.0911
Y	0.2386	0.0294	-0.1455
Yb	0.1588	0.0946	0.1061
Zn	0.0782	-0.3245	-0.0224
Zr	0.1038	0.0149	-0.0165

A.3 Heatmaps

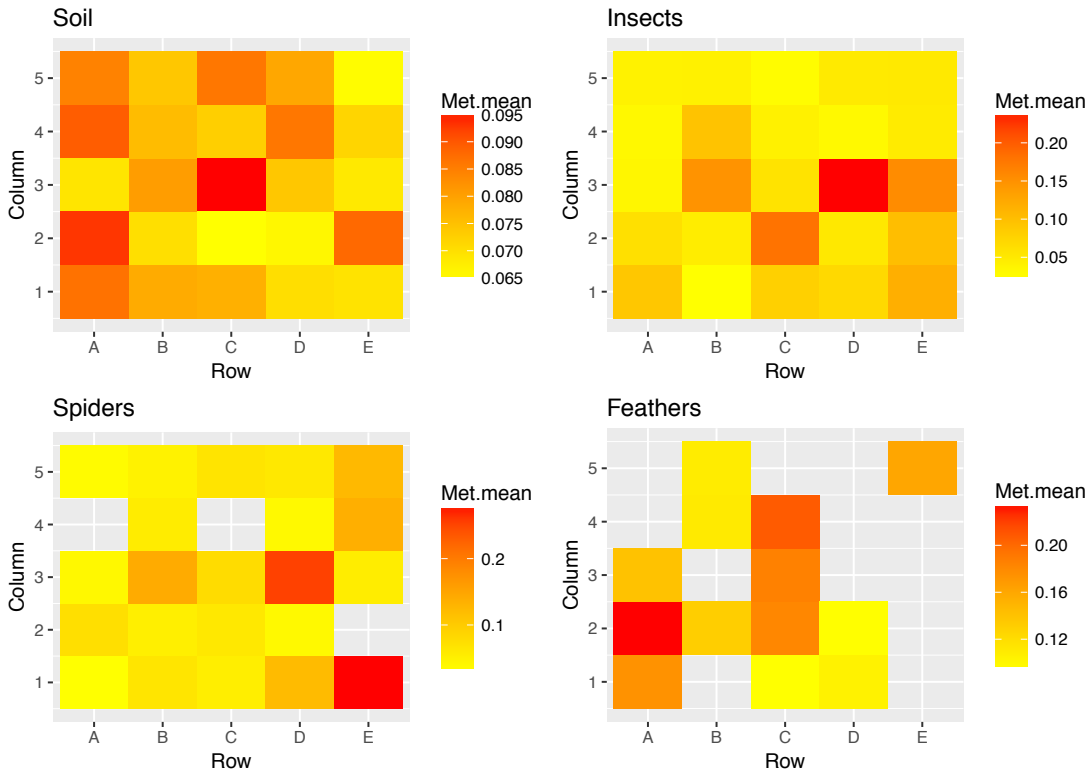
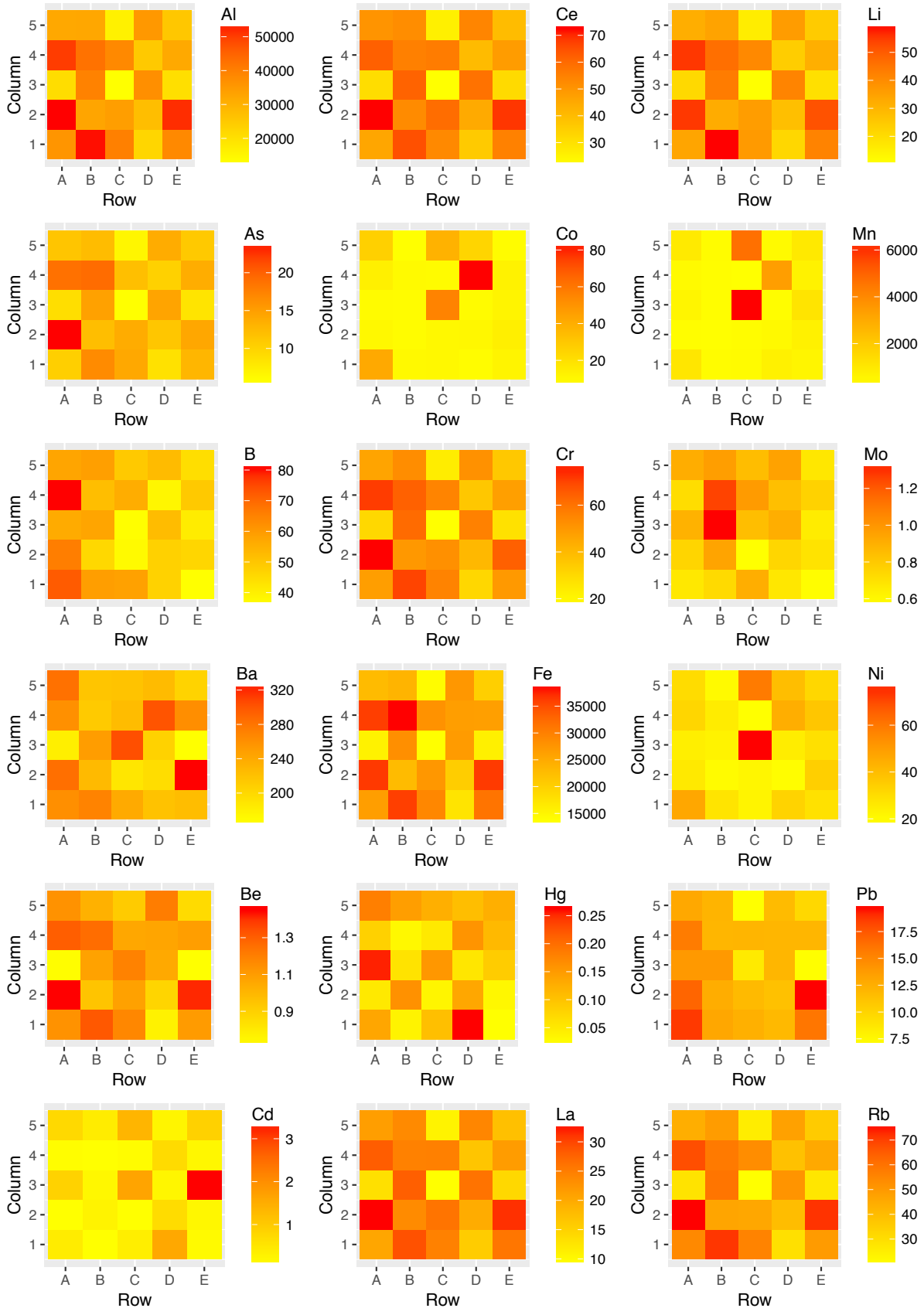


Figure A.5. Mean concentrations (µg/g) in soil, insects, spiders, and feathers based on standardized concentrations of all selected elements Al, As, B, Ba, Be, Cd, Ce, Co, Cr, Fe, Hg, La, Li, Mn, Mo, Ni, Pb, Rb, S, Sb, Se, Sm, Th, Tl, U, V, Y, Yb, Zn, and Zr. Rows and columns are consistent with the cells in figure 1. Data below LOD is replaced with LOD*DF.


Al
50000
40000
30000
20000
Ce
70
60
50
40
30
Li
50
40
30
20
As
20
15
10
Co
80
60
40
20
Mn
6000
4000
2000
B
80
70
60
50
40
Cr
60
40
20
Mo
1.2
1.0
0.8
0.6
Ba
320
280
240
200
Fe
35000
30000
25000
20000
15000
Ni
60
40
20
Be
1.3
1.1
0.9
Hg
0.25
0.20
0.15
0.10
0.05
Pb
17.5
15.0
12.5
10.0
7.5
Cd
3
2
1
La
30
25
20
15
10
Rb
70
60
50
40
30
Column
Row
1
2
3
4
5
A
B
C
D
E

