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Tracing the footprints of Arctic pollution: Spatial variations in toxic and essential elements in Svalbard reindeer (*Rangifer tarandus platyrhynchus*) faeces

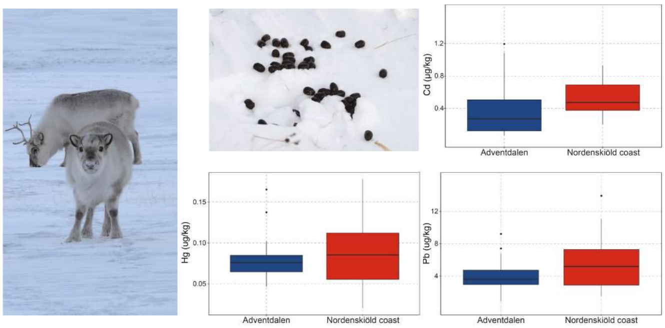
Malin Andersson Stavridis [a,b,*], Susanne Brix Røed [b], Brage Bremset Hansen [b,c], Øyvind Mikkelsen [a,d], Tomasz Maciej Ciesielski [a,b], Bjørn Munro Jenssen [a,b,*]

[a] *Department of Arctic Technology, University Centre in Svalbard (UNIS), P.O. Box 156, N-9171 Longyearbyen, Norway*
[b] *Department of Biology, Norwegian University of Science and Technology (NTNU), NO-7491 Trondheim, Norway*
[c] *Department of Terrestrial Ecology, Norwegian Institute for Nature Research (NINA), NO-7485 Trondheim, Norway*
[d] *Department of Chemistry, Norwegian University of Science and Technology (NTNU), NO-7491 Trondheim, Norway*

HIGHLIGHTS

- The elemental composition of Svalbard reindeer faeces was analysed.
- There were significant differences in several elements between populations.
- Local Arctic settlements may influence the bioavailability of toxic metals.
- Dietary differences between populations may also affect their toxic metal exposure.
- Faeces could be used to biomonitor trends in environmental pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

The Arctic is an accumulation zone of long-range transported pollution. In addition, local anthropogenic activities further contribute to regional pollution levels. The Svalbard reindeer (*Rangifer tarandus platyrhynchus*) is a suitable organism for studying and monitoring exposure to anthropogenic pollutants at the base of the terrestrial Arctic food web, and reindeer faeces have been promoted as non-invasive means of biomonitoring contaminant exposure. This study used HR-ICP-MS to analyse levels and composition of 16 elements in Svalbard reindeer faeces ($n = 96$) and soil ($n = 9$) from two locations on Svalbard, with the aim to assess whether local anthropogenic pollution influences element bioavailability. One of the sampling areas, the Nordenskiöld coast, is situated on the west coast of Spitsbergen, close to the Arctic Ocean and relatively far from local anthropogenic sources. The other sampling area, Adventdalen, is located further inland and close to Longyearbyen, the largest settlement of the archipelago. There was a significant difference in faecal elemental concentration and composition between the Adventdalen and Nordenskiöld coast reindeer populations. Elements of geogenic origin (e.g.,

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[*] Corresponding authors at: Department of Biology, Norwegian University of Science and Technology (NTNU), NO-7491 Trondheim, Norway. Department of Arctic Technology, University Centre in Svalbard (UNIS), P.O. Box 156, N-9171 Longyearbyen, Norway.
E-mail addresses: malins@unis.no (M. Andersson Stavridis), bjorn.munro.jenssen@ntnu.no (B.M. Jenssen).

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Al, Cu and Fe) were found at higher levels in faeces from Adventdalen. In comparison, levels of Ca, Se and the toxic elements Cd and Pb were higher in faecal samples from the Nordenskiöld coast. The significantly higher levels of faecal Cd and Pb at Nordenskiöld coast may be due to marine input, dietary differences between the populations, or possible anthropogenic influence from the nearby settlement of Barentsburg. There was, however, a decoupling in elemental composition between faecal and soil samples, which may derive from a selective vegetational uptake of elements from the soil. The results suggest that reindeer are exposed to a range of elements and that faeces can be used to monitor the exposure to bioavailable environmental levels of both essential and toxic elements in terrestrial ecosystems.