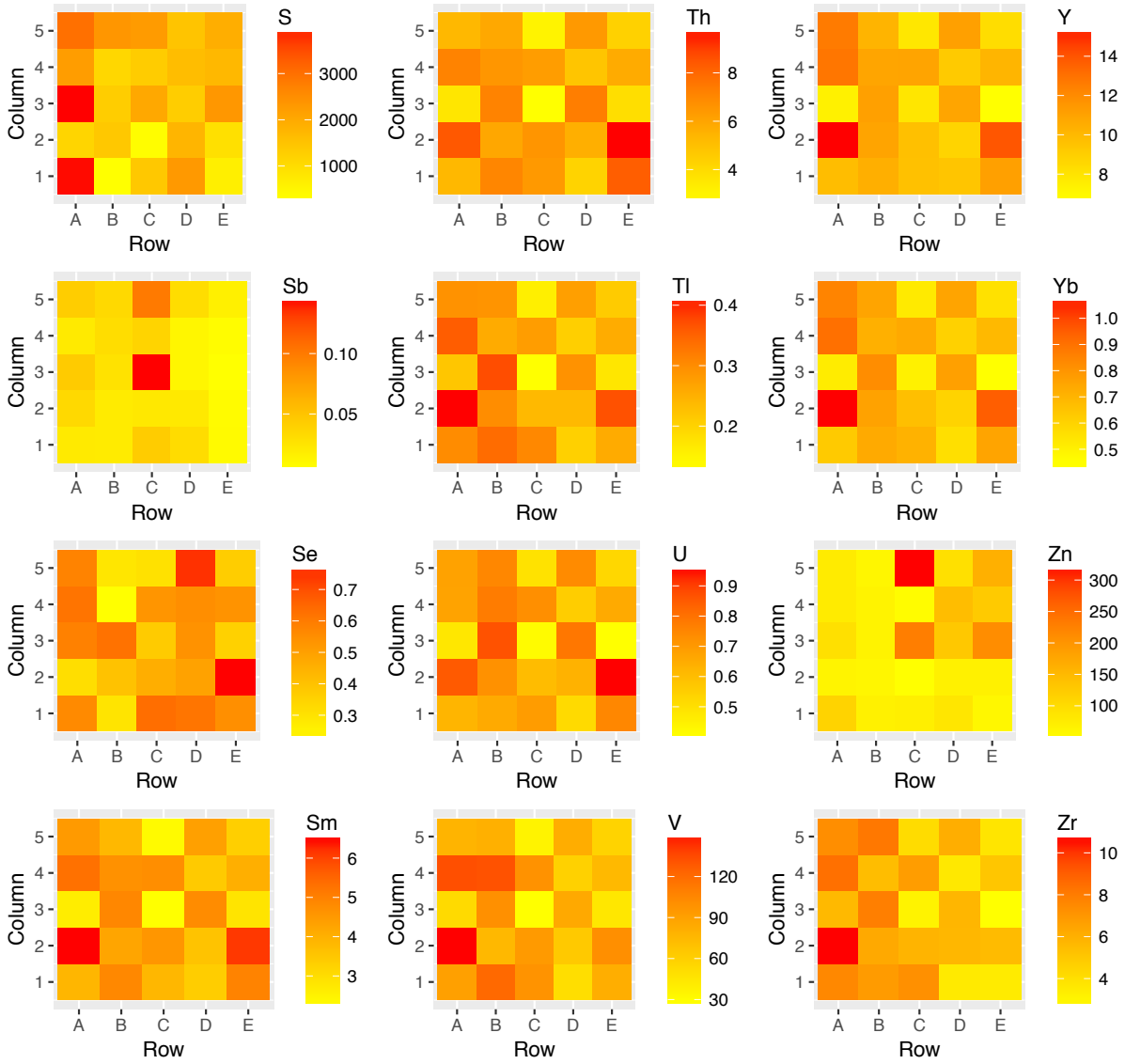
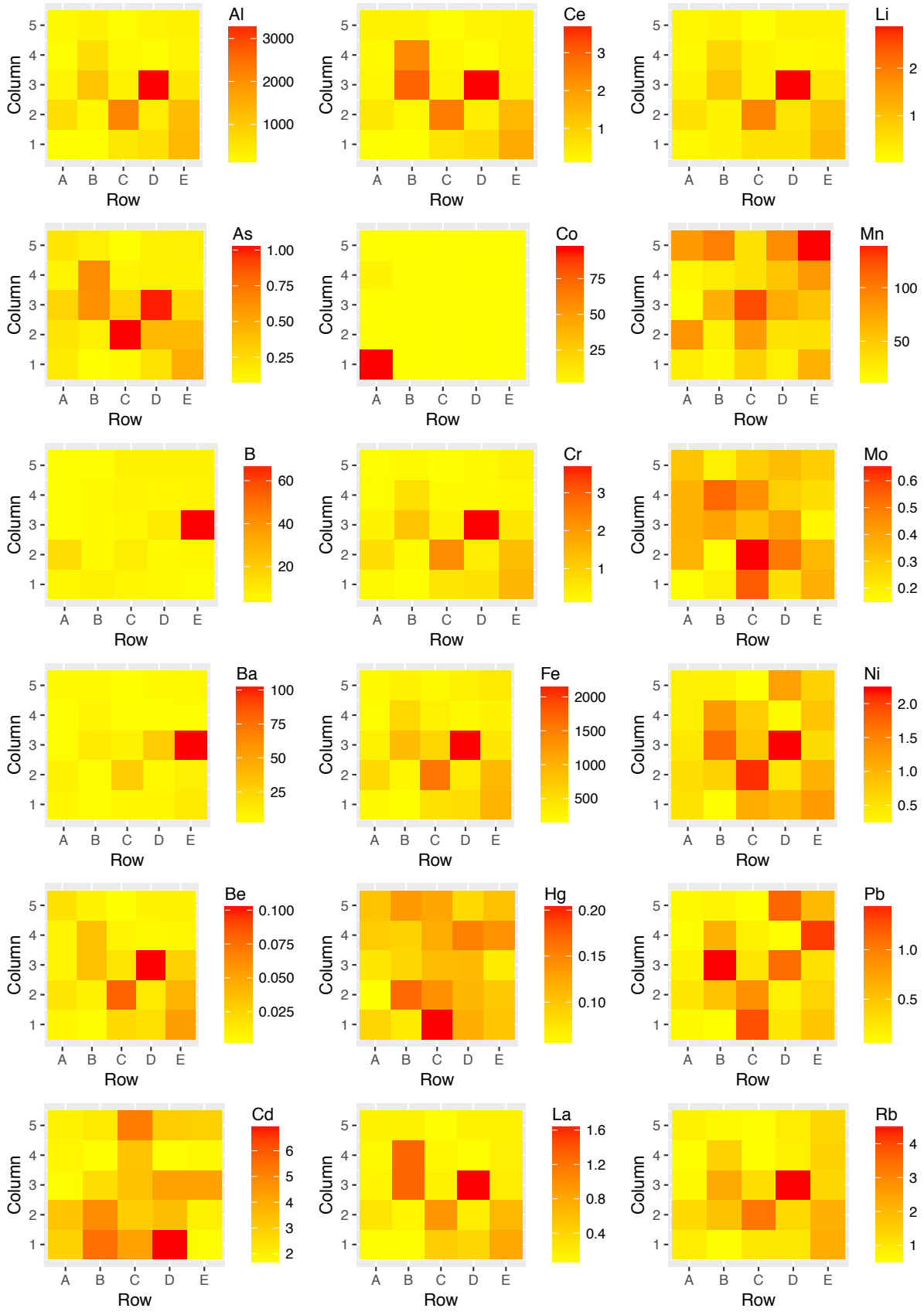


Figure A.6. Mean concentrations (µg/g) of Al, As, B, Ba, Be, Cd, Ce, Co, Cr, Fe, Hg, La, Li, Mn, Mo, Ni, Pb, Rb, S, Sb, Se, Sm, Th, Tl, U, V, Y, Yb, Zn, and Zr in soil. Rows and columns are consistent with the cells in figure 1. Data below LOD is replaced with LOD*DF.


Al
3000
2000
1000
Ce
3
2
1
Li
2
1
As
1.00
0.75
0.50
0.25
Co
75
50
25
Mn
100
50
B
60
40
20
Cr
3
2
1
Mo
0.6
0.5
0.4
0.3
0.2
Ba
100
75
50
25
Fe
2000
1500
1000
500
Ni
2.0
1.5
1.0
0.5
Be
0.100
0.075
0.050
0.025
Hg
0.20
0.15
0.10
Pb
1.0
0.5
Cd
6
5
4
3
2
La
1.6
1.2
0.8
0.4
Rb
4
3
2
1
Column
Row
1
2
3
4
5
A
B
C
D
E

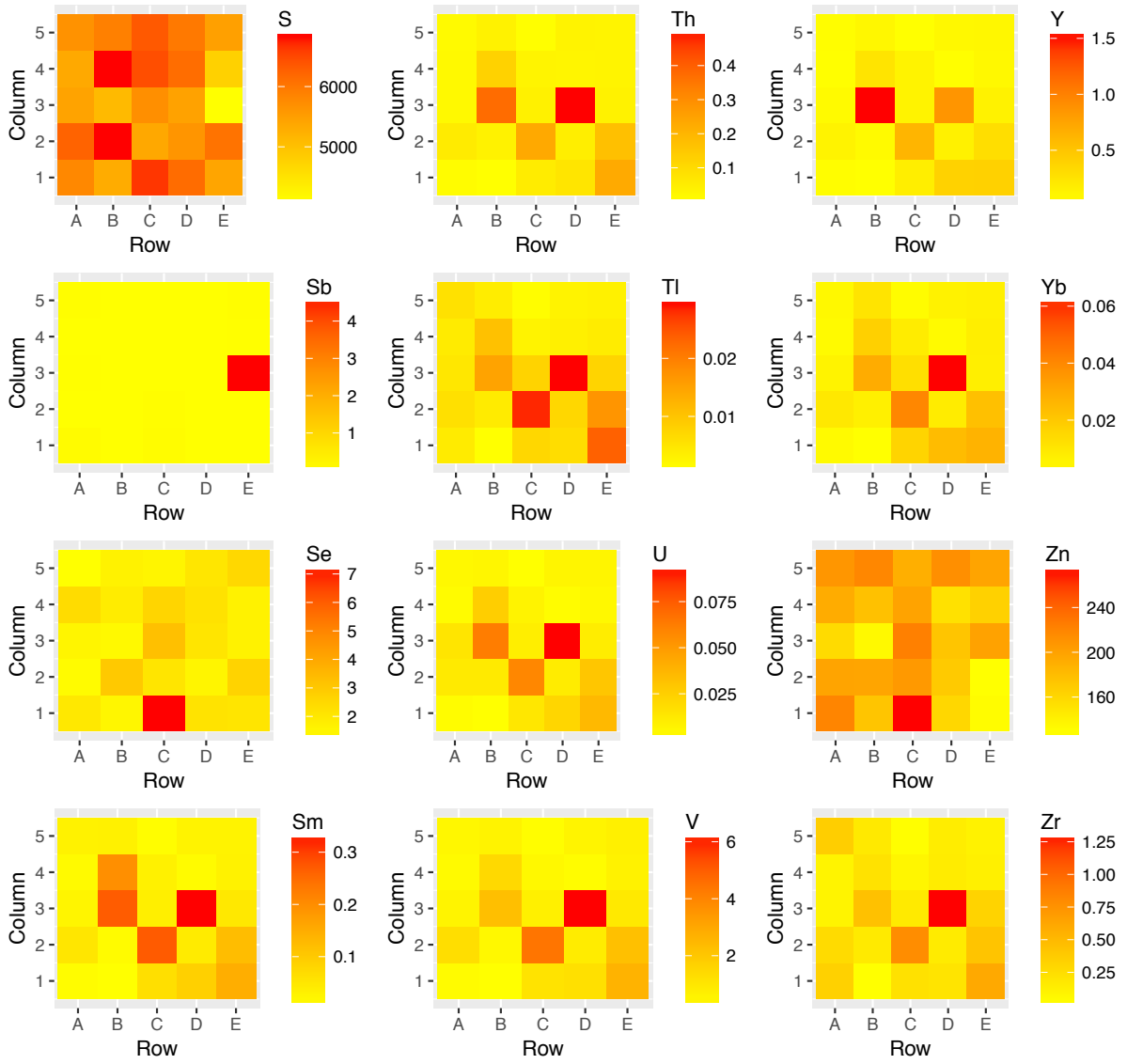
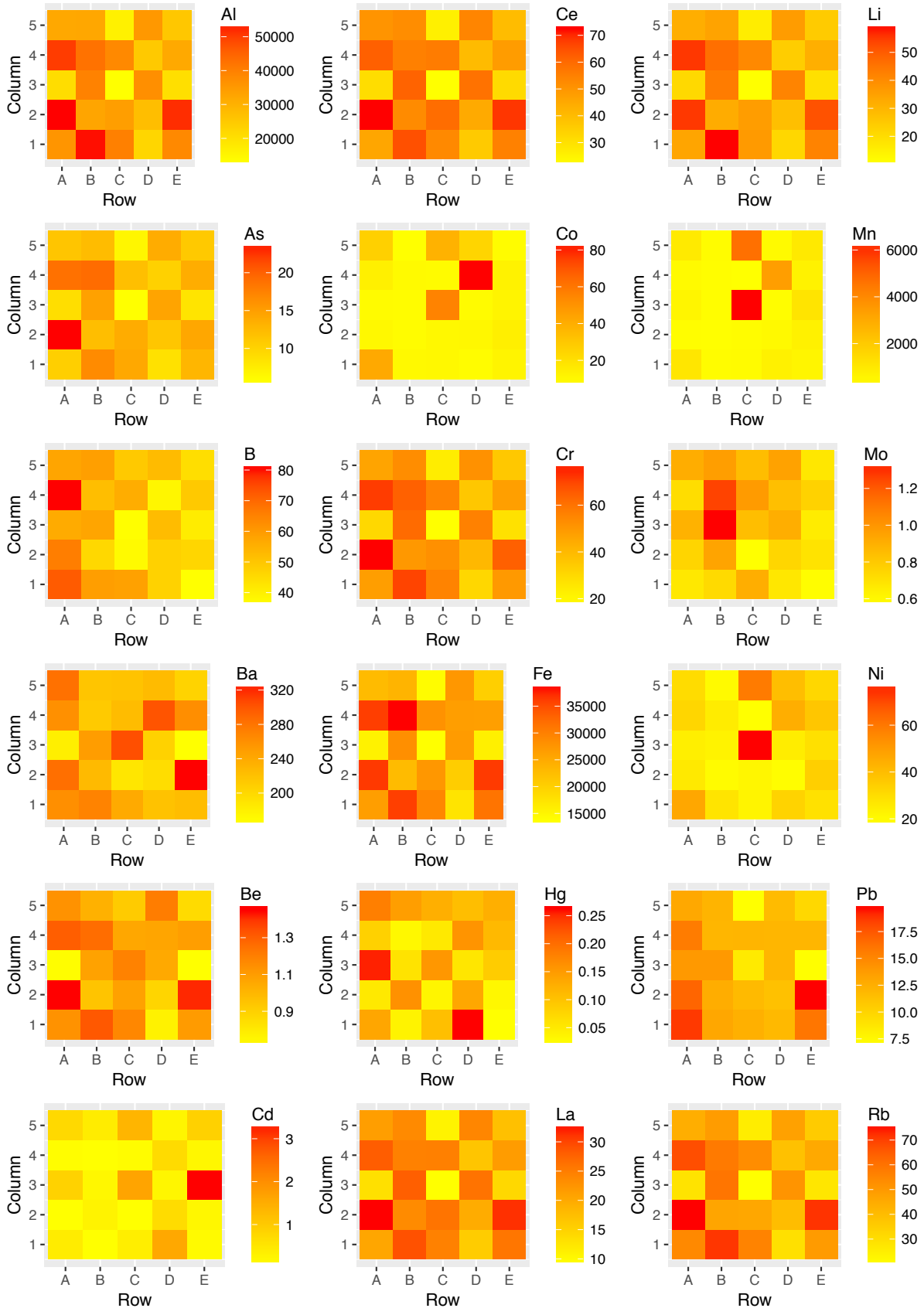