1. Introduction

The anthropogenic disruption of the biogeochemical cycles of several heavy metals, such as mercury (Hg), cadmium (Cd) and lead (Pb), has led to their increase in the biosphere (Amos et al., 2013; Johansson et al., 2001). Besides several natural sources (e.g., volcanic eruptions or wildfires), there are many anthropogenic contributions to the presence of these metals in the environment. Emissions, for instance, occur during fossil fuel burning, waste incineration, chemical production and mining (Pacyna and Pacyna, 2001; Raj and Maiti, 2020). Heavy metal emissions occur globally, but the main contributions derive from highly industrialised regions. The chemical properties of these metals (e.g., high volatility or association with particles) allow them to travel long distances from their source of origin to pristine areas without much local pollution input, such as the Arctic (AMAP, 2005).

Once emitted to the atmosphere, the transport of these metals takes place either in gaseous (e.g., Hg) or fine particulate form (e.g., Cd and Pb) before being deposited through wet or dry deposition (Carpi, 1997; Thorne et al., 2018). Furthermore, Hg and Cd deposition is strongly associated with marine vectors such as sea spray aerosols and fog droplets (Garbarino et al., 2002). The residence time of gaseous Hg in the atmosphere is up to 18 months, which allows for long-range atmospheric transport to areas with few local emissions (Gworek et al., 2020). In contrast, the atmospheric transportation range of particulate matter strongly depends on variables such as the size of the particle, wind conditions and precipitation patterns. Elevated levels of metals such as Cd and Pb are mainly found close to their emission sources (AMAP, 2005). However, studies have found traceable levels of these metals several hundred km from where they were first emitted, demonstrating a gradient of pollution originating from highly polluting areas. This gradient has been observed in a range of matrices, such as moss (Berg and Steinnes, 1997), air (Strizhkina et al., 2022) and snow (Barrie et al., 1992), which indicates that low levels of these metals may travel from central Europe all the way to the Arctic (AMAP, 2005).

Svalbard is an archipelago in the high Arctic (74–81° N). The unique meteorological conditions, ice-bound environment and extreme changes in the light regime between seasons make the Arctic an accumulation zone of long-range transported (LRT) pollutants and thus a recipient of many toxic elements, including Hg, Cd and Pb (AMAP, 1997). In addition, local sources of emissions further contribute to the pollution of the Svalbard biosphere. These sources include burning fossil fuels in the local coal power plants in Longyearbyen and Barentsburg, vehicle exhaust and mining activities (Drotikova et al., 2020a, 2020b).

After being deposited, the pollutants can either undergo re-emission to the atmosphere, bind to soil organic matter or be taken up by vegetation via, for instance, leaf stomata or roots (Schaefer et al., 2020). The primary pathway of Hg into the terrestrial ecosystem is via vegetational (both vascular and non-vascular plants) atmospheric uptake, which accounts for approximately 90 % of all terrestrial Hg sequestration (Obrist et al., 2017; Zhou et al., 2021). In contrast, the major pathway for Cd and Pb accumulation in terrestrial environments is via root uptake from surrounding soils in vascular vegetation (Ismael et al., 2019; Pourrut et al., 2011). In general, non-vascular vegetation (e.g., bryophytes and lichens) efficiently accumulate atmospherically deposited heavy metals due to their overall high surface-to-mass ratio, long lifespan, and slow growth rates (Bargagli, 2016; Gjengedal and Steinnes, 1990; Steinnes, 1995). This vegetational uptake of toxic elements subsequently acts as a pathway of exposure to herbivorous animals, such as the Svalbard reindeer (*Rangifer tarandus platyrhynchus*).