Figure A.7. Mean concentrations (µg/g) of Al, As, B, Ba, Be, Cd, Ce, Co, Cr, Fe, Hg, La, Li, Mn, Mo, Ni, Pb, Rb, S, Sb, Se, Sm, Th, Tl, U, V, Y, Yb, Zn, and Zr in insects. Rows and columns are consistent with the cells in figure 1. Data below LOD is replaced with LOD*DF.


Al
50000
40000
30000
20000
Ce
70
60
50
40
30
Li
50
40
30
20
As
20
15
10
Co
80
60
40
20
Mn
6000
4000
2000
B
80
70
60
50
40
Cr
60
40
20
Mo
1.2
1.0
0.8
0.6
Ba
320
280
240
200
Fe
35000
30000
25000
20000
15000
Ni
60
40
20
Be
1.3
1.1
0.9
Hg
0.25
0.20
0.15
0.10
0.05
Pb
17.5
15.0
12.5
10.0
7.5
Cd
3
2
1
La
30
25
20
15
10
Rb
70
60
50
40
30
Column
Row
A B C D E
1 2 3 4 5

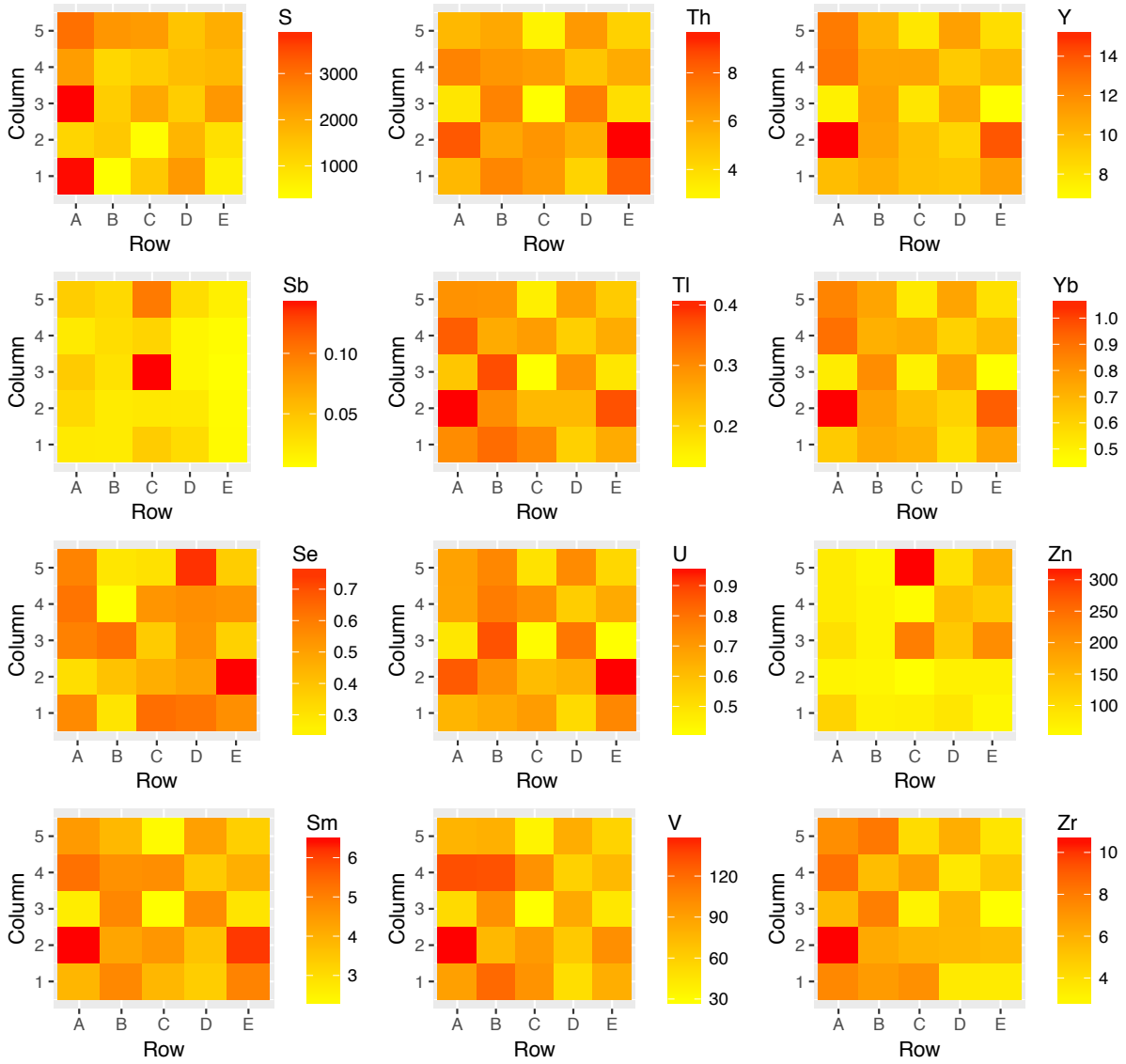
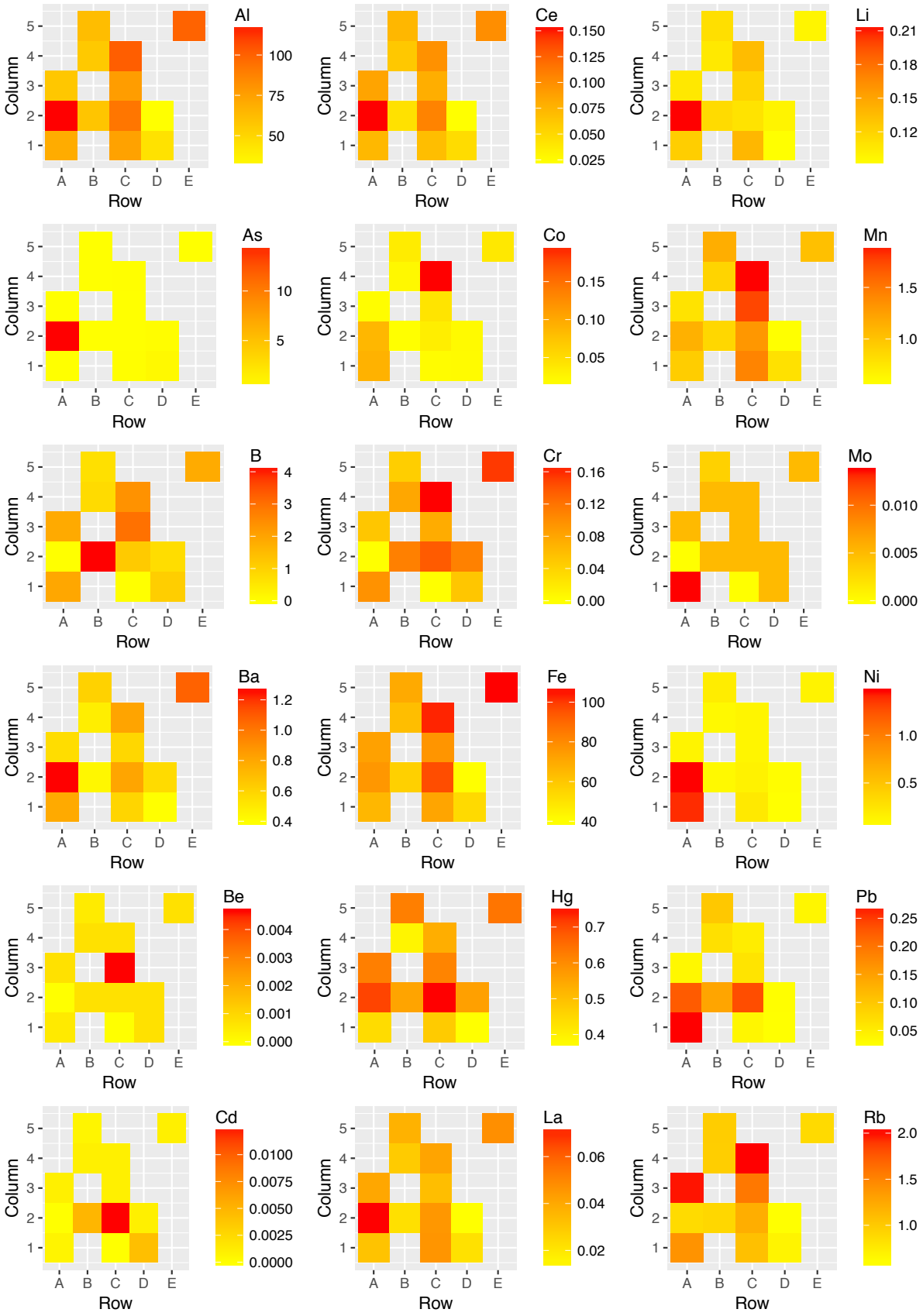


Figure A.8. Mean concentrations (µg/g) of Al, As, B, Ba, Be, Cd, Ce, Co, Cr, Fe, Hg, La, Li, Mn, Mo, Ni, Pb, Rb, S, Sb, Se, Sm, Th, Tl, U, V, Y, Yb, Zn, and Zr in spiders. Rows and columns are consistent with the cells in figure 1. Data below LOD is replaced with LOD*DF.


Al
100
75
50
Ce
0.150
0.125
0.100
0.075
0.050
0.025
Li
0.21
0.18
0.15
0.12
As
10
5
Co
0.15
0.10
0.05
Mn
1.5
1.0
B
4
3
2
1
0
Cr
0.16
0.12
0.08
0.04
0.00
Mo
0.010
0.005
0.000
Ba
1.2
1.0
0.8
0.6
0.4
Fe
100
80
60
40
Ni
1.0
0.5
Be
0.004
0.003
0.002
0.001
0.000
Hg
0.7
0.6
0.5
0.4
Pb
0.25
0.20
0.15
0.10
0.05
Cd
0.0100
0.0075
0.0050
0.0025
0.0000
La
0.06
0.04
0.02
Rb
2.0
1.5
1.0
Column
Row
1
2
3
4
5
A
B
C
D
E

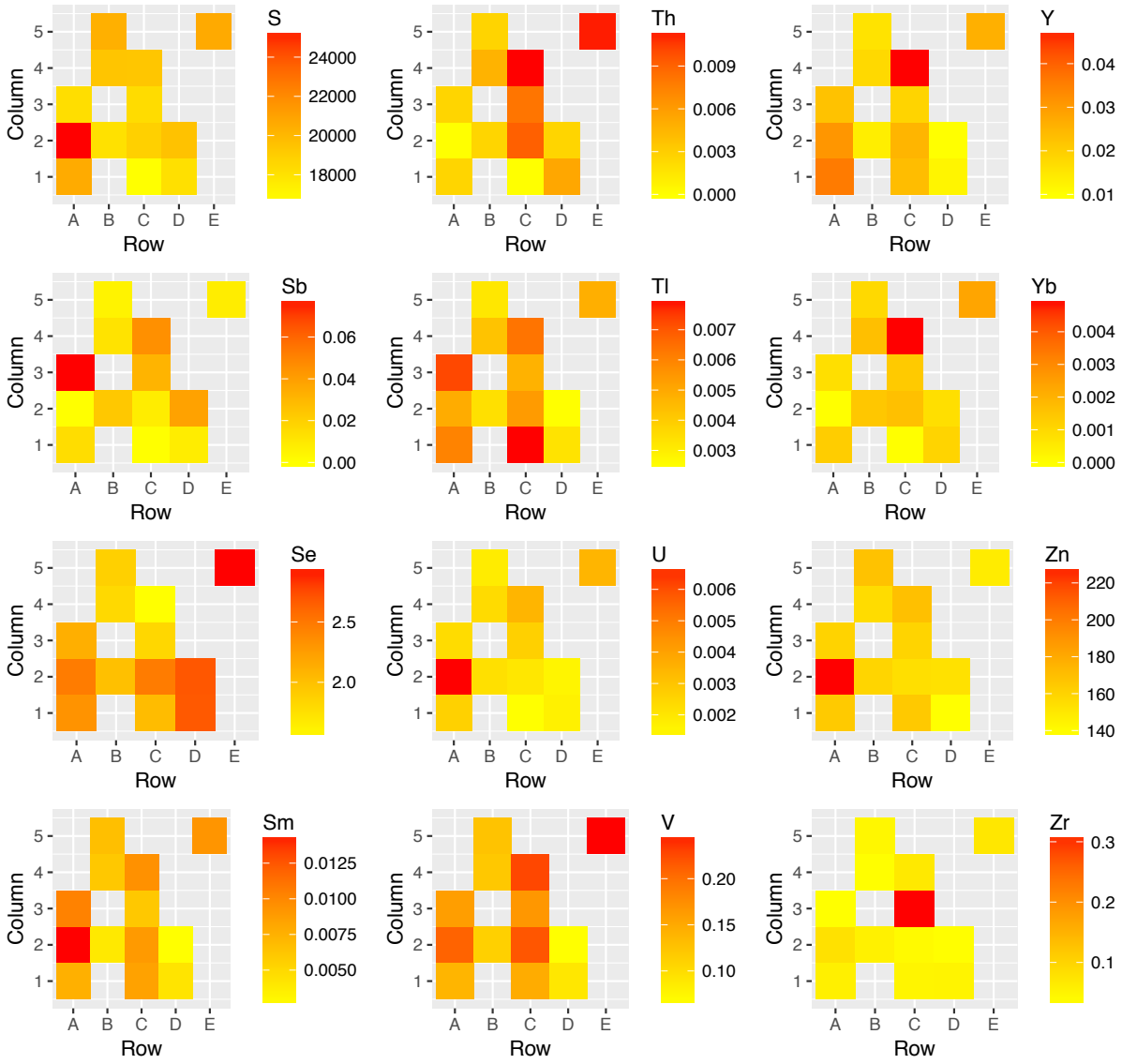
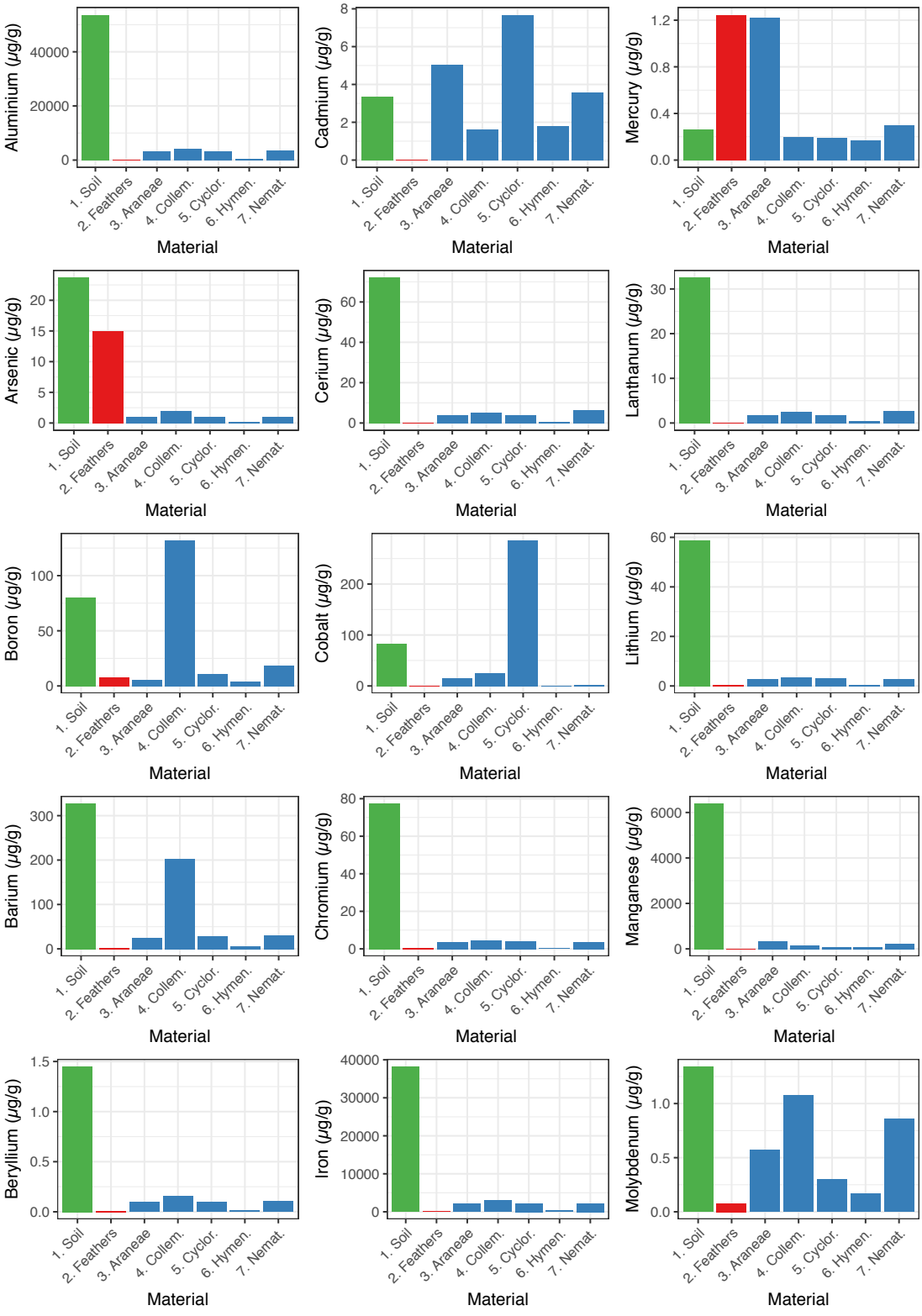


Figure A.9. Mean concentrations (µg/g) of Al, As, B, Ba, Be, Cd, Ce, Co, Cr, Fe, Hg, La, Li, Mn, Mo, Ni, Pb, Rb, S, Sb, Se, Sm, Th, Tl, U, V, Y, Yb, Zn, and Zr in feathers. Rows and columns are consistent with the cells in figure 1. Data below LOD is replaced with LOD*DF.