The Svalbard reindeer is the largest herbivore on Svalbard. It is found all over Spitsbergen and is mainly non-migratory (Le Moullec et al., 2019). It rarely leaves its limited home range unless motivated by restricted food accessibility in winter (Hansen et al., 2010). The primary diet of the reindeer consists of vascular plants (e.g., graminoids or polar willow) and bryophytes (Bjørkvoll et al., 2009), which may act as vectors of exposure to long-range and local pollution. It is therefore argued that the Svalbard reindeer is a suitable species for biomonitoring pollution levels in the Svalbard terrestrial ecosystem (Pacyna et al., 2018). As the Svalbard reindeer inhabit a relatively restricted area throughout their lifespan, their pollution levels further reflect any potential local contributions to the background environmental pollution load (Kinck, 2014).

Excretion via faeces is one of the main pathways for eliminating pollutants and excess nutrients from an organism. Faeces is, therefore, considered an indicator of environmental dietary exposure to elements. Faeces has also been promoted as a suitable matrix for biomonitoring since it can be collected using non-invasive methods (Pacyna-Kuchta et al., 2020).

There is currently a limited number of studies evaluating pollution levels in Arctic terrestrial ecosystems and organisms, especially few focusing on endemic species that do not migrate, such as Arctic herbivores (Dietz et al., 2022; Scheuhammer et al., 2015). In this study, we, therefore, investigated the composition of toxic (e.g., Hg, Cd and Pb) and essential elements in Svalbard reindeer faeces with the key aims to I) assess the Svalbard reindeer exposure to pollutants and II) evaluate whether reindeer inhabiting a presumed pristine area have a different faecal elemental composition as compared to reindeer living in areas more influenced by anthropogenic activities. Previous studies on Svalbard reindeer have not compared the faecal elemental composition between different reindeer populations to assess the potential anthropogenic influence on the bioavailability of elements, making this study design a novel approach to Arctic biomonitoring.

In addition to faeces, the elemental composition of soil was also analysed to evaluate to which extent spatial differences in soil composition are reflected in reindeer faeces.

2. Materials and methods

2.1. Study areas

The field sampling of faeces and soil was conducted at two separate areas on central Spitsbergen, the main island of the high Arctic archipelago of Svalbard. Adventdalen (78.2°N, 15.8°E) and Nordenskiöld coast (78.1°N, 13.6°E) were chosen as sampling sites due to the presumed difference in anthropogenic influence between them. Adventdalen is a valley close to Longyearbyen (Fig. 1), the largest settlement on the island (ca. 2500 inhabitants), where local sources of pollution include a coal power plant, an airport, vehicle exhaust, mining activities and releases from other local anthropogenic activities (Warner et al., 2019; Drotikova et al., 2020a, 2020b). In contrast, the Nordenskiöld coast is likely more pristine as it is located near protected wildlife areas and further away from any direct point sources of pollution. Barentsburg, the second-largest settlement on Svalbard (ca. 500 inhabitants), is, however, situated approximately 15 km east of Nordenskiöld coast (Fig. 1), with pollution sources similar to Longyearbyen (Warner et al., 2019).

Although the sites are relatively close (~50 km), the weather conditions differ considerably between the two locations. The annual average precipitation in Adventdalen is around 190 mm, compared to 410 mm at the Nordenskiöld coast. Likewise, the annual average snow depth in Adventdalen is only half of that at Kapp Linné on the northern cape of the Nordenskiöld coast, 10.2 cm versus 20.1 cm (Førland et al., 2011; Norsk Klimaservicesenter, n.d.).

2.2. Field sampling

Faecal samples were collected in the spring (Feb-May) of 2014, 2015, and 2017. Reindeer were observed from 50 to 100 m away until faeces were excreted. Fresh faecal samples were subsequently collected within approximately 10 min. Nitrile gloves were worn during the collection to avoid sample contamination. Reindeer age (adult or calf), sex, and sampling coordinates were noted for each faecal sample. In total, 63 samples were collected from Adventdalen and 33 samples from the Nordenskiöld coast. The samples were kept in polyethylene zip-lock bags and stored frozen (−20 °C) until further analysis. Nine soil samples were collected from Adventdalen ($n = 5$) and Nordenskiöld coast ($n = 4$) in August 2021. The selected soil sampling sites had a developed organic surface layer and were entirely covered with vegetation. All samples were collected by cutting a square measuring 10 × 10 × 10 cm using a stainless-steel knife. The samples were stored in paper bags until further sample preparations. For further details of the sampling, see Table A1-A2 and Fig. A1.

2.3. Element analysis

All faecal samples ($n = 96$) were freeze-dried using an Alpha 1–2 LDplus (Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany) for 24 h before being homogenised using a TissueLyser II with Teflon® chambers (Qiagen, Hilden, Germany).

The soil samples ($n = 9$) were left to airdry until all the water had evaporated (which was determined when weight changes were within ±5 % over a week). The edges of the dried samples were removed to avoid cross- or external contamination. The cores were homogenised using a cutting mill (RETSCH SM 100, 2-mm sieve, Retsch GmbH, Haan, Germany) and stored in polyethylene bags.

The dried, homogenised soil and faecal samples were digested in HNO_3. Approximately 0.5 g of faeces or 0.3 g of soil was transferred to an 18 ml polytetrafluoroethylene (PTFE) vial with 6.25 ml or 9 ml 50 % (v/v) HNO_3 acid (ultrapure grade, purified from HNO_3, AnalaR NORMAPUR®, VWR using a sub-boiling distillation system (Milestone, SubPur, Sorisole, BG, Italy)), respectively. The samples were digested using a high-pressure microwave system (Milestone Ultraclave, EMLS, Leutkirch, Germany) for 150 min and thereafter diluted with ultrapure water (Elga® Purelab Flex 4) up to 60 g (faecal samples) or 110 g (soil samples).

Additional blank (ultrapure water and HNO_3) and certified reference material samples (Virginia Tobacco leaves, INCT-PVTL-6, Institute of Nuclear Chemistry and Technology, Warszawa, Poland) were digested and analysed for quality assurance (for the results of the CRM analysis, see Table A3).

High-resolution inductively coupled plasma mass spectrometry (HR-ICP-MS, Thermo Finnigan model Element 2 instrument, Bremen, Germany) was used to identify and quantify the elemental composition of the samples. Sixteen elements (Al, As, Ca, Cd, Cu, Fe, Hg, Mg, Na, Ni, P, Pb, S, Se, Si, and Zn) were analysed, and all were detected above the limit of detection (LOD). The final concentration of each element was obtained by subtracting the average of the method blank concentration