A.4 Bar charts



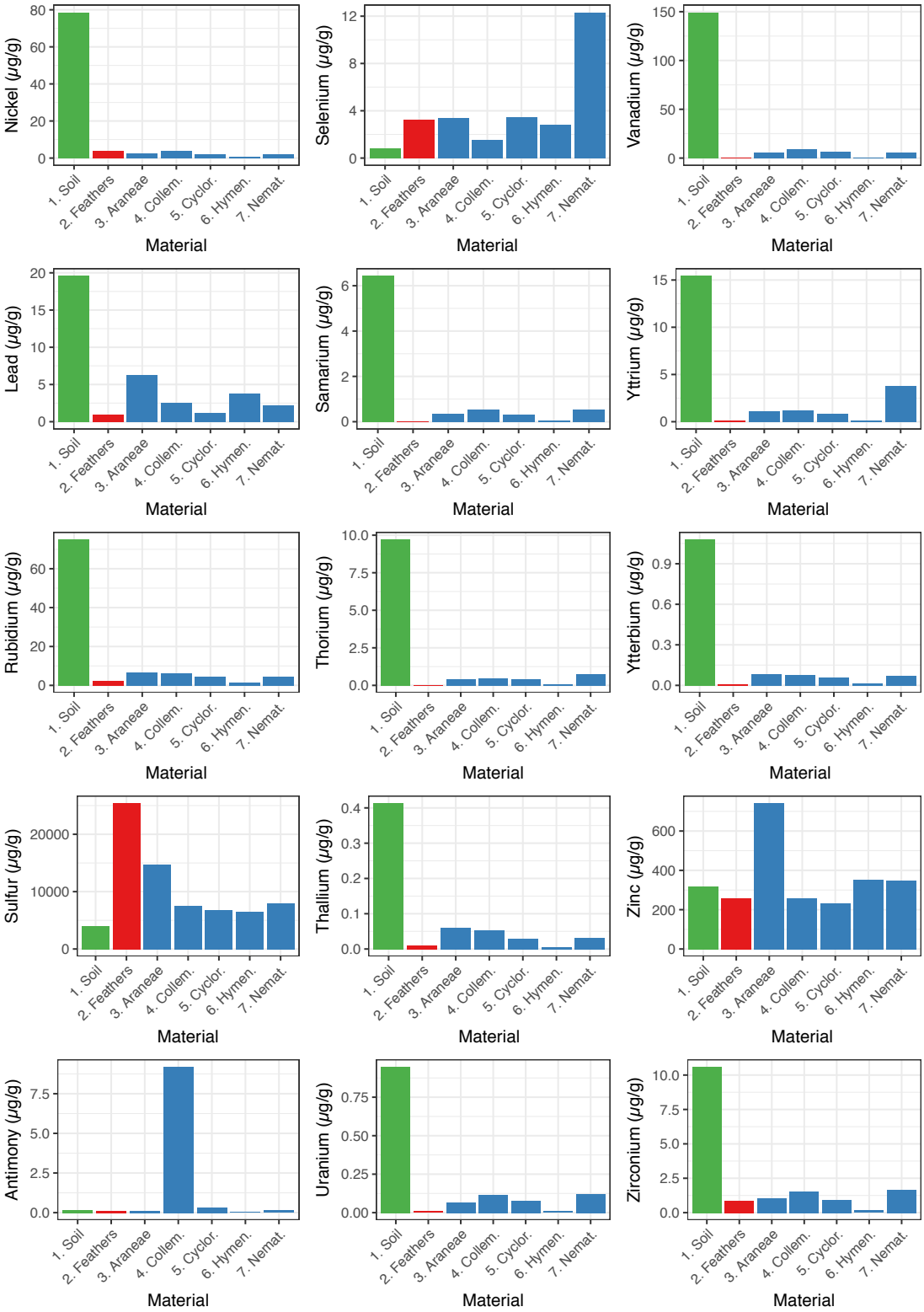


Figure A.10. Bar charts showing the accumulation of all investigated metals in different sample materials. All numbers are given in µg/g. Data below LOD is replaced with LOD*DF.

A.5 Detailed results

Table A.11. Concentrations (µg/g, dw) of the 30 analysed chemical elements in soil in the 25 cells resulting from HR-ICP-MS.

Cell	1	2	3	4	5	6	7	8	9	10	11	12	13
Al	25601.16	32015.77	20333.94	51635.21	38302.85	22354.75	27615.18	37138.60	25191.70	35445.98	16420.98	38278.17	13266.39
As	11.06	13.71	8.47	14.24	12.88	8.75	11.39	14.55	10.33	13.82	6.66	11.99	5.78
B	44.56	49.54	41.31	46.53	36.62	47.38	47.59	52.48	38.85	52.69	49.53	55.99	36.96
Ba	204.80	262.21	166.30	327.33	223.94	220.16	196.36	206.42	301.58	225.92	219.02	225.27	304.35
Be	0.86	1.10	0.71	1.42	1.11	0.77	0.88	1.06	1.07	1.21	0.92	1.06	1.20
Cd	0.88	0.19	3.34	0.19	0.15	1.60	0.69	0.23	0.70	0.24	1.37	0.11	1.66
Ce	40.26	48.48	32.12	68.51	54.84	36.57	44.96	58.22	40.54	54.06	26.11	56.07	22.02
Co	8.49	12.57	11.72	14.00	11.98	8.66	10.57	8.99	82.55	24.64	38.63	9.00	56.08
Cr	35.09	47.88	26.82	65.00	49.86	30.17	40.09	56.68	35.48	52.25	22.86	55.74	16.87
Fe	20306.64	26427.27	15962.45	36302.77	31345.83	17596.76	20633.40	26925.85	26581.01	27391.27	14342.33	27963.84	13998.42
Hg	0.13	0.11	0.09	0.04	0.02	0.26	0.14	0.06	0.16	0.11	0.13	0.05	0.15
La	17.36	21.69	14.47	31.19	25.75	15.94	19.75	26.05	16.80	24.11	10.99	24.78	9.48
Li	24.77	31.82	19.44	51.45	42.03	21.74	27.90	41.86	24.30	36.41	15.89	40.63	11.31
Mn	923.20	641.37	1104.50	676.69	541.80	659.46	399.71	269.16	3393.74	352.64	4737.34	185.46	6393.36
Mo	0.67	0.76	0.65	0.69	0.58	0.67	0.74	0.91	0.84	0.96	0.85	0.98	0.85
Ni	31.54	37.19	28.60	34.10	28.22	33.04	19.14	24.48	44.74	39.70	59.58	18.34	78.64
Pb	9.81	12.09	7.26	19.58	15.95	11.87	11.34	12.06	12.11	11.77	7.18	12.15	8.61
Rb	36.05	45.79	28.05	71.44	49.59	29.92	40.02	51.48	38.98	48.04	25.97	53.34	20.53
S	1921.39	1734.29	2355.15	940.80	640.48	2308.12	1786.69	1328.54	1639.78	1504.97	2274.53	1346.42	2032.49
Sb	0.02	0.01	0.01	0.01	0.01	0.03	0.02	0.01	0.01	0.03	0.10	0.04	0.14
Se	0.37	0.54	0.35	0.79	0.55	0.62	0.50	0.55	0.55	0.76	0.30	0.53	0.37
Sm	3.33	4.05	2.85	6.11	4.95	3.39	3.60	4.77	3.45	4.38	2.35	4.74	2.24
Th	4.36	5.81	3.86	9.71	8.30	4.29	5.66	7.36	4.81	6.43	3.02	6.28	2.56
Tl	0.21	0.26	0.17	0.37	0.26	0.20	0.24	0.29	0.21	0.28	0.16	0.28	0.13
U	0.53	0.66	0.41	0.95	0.75	0.52	0.64	0.79	0.56	0.74	0.50	0.73	0.43
V	58.53	74.73	44.89	100.21	81.49	50.90	64.51	84.75	59.65	82.10	37.04	98.72	29.30
Y	8.31	10.17	6.69	13.99	11.12	9.44	8.75	10.91	9.24	11.12	7.87	10.97	7.87
Yb	0.55	0.69	0.45	0.96	0.76	0.56	0.60	0.77	0.61	0.76	0.52	0.74	0.50
Zn	164.22	128.31	211.62	71.47	62.94	88.42	71.90	130.44	147.25	96.31	317.13	55.45	229.62
Zr	3.71	5.07	2.58	5.56	3.46	3.50	5.69	5.75	3.60	6.06	4.10	6.76	3.11

Cell	14	15	16	17	18	19	20	21	22	23	24	25
Al	34160.48	40053.27	53346.07	32762.03	39722.98	42610.92	32432.82	32099.60	50241.94	20764.71	53697.54	36039.87
As	13.83	14.16	16.74	12.07	14.83	19.18	12.32	11.47	18.83	9.07	23.69	10.48
B	37.93	58.54	59.14	45.93	57.78	52.06	59.01	57.60	80.26	56.26	65.90	72.28
Ba	188.65	239.85	270.71	226.94	250.28	212.75	216.95	283.35	260.99	180.24	284.99	261.66
Be	1.09	1.18	1.32	0.95	1.08	1.26	1.02	1.14	1.31	0.72	1.45	1.14
Cd	0.06	0.36	0.06	0.27	0.20	0.06	0.41	0.74	0.09	0.87	0.06	0.41
Ce	59.21	53.07	64.74	52.47	61.27	55.20	52.21	50.51	61.80	31.37	72.05	45.95
Co	10.35	10.09	9.94	8.66	8.46	9.44	7.27	26.73	13.43	8.99	10.38	42.22
Cr	52.39	56.18	70.93	49.91	62.43	64.83	53.16	46.47	72.15	29.75	77.42	48.20
Fe	27447.32	29241.74	35931.53	22923.29	28369.29	38247.21	23793.31	22957.89	36055.38	15697.37	36320.68	26529.60
Hg	0.04	0.11	0.04	0.16	0.06	0.03	0.15	0.18	0.08	0.25	0.05	0.14
La	25.94	24.68	29.09	23.53	27.97	24.64	23.66	21.48	28.10	13.40	32.53	20.51
Li	36.08	36.78	58.69	32.41	43.45	46.12	34.76	32.06	55.21	21.23	55.06	34.24
Mn	273.84	328.67	272.63	209.07	265.56	256.31	286.71	894.86	363.80	520.99	239.57	1032.85
Mo	0.57	0.91	0.73	0.95	1.34	1.26	0.97	0.91	0.72	0.89	0.75	0.67
Ni	21.29	22.14	27.30	19.73	22.30	24.72	20.09	29.85	31.83	23.82	25.77	46.92
Pb	11.85	12.48	13.01	12.67	13.90	12.05	12.21	12.98	15.59	13.90	16.80	18.60
Rb	46.14	55.91	71.10	46.90	59.11	57.98	49.27	44.24	67.45	29.03	74.97	54.41
S	347.61	1415.30	315.06	1417.73	1318.07	1112.81	2359.21	3005.75	2237.67	3992.18	1165.67	3970.03
Sb	0.02	0.04	0.02	0.02	0.03	0.03	0.04	0.04	0.02	0.05	0.03	0.02
Se	0.46	0.64	0.29	0.40	0.63	0.20	0.28	0.59	0.63	0.59	0.31	0.57
Sm	4.57	3.86	4.86	4.26	4.88	4.69	3.84	4.49	5.29	2.65	6.43	3.89
Th	6.58	6.45	7.10	5.93	7.21	6.53	5.91	5.30	7.20	3.53	8.46	5.33
Tl	0.24	0.31	0.34	0.30	0.37	0.26	0.29	0.29	0.36	0.22	0.41	0.30
U	0.61	0.70	0.66	0.72	0.87	0.78	0.75	0.68	0.68	0.48	0.85	0.63
V	93.78	97.84	120.83	75.82	99.81	131.22	80.82	78.25	132.22	53.64	149.37	90.38
Y	9.71	9.66	10.44	10.95	11.13	10.92	10.25	12.68	12.88	7.39	15.43	9.82
Yb	0.67	0.71	0.74	0.76	0.83	0.72	0.76	0.85	0.91	0.51	1.08	0.63
Zn	51.24	74.69	71.24	65.21	69.51	68.59	63.78	80.16	80.70	96.17	66.76	115.65
Zr	5.84	7.26	6.85	6.30	7.93	5.44	8.17	7.35	8.44	5.61	10.59	7.59

Table A.12. Concentrations (µg/g, dw) of the 30 analysed chemical elements in insects in the 25 cells resulting from HR-ICP-MS.

Cell	1	2	3	4	5	6	7	8	9	10	11	12	13
Al	528.13	698.44	975.87	2520.21	2671.20	620.39	1199.02	6781.94	194.16	455.81	76.43	334.90	638.22
As	0.29	0.23	0.54	0.84	0.96	0.21	1.12	1.99	0.14	0.14	0.06	0.10	0.58
B	12.69	17.49	136.75	8.84	5.81	4.23	10.20	18.62	9.81	13.09	6.43	11.61	9.91
Ba	11.39	12.38	204.82	21.62	26.66	6.99	17.16	57.48	4.94	9.26	1.80	6.82	18.13
Be	0.02	0.01	0.05	0.09	0.11	0.02	0.03	0.20	0.00	0.01	0.00	0.01	0.03
Cd	5.96	5.31	8.58	4.11	3.45	6.91	10.48	8.49	3.32	6.14	5.17	6.71	6.72
Ce	0.56	1.05	0.89	2.98	3.52	0.78	1.20	7.56	0.21	0.53	0.08	0.42	0.65
Co	0.85	1.08	1.00	1.15	1.35	0.23	0.60	1.71	0.66	0.97	0.38	0.98	1.01
Cr	0.75	0.52	1.04	2.67	3.04	0.65	1.04	7.42	0.15	0.21	0.11	0.17	0.59
Fe	797.39	859.65	838.53	1850.52	1945.59	522.47	1169.81	4342.15	415.40	639.55	219.30	619.04	1223.47
Hg	0.21	0.42	0.14	0.20	0.20	0.12	0.34	0.22	0.31	0.17	0.12	0.24	0.22
La	0.25	0.52	0.40	1.33	1.60	0.38	0.57	3.36	0.10	0.24	0.04	0.19	0.28
Li	0.52	0.68	0.98	2.08	2.22	0.54	1.50	5.75	0.32	0.50	0.08	0.49	0.52
Mn	279.90	251.76	110.79	71.05	133.61	25.10	102.73	141.55	108.68	180.88	35.95	68.66	246.12
Mo	0.58	0.58	0.20	0.70	0.74	0.24	1.49	0.80	0.57	0.67	0.29	0.90	0.64
Ni	1.39	2.50	1.18	2.05	2.50	0.95	1.49	4.41	0.52	2.46	0.21	1.50	1.67
Pb	1.17	4.05	0.55	0.77	0.97	0.24	0.50	2.23	0.13	2.32	0.05	0.33	0.49
Rb	2.78	4.39	2.72	4.38	4.58	0.99	3.87	8.97	1.60	1.78	0.56	0.98	2.36
S	10955.37	14344.10	8210.80	12094.83	10814.70	6132.68	16897.31	10900.22	12229.36	11950.02	6328.24	12857.59	11421.52
Sb	0.09	0.07	9.19	<LOD	0.02	<LOD	<LOD	0.02	0.00	0.00	0.00	0.00	<LOD
Se	4.95	4.18	2.58	5.25	4.11	2.10	4.52	4.08	4.26	4.02	1.51	5.12	6.50
Sm	0.05	0.09	0.10	0.25	0.30	0.09	0.12	0.66	0.03	0.05	0.01	0.06	0.07
Th	0.07	0.10	0.08	0.35	0.46	0.08	0.16	0.99	0.06	0.07	0.01	0.07	0.09
Tl	0.01	0.01	0.02	0.03	0.05	0.01	0.02	0.06	0.01	0.01	0.00	0.01	0.02
U	0.01	0.02	0.02	0.06	0.07	0.02	0.03	0.18	0.01	0.01	0.00	0.02	0.02
V	1.26	1.66	1.75	4.43	5.30	1.17	2.34	12.52	0.45	1.01	0.18	0.73	1.25
Y	0.17	0.24	0.26	0.59	0.79	0.39	0.41	1.72	0.05	0.16	0.02	0.24	0.24
Yb	0.01	0.02	0.01	0.05	0.05	0.02	0.02	0.13	0.00	0.01	0.00	0.02	0.02
Zn	397.30	493.92	400.21	257.00	262.18	160.46	510.45	348.22	304.23	427.03	191.02	398.84	447.57
Zr	0.23	0.39	0.66	0.87	1.23	0.21	0.48	2.59	0.32	0.31	0.03	0.18	0.35

Cell	14	15	16	17	18	19	20	21	22	23	24	25
Al	4401.25	928.39	42.54	186.02	3311.58	1264.00	797.74	489.72	490.90	916.32	1304.79	318.82
As	2.04	<LOD	<LOD	<LOD	1.87	1.29	0.17	0.38	<LOD	1.02	0.40	0.23
B	17.06	10.48	7.19	3.76	10.11	8.29	5.18	3.97	5.70	7.95	28.90	13.55
Ba	57.15	12.00	2.98	4.21	38.59	14.76	13.47	14.92	10.67	16.93	16.42	20.24
Be	0.16	0.05	<LOD	<LOD	0.11	0.07	0.01	0.04	<LOD	0.01	0.03	<LOD
Cd	6.12	8.67	5.50	4.82	7.83	3.21	6.67	5.84	7.54	6.94	6.59	8.69
Ce	5.16	1.20	0.05	0.20	8.88	4.76	0.92	0.97	0.58	0.90	1.05	0.29
Co	1.60	1.79	0.11	0.18	1.70	1.02	0.89	2.60	24.43	0.57	0.96	285.52
Cr	4.57	1.15	<LOD	<LOD	3.60	1.37	0.24	<LOD	<LOD	0.98	1.41	0.25
Fe	3154.93	965.79	151.41	248.05	2667.12	1173.24	873.17	626.46	760.19	1174.41	1142.01	633.33
Hg	0.28	0.41	0.07	0.17	0.26	0.18	0.41	0.31	0.38	0.29	0.11	0.26
La	1.91	0.93	0.02	0.11	3.92	2.62	0.43	0.38	0.31	0.45	0.50	0.17
Li	3.73	1.05	0.27	0.27	2.95	1.40	0.79	0.64	0.76	1.16	1.07	0.60
Mn	160.28	91.53	18.18	24.74	206.64	55.67	293.66	246.51	80.52	58.30	171.10	79.55
Mo	1.31	1.12	0.20	<LOD	1.23	1.07	0.27	0.95	1.28	1.30	0.71	0.14
Ni	4.18	2.21	0.21	0.71	5.12	2.55	1.06	1.16	1.52	1.84	1.11	1.34
Pb	1.77	2.54	0.05	0.52	4.43	1.28	0.38	0.29	0.32	0.62	0.48	0.29
Rb	6.58	1.96	0.50	1.81	6.94	2.97	1.78	2.30	1.92	2.38	2.56	2.60
S	10752.85	13228.31	5316.36	6810.06	15364.27	13619.33	17691.83	17009.69	21444.29	21732.13	12456.05	17385.48
Sb	0.10	0.16	<LOD	<LOD	<LOD	0.00	0.01	0.18	<LOD	0.05	<LOD	0.30
Se	4.07	14.47	1.49	2.97	4.13	3.64	4.86	2.90	9.32	5.35	2.62	5.83
Sm	0.54	0.13	0.01	0.01	0.81	0.40	0.09	0.08	0.06	0.09	0.10	0.03
Th	0.46	0.12	0.01	0.04	1.12	0.25	0.12	0.04	0.05	0.07	0.12	0.01
Tl	0.06	0.01	0.00	0.00	0.05	0.02	0.01	0.02	0.02	0.02	0.01	0.01
U	0.12	0.03	0.00	0.01	0.19	0.05	0.02	0.02	0.01	0.06	0.02	0.01
V	8.95	2.15	0.12	0.36	6.73	2.79	1.61	1.17	1.17	2.00	2.45	0.76
Y	1.25	0.33	0.02	0.06	4.56	0.49	0.25	0.12	0.13	0.26	0.24	0.12
Yb	0.08	0.03	<LOD	0.01	0.09	0.03	0.03	0.01	0.00	0.02	0.02	0.00
Zn	414.10	545.20	174.18	197.93	400.59	354.86	657.04	627.58	771.29	628.61	397.24	661.61
Zr	1.58	0.47	0.02	0.17	1.36	0.45	0.53	1.10	0.40	0.42	0.55	1.03

<LOD: Value below LOD

Table A.13. Concentrations (µg/g, dw) of the 30 analysed chemical elements in spiders in the 25 cells resulting from HR-ICP-MS.

Cell	1	2	3	4	5	6	7	8	9	10	11	12	13
Al	715.08	255.70	90.30	NA	2613.10	789.50	47.02	3383.08	36.41	75.51	28.70	NA	216.46
As	0.75	0.71	0.28	NA	0.91	0.59	<LOD	0.99	<LOD	0.66	<LOD	NA	0.39
B	2.82	4.95	1.30	NA	2.99	1.41	<LOD	5.00	<LOD	1.36	<LOD	NA	1.71
Ba	11.84	7.66	2.69	NA	20.64	13.19	2.27	24.23	2.93	3.67	1.52	NA	6.75
Be	0.03	<LOD	0.01	NA	0.09	0.03	<LOD	0.09	<LOD	0.01	<LOD	NA	0.01
Cd	1.07	1.96	1.03	NA	1.51	3.70	5.05	0.86	1.10	3.20	1.48	NA	1.58
Ce	1.12	0.30	0.66	NA	3.73	1.10	0.06	2.57	0.07	0.15	0.06	NA	0.30
Co	0.52	0.76	0.30	NA	0.79	0.34	0.09	0.68	0.38	0.44	0.39	NA	0.67
Cr	0.97	<LOD	<LOD	NA	3.49	0.86	<LOD	2.76	<LOD	0.11	<LOD	NA	2.03
Fe	838.48	688.18	284.44	NA	1993.95	723.80	288.36	2108.49	199.24	319.67	271.46	NA	575.52
Hg	0.47	1.22	0.20	NA	0.15	0.34	0.18	0.21	0.39	0.29	0.36	NA	0.24
La	0.48	0.40	0.28	NA	1.67	0.43	<LOD	1.14	<LOD	0.06	0.03	NA	0.14
Li	0.70	0.49	0.20	NA	2.30	0.67	0.20	2.79	0.16	0.18	0.07	NA	0.20
Mn	305.30	195.98	82.82	NA	51.02	86.03	16.84	53.14	68.27	108.21	79.90	NA	89.71
Mo	0.25	0.57	0.16	NA	0.30	0.25	<LOD	<LOD	<LOD	0.29	0.42	NA	0.48
Ni	1.56	2.21	0.97	NA	2.00	1.80	0.42	2.30	0.84	1.53	0.61	NA	2.44
Pb	0.29	1.08	0.07	NA	0.89	0.43	0.06	0.93	0.07	0.26	6.33	NA	0.45
Rb	4.20	4.78	2.14	NA	3.85	1.73	1.01	6.58	0.98	1.28	0.43	NA	0.84
S	7411.42	14643.68	5946.12	NA	6511.61	7681.64	8273.90	7668.07	7200.61	7954.75	7830.21	NA	6148.09
Sb	0.03	0.09	<LOD	NA	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	NA	<LOD
Se	1.86	3.39	1.16	NA	3.02	2.26	2.37	1.19	1.52	3.03	1.34	NA	1.68
Sm	0.10	0.04	0.07	NA	0.35	0.15	0.01	0.20	0.01	0.02	0.01	NA	0.04
Th	0.10	0.04	0.15	NA	0.41	0.11	<LOD	0.26	0.03	0.03	<LOD	NA	0.04
Tl	0.03	0.06	0.01	NA	0.04	0.03	0.01	0.04	0.01	0.03	0.02	NA	0.03
U	0.02	0.02	0.01	NA	0.07	0.03	<LOD	0.07	0.01	0.02	0.00	NA	0.02
V	1.53	0.52	0.24	NA	5.52	1.54	0.12	5.71	0.11	0.20	0.12	NA	0.52
Y	0.23	0.13	0.08	NA	1.08	0.51	0.03	0.64	0.05	0.08	0.03	NA	0.15
Yb	0.02	0.01	0.00	NA	0.08	0.03	<LOD	0.05	<LOD	0.01	<LOD	NA	0.01
Zn	290.78	741.21	288.04	NA	254.50	329.13	242.71	245.44	287.74	393.73	297.27	NA	341.48
Zr	0.24	0.13	0.04	NA	0.78	0.34	0.06	1.02	0.13	0.06	0.13	NA	0.13

Cell	14	15	16	17	18	19	20	21	22	23	24	25
Al	154.84	126.34	68.88	58.41	1075.15	130.73	137.07	26.22	NA	63.29	105.68	21.40
As	0.33	0.30	<LOD	0.47	0.80	<LOD	0.34	<LOD	NA	<LOD	<LOD	0.25
B	1.08	0.87	<LOD	0.87	2.00	<LOD	<LOD	<LOD	NA	<LOD	<LOD	0.63
Ba	5.03	5.76	3.85	4.99	13.62	2.66	4.28	2.69	NA	2.32	2.60	1.37
Be	<LOD	0.01	<LOD	0.01	0.05	<LOD	<LOD	<LOD	NA	<LOD	<LOD	<LOD
Cd	3.11	2.99	2.42	2.28	2.51	1.56	1.86	2.21	NA	2.49	1.87	1.31
Ce	0.24	0.19	0.07	0.14	1.52	0.63	0.18	0.04	NA	0.07	0.12	0.03
Co	0.29	0.25	8.68	0.24	0.54	0.33	0.54	0.68	NA	<LOD	15.25	0.33
Cr	0.17	0.13	0.93	<LOD	1.13	<LOD	<LOD	<LOD	NA	<LOD	<LOD	<LOD
Fe	312.01	264.73	165.40	304.53	984.81	246.77	263.94	177.37	NA	204.16	206.98	161.50
Hg	0.39	0.26	0.31	0.46	0.24	0.18	0.14	0.22	NA	0.27	0.17	0.19
La	0.10	0.08	0.49	0.06	0.82	0.27	0.20	<LOD	NA	0.03	0.07	0.01
Li	0.20	0.22	0.19	0.16	0.89	0.17	0.21	0.10	NA	0.15	0.33	0.08
Mn	57.57	65.37	32.24	63.95	37.20	24.20	45.15	39.69	NA	18.09	25.81	24.80
Mo	0.28	0.21	<LOD	0.24	0.32	<LOD	<LOD	0.31	NA	<LOD	0.29	0.25
Ni	0.71	0.70	1.12	0.71	2.12	1.00	1.01	0.73	NA	0.52	0.73	0.51
Pb	0.30	0.30	0.29	0.25	1.26	1.42	0.31	0.06	NA	0.06	0.13	0.04
Rb	0.56	1.08	0.68	1.69	2.42	0.39	0.71	0.63	NA	0.43	1.07	2.30
S	7180.98	10036.30	7166.00	7934.49	7054.47	6696.83	8146.49	7535.02	NA	8750.66	9657.12	7353.11
Sb	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	NA	<LOD	<LOD	<LOD
Se	2.19	2.14	1.96	2.52	2.28	2.09	2.00	1.47	NA	2.54	1.55	1.81
Sm	0.03	0.03	0.01	0.03	0.15	0.05	0.02	<LOD	NA	0.01	0.02	0.00
Th	0.03	0.02	<LOD	0.03	0.14	0.06	0.03	<LOD	NA	<LOD	<LOD	<LOD
Tl	0.03	0.02	0.02	0.02	0.03	0.03	0.02	0.01	NA	0.02	0.01	0.02
U	0.01	0.01	0.01	0.01	0.04	0.01	0.01	<LOD	NA	0.01	0.00	0.00
V	0.34	0.26	0.14	0.14	2.19	0.30	0.32	0.08	NA	0.16	0.25	0.06
Y	0.08	0.07	0.03	0.08	0.41	0.09	0.08	0.02	NA	0.14	0.09	0.01
Yb	0.01	0.00	<LOD	0.00	0.03	<LOD	<LOD	<LOD	NA	0.01	<LOD	<LOD
Zn	339.61	390.90	374.15	411.78	281.41	308.91	339.44	307.95	NA	303.32	274.40	303.58
Zr	0.80	0.05	0.07	0.05	0.34	0.13	0.12	0.03	NA	0.05	0.26	0.02

NA: Not available, <LOD: Value below LOD

Table A.14. Concentrations (μg/g, dw) of the 30 analysed chemical elements in feathers in the 25 cells resulting from HR-ICP-MS.

Cell	1	2	3	4	5	6	7	8	9	10	11	12	13
Al	99.82	NA	NA	NA	NA	88.65	30.25	NA	NA	48.92	NA	101.47	229.90
As	<LOD	NA	NA	NA	NA	1.04	0.20	NA	NA	<LOD	NA	0.08	<LOD
B	1.84	NA	NA	NA	NA	2.17	0.77	NA	NA	0.66	NA	2.36	8.45
Ba	1.08	NA	NA	NA	NA	0.80	0.57	NA	NA	0.39	NA	0.82	1.76
Be	<LOD	NA	NA	NA	NA	<LOD	<LOD	NA	NA	<LOD	NA	<LOD	0.01
Cd	<LOD	NA	NA	NA	NA	0.01	<LOD	NA	NA	<LOD	NA	<LOD	<LOD
Ce	0.10	NA	NA	NA	NA	0.10	0.02	NA	NA	0.05	NA	0.10	0.24
Co	0.04	NA	NA	NA	NA	0.04	0.02	NA	NA	0.02	NA	0.20	0.13
Cr	0.15	NA	NA	NA	NA	<LOD	0.11	NA	NA	<LOD	NA	0.17	0.12
Fe	106.05	NA	NA	NA	NA	105.92	39.50	NA	NA	49.51	NA	103.74	231.01
Hg	0.64	NA	NA	NA	NA	0.74	0.56	NA	NA	0.42	NA	0.54	1.84
La	0.05	NA	NA	NA	NA	0.04	0.01	NA	NA	0.02	NA	0.04	0.10
Li	0.10	NA	NA	NA	NA	0.18	0.10	NA	NA	0.10	NA	0.14	0.36
Mn	1.04	NA	NA	NA	NA	1.61	0.59	NA	NA	0.77	NA	1.89	5.23
Mo	<LOD	NA	NA	NA	NA	<LOD	<LOD	NA	NA	<LOD	NA	<LOD	<LOD
Ni	0.16	NA	NA	NA	NA	0.12	0.08	NA	NA	0.17	NA	0.14	0.43
Pb	0.04	NA	NA	NA	NA	0.06	0.03	NA	NA	0.03	NA	0.05	0.19
Rb	0.88	NA	NA	NA	NA	1.35	0.59	NA	NA	0.73	NA	2.01	4.75
S	20650.04	NA	NA	NA	NA	36273.37	19519.65	NA	NA	21865.19	NA	19361.36	54936.97
Sb	<LOD	NA	NA	NA	NA	<LOD	0.04	NA	NA	<LOD	NA	0.05	0.09
Se	2.94	NA	NA	NA	NA	5.37	2.70	NA	NA	2.24	NA	1.50	5.46
Sm	0.01	NA	NA	NA	NA	0.01	<LOD	NA	NA	<LOD	NA	0.01	0.02
Th	0.01	NA	NA	NA	NA	0.01	<LOD	NA	NA	<LOD	NA	0.01	0.02
Tl	<LOD	NA	NA	NA	NA	0.01	<LOD	NA	NA	<LOD	NA	0.01	0.01
U	<LOD	NA	NA	NA	NA	<LOD	<LOD	NA	NA	<LOD	NA	<LOD	0.01
V	0.25	NA	NA	NA	NA	0.18	0.07	NA	NA	0.09	NA	0.23	0.50
Y	0.03	NA	NA	NA	NA	0.02	0.01	NA	NA	0.01	NA	0.05	0.06
Yb	<LOD	NA	NA	NA	NA	<LOD	<LOD	NA	NA	<LOD	NA	<LOD	<LOD
Zn	148.02	NA	NA	NA	NA	276.64	153.12	NA	NA	156.78	NA	169.34	479.72
Zr	0.07	NA	NA	NA	NA	0.10	0.03	NA	NA	0.03	NA	0.07	0.96

Cell	14	15	16	17	18	19	20	21	22	23	24	25
Al	92.73	74.47	NA	116.59	NA	113.09	322.37	NA	NA	57.22	118.78	488.07
As	<LOD	<LOD	NA	<LOD	NA	<LOD	<LOD	NA	NA	<LOD	14.92	<LOD
B	1.18	<LOD	NA	8.16	NA	1.14	2.68	NA	NA	1.86	<LOD	13.56
Ba	0.81	0.60	NA	0.87	NA	0.95	3.28	NA	NA	0.56	1.26	5.54
Be	<LOD	<LOD	NA	<LOD	NA	<LOD	<LOD	NA	NA	<LOD	<LOD	<LOD
Cd	0.01	<LOD	NA	0.01	NA	<LOD	<LOD	NA	NA	<LOD	<LOD	<LOD
Ce	0.11	0.07	NA	0.09	NA	0.13	0.39	NA	NA	0.09	0.15	0.51
Co	0.03	<LOD	NA	0.02	NA	0.05	0.18	NA	NA	0.02	0.09	0.61
Cr	0.14	<LOD	NA	0.17	NA	0.11	0.11	NA	NA	<LOD	<LOD	0.57
Fe	97.02	71.67	NA	113.30	NA	126.39	369.55	NA	NA	72.94	76.16	457.79
Hg	0.75	0.48	NA	1.11	NA	0.79	3.31	NA	NA	0.62	0.71	3.11
La	0.05	0.05	NA	0.04	NA	0.06	0.18	NA	NA	0.04	0.07	0.19
Li	0.11	0.14	NA	0.23	NA	0.21	0.58	NA	NA	0.11	0.21	0.87
Mn	1.31	1.43	NA	1.76	NA	1.79	6.20	NA	NA	0.79	1.14	6.61
Mo	<LOD	<LOD	NA	<LOD	NA	<LOD	<LOD	NA	NA	<LOD	<LOD	0.07
Ni	0.16	0.23	NA	0.22	NA	0.22	0.84	NA	NA	0.15	1.45	9.68
Pb	0.24	0.04	NA	0.29	NA	0.14	0.58	NA	NA	0.04	0.22	1.85
Rb	1.21	1.08	NA	1.79	NA	1.93	5.16	NA	NA	1.99	0.87	9.85
S	18861.52	16619.62	NA	36118.68	NA	38763.63	100393.19	NA	NA	18231.76	25409.45	144058.87
Sb	<LOD	<LOD	NA	0.05	NA	0.02	<LOD	NA	NA	0.08	<LOD	0.09
Se	2.49	2.03	NA	3.99	NA	3.60	8.99	NA	NA	2.13	2.49	16.31
Sm	0.01	0.01	NA	0.01	NA	0.01	0.03	NA	NA	0.01	0.01	0.05
Th	0.01	<LOD	NA	<LOD	NA	0.01	0.01	NA	NA	<LOD	<LOD	0.01
Tl	0.01	0.01	NA	0.01	NA	0.01	0.01	NA	NA	0.01	<LOD	0.04
U	<LOD	<LOD	NA	<LOD	NA	<LOD	0.01	NA	NA	<LOD	0.01	0.02
V	0.22	0.15	NA	0.23	NA	0.24	0.66	NA	NA	0.16	0.21	0.97
Y	0.03	0.02	NA	0.03	NA	0.04	0.08	NA	NA	0.02	0.03	0.25
Yb	<LOD	<LOD	NA	<LOD	NA	<LOD	0.01	NA	NA	<LOD	<LOD	0.01
Zn	154.25	165.09	NA	318.67	NA	312.04	854.56	NA	NA	160.06	229.30	1153.08
Zr	0.04	0.05	NA	0.11	NA	0.07	0.23	NA	NA	0.03	0.08	0.39

NA: Not available, <LOD: Value below LOD

Table A.15. Standard deviation, mean, median, minimum and maximum concentrations for all selected metals and sample materials. N equals number of values per sample material above LOD. All concentrations are given in µg/g, dw.

Metal	N	Sample material	Mean (µg/g)	Median (µg/g)	Min. (µg/g)	Max. (µg/g)	Standard deviation
Al	28	Feathers	68.36	64.79	28.50	118.78	25.79
	78	Insects/spiders	543.15	176.53	21.40	4259.02	857.87
	25	Soil	34061.32	34160.48	13266.39	53697.54	11124.93
As	4*	Feathers	4.06	0.62	0.08	14.92	7.25
	50	Insects/spiders	0.45	0.37	0.03	1.90	0.37
	25	Soil	12.81	12.32	5.78	23.69	3.98
B	19	Feathers	2.36	1.86	0.56	7.60	1.86
	55	Insects/spiders	7.54	5.18	0.63	131.77	17.35
	25	Soil	52.11	52.06	36.62	80.26	10.74
Ba	28	Feathers	0.66	0.58	0.32	1.26	0.26
	78	Insects/spiders	9.85	4.22	0.80	202.77	23.53
	25	Soil	238.44	225.92	166.30	327.33	41.19
Be	2*	Feathers	0.01	0.01	0.01	0.01	0.00
	40	Insects/spiders	0.03	0.03	0.00	0.16	0.03
	25	Soil	1.08	1.09	0.71	1.45	0.20
Cd	3*	Feathers	0.01	0.01	0.01	0.01	0.00
	78	Insects/spiders	2.65	2.25	0.06	7.69	1.82
	25	Soil	0.60	0.27	0.06	3.34	0.74
Ce	28	Feathers	0.07	0.06	0.02	0.20	0.04
	78	Insects/spiders	0.75	0.24	0.03	6.24	1.23
	25	Soil	49.70	52.47	22.02	72.05	12.94
Co	24	Feathers	0.06	0.03	0.01	0.23	0.06
	76	Insects/spiders	4.81	0.39	0.06	285.26	32.77
	25	Soil	18.55	10.38	7.27	82.55	18.22
Cr	11*	Feathers	0.15	0.15	0.11	0.23	0.04
	44	Insects/spiders	1.01	0.76	0.08	4.33	1.08
	25	Soil	48.74	49.91	16.87	77.42	15.95
Fe	28	Feathers	69.67	67.85	35.24	109.22	21.66
	78	Insects/spiders	520.47	286.40	112.25	2947.87	541.40
	25	Soil	25971.70	26581.01	13998.42	38247.21	7242.48
Hg	28	Feathers	0.54	0.50	0.28	1.25	0.19
	78	Insects/spiders	0.17	0.13	0.03	1.22	0.15
	25	Soil	0.11	0.11	0.02	0.26	0.06
La	25	Feathers	0.04	0.04	0.01	0.10	0.02
	74	Insects/spiders	0.38	0.12	0.01	2.71	0.56
	25	Soil	22.16	23.66	9.48	32.53	6.11

Metal	N	Sample material	Mean (µg/g)	Median (µg/g)	Min. (µg/g)	Max. (µg/g)	Standard deviation
Li	28	Feathers	0.12	0.11	0.06	0.21	0.03
	78	Insects/spiders	0.53	0.26	0.07	3.53	0.69
	25	Soil	35.03	34.76	11.31	58.69	12.59
Mn	28	Feathers	1.11	1.00	0.59	3.12	0.50
	78	Insects/spiders	61.73	40.67	6.77	305.30	56.17
	25	Soil	1008.93	399.71	185.46	6393.36	1531.55
Mo	1*	Feathers	0.07	0.07	0.07	0.07	#DIV/0!
	62	Insects/spiders	0.36	0.29	0.14	1.08	0.18
	25	Soil	0.83	0.84	0.57	1.34	0.19
Ni	27	Feathers	0.50	0.16	0.05	3.76	0.90
	77	Insects/spiders	0.94	0.73	0.12	3.81	0.71
	25	Soil	31.72	28.22	18.34	78.64	13.89
Pb	27	Feathers	0.14	0.06	0.02	0.97	0.20
	78	Insects/spiders	0.54	0.24	0.04	6.33	0.92
	25	Soil	12.71	12.15	7.18	19.58	2.97
Rb	28	Feathers	1.18	1.06	0.48	2.38	0.54
	78	Insects/spiders	1.46	0.98	0.25	6.58	1.29
	25	Soil	47.83	48.04	20.53	74.97	14.71
S	28	Feathers	19691.84	19361.36	16619.62	25409.45	1892.11
	78	Insects/spiders	6317.04	6111.62	2145.78	14643.68	1593.12
	25	Soil	1778.83	1639.78	315.06	3992.18	927.93
Sb	11*	Feathers	0.04	0.03	0.02	0.08	0.02
	26*	Insects/spiders	0.40	0.02	0.00	9.19	1.79
	25	Soil	0.03	0.02	0.01	0.14	0.03
Se	28	Feathers	2.15	2.18	1.06	3.22	0.54
	74	Insects/spiders	2.16	2.04	0.72	12.33	1.38
	25	Soil	0.50	0.54	0.20	0.79	0.15
Sm	25	Feathers	0.01	0.01	0.00	0.02	0.00
	75	Insects/spiders	0.07	0.03	0.00	0.55	0.11
	25	Soil	4.16	4.26	2.24	6.43	1.05
Th	11*	Feathers	0.01	0.01	0.01	0.01	0.00
	63	Insects/spiders	0.10	0.04	0.01	0.77	0.15
	25	Soil	5.92	5.93	2.56	9.71	1.73
Tl	26	Feathers	0.00	0.00	0.00	0.01	0.00
	77	Insects/spiders	0.01	0.01	0.00	0.06	0.01
	25	Soil	0.27	0.28	0.13	0.41	0.07
U	25	Feathers	0.00	0.00	0.00	0.01	0.00
	74	Insects/spiders	0.02	0.01	0.00	0.12	0.03
	25	Soil	0.66	0.68	0.41	0.95	0.14
V	28	Feathers	0.14	0.14	0.05	0.25	0.06
	78	Insects/spiders	1.09	0.37	0.06	8.65	1.63
	25	Soil	82.83	81.49	29.30	149.37	30.12

Metal	N	Sample material	Mean (µg/g)	Median (µg/g)	Min. (µg/g)	Max. (µg/g)	Standard deviation
Y	28	Feathers	0.02	0.02	0.01	0.10	0.02
	78	Insects/spiders	0.22	0.08	0.01	3.75	0.47
	25	Soil	10.31	10.25	6.69	15.43	2.03
Yb	16	Feathers	0.00	0.00	0.00	0.00	0.00
	58	Insects/spiders	0.02	0.01	0.00	0.08	0.02
	25	Soil	0.70	0.72	0.45	1.08	0.15
Zn	28	Feathers	163.13	156.96	114.62	255.31	26.85
	78	Insects/spiders	227.90	198.04	81.71	741.21	99.33
	25	Soil	107.15	80.16	51.24	317.13	64.14
Zr	28	Feathers	0.08	0.04	0.02	0.86	0.15
	78	Insects/spiders	0.27	0.13	0.02	1.66	0.33
	25	Soil	5.85	5.75	2.58	10.59	1.95

* ≥50 % of the values were below LOD, and the corresponding metal was deleted from the dataset

A.6 Linear mixed-effect model

Table A.16. Estimate. standard error, degrees of freedom (df), t-value, and p-value demonstrating the relationship between the 25 cells in the grid. Numbers are resulting from linear mixed-effect model conducted in R, and cell 25 is used as intercept for comparison.

Cell	Estimate	Standard error	df	t-value	p-value
1	1.85e-02	5.60e-03	2.43e+03	3.30	0.000986***
2	2.37e-02	6.09e-03	2.43e+03	3.89	0.000101***
3	1.17e-02	6.09e-03	2.43e+03	1.93	0.053949
4	2.45e-02	8.98e-03	2.43e+03	2.73	0.006472**
5	7.70e-02	6.09e-03	2.43e+03	12.66	<2e-16***
6	1.34e-02	5.60e-03	2.43e+03	2.39	0.016753*
7	-1.01e-02	5.60e-03	2.43e+03	-1.80	0.071783
8	8.86e-02	6.09e-03	2.43e+03	14.56	<2e-16***
9	-1.04e-02	6.09e-03	2.43e+03	-1.71	0.087364
10	2.78e-04	6.09e-03	2.43e+03	0.046	0.963560
11	-2.74e-03	6.09e-03	2.43e+03	-0.45	0.652128
12	1.39e-02	6.93e-03	2.43e+03	2.01	0.044389*
13	1.04e-02	5.60e-03	2.43e+03	1.86	0.062909
14	2.09e-02	5.60e-03	2.43e+03	3.72	0.000202***
15	-3.22e-03	5.60e-03	2.43e+03	-0.58	0.565035
16	-3.25e-03	6.09e-03	2.43e+03	-0.53	0.593198
17	-3.41e-03	5.60e-03	2.43e+03	-0.61	0.543357
18	4.15e-02	6.09e-03	2.43e+03	6.83	1.1e-11**
19	1.30e-03	5.60e-03	2.43e+03	0.23	0.817027
20	-7.30e-03	5.60e-03	2.43e+03	-1.30	0.192543
21	-9.92e-03	6.09e-03	2.43e+03	-1.63	0.103365
22	-9.17e-03	8.98e-03	2.43e+03	-1.02	0.307577
23	-6.575e-03	5.60e-03	2.43e+03	-1.17	0.240728
24	1.28e-02	5.60e-03	2.43e+03	2.29	0.022269*
25	3.91e-02	1.033e-02	4.00e+00	3.784	0.019247*
	(Intercept)				