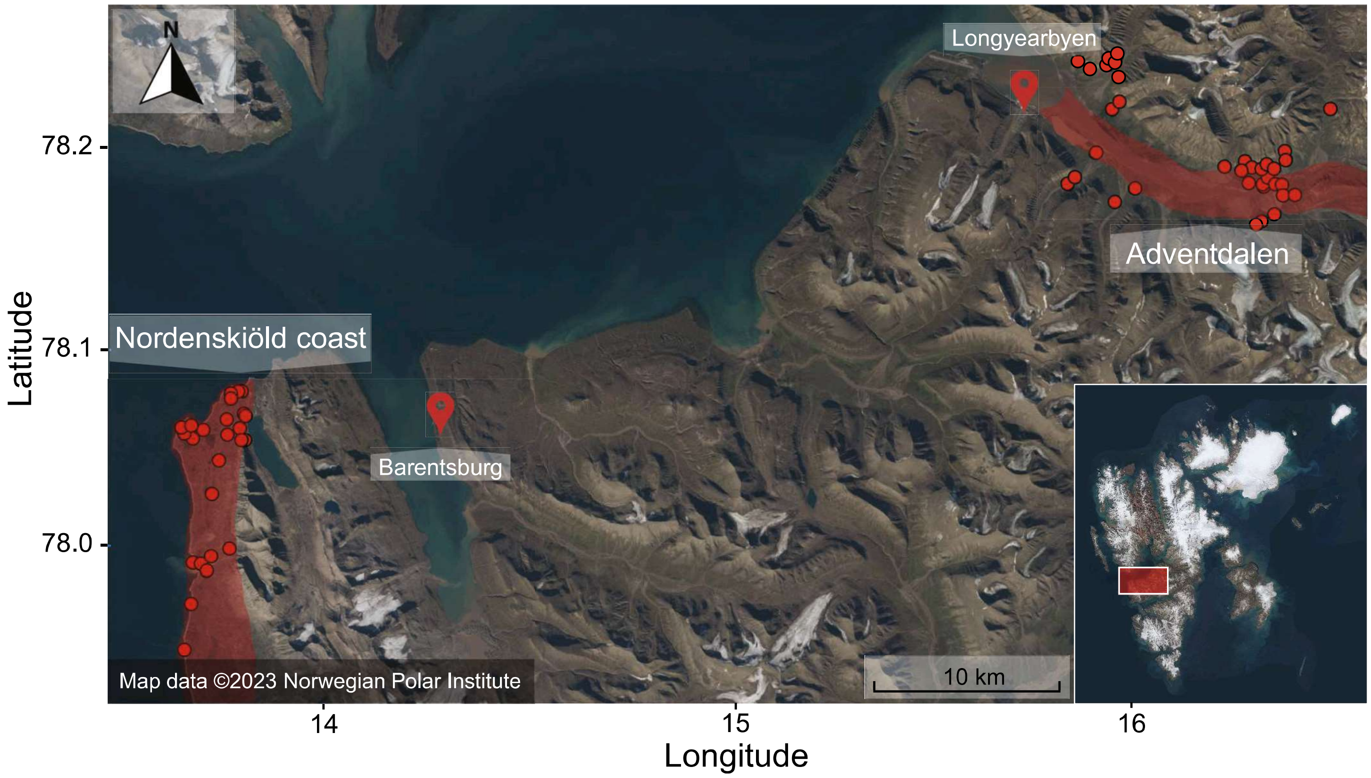


Fig. 1. Central Nordenskiöld land with Nordenskiöld coast and Adventdalen areas marked in dark red and the faecal sampling sites marked with red dots. The settlements of Barentsburg and Longyearbyen are indicated with red geotags.

of the respective element in all analysed samples.

The method detection limits for each element were either based on three times the standard deviation of the blanks or the instrument detection limits (IDL), depending on which method resulted in the highest value. The IDLs were estimated by analysing solutions with decreasing concentrations of the elements (see Table A4). The concentration that resulted in a relative standard deviation of approximately 25 % ($n = 3$ scans) was selected as IDL, with baseline corrections applied for these values.

The sixteen analysed elements include toxic metals (e.g., Cd, Pb, As and Hg) and essential elements (e.g., Se). Bedrock-related elements (i.e., Al, Ca, Cu, Fe, Mg, P, Si, Na, Ni, S and Zn) were primarily included as means to evaluate the origin of the toxic elements (Halbach et al., 2017; Klaassen et al., 2013).

2.4. Data treatment

All data were analysed for normal distribution using a Shapiro-Wilk test and for equal variances using Levene's test. None of the faecal elemental data was normally distributed, and a BoxCox transformation was applied to normalise the variables (using logarithmic or square root transformations). The transformed data did not fulfil the condition of equal variances, and non-parametric tests of analysis were therefore applied. A Mann-Whitney *U* test was employed to evaluate significant differences in faecal elemental concentrations between the two populations. Additionally, Spearman rank correlations were carried out to determine relationships among elements and between the elements and distance to the ocean at Nordenskiöld coast and Adventdalen, respectively. The results of the Spearman rank correlation were subsequently followed up using linear regression to plot the correlations. All soil elemental data (except for Pb) and parameters (TOC and pH) fulfilled the assumptions of parametric tests. A Student *t*-test was employed to evaluate significant differences in element concentration and soil parameters between the two locations (the Mann-Whitney U test was used for Pb).

Ocean distance was determined as the closest linear distance to the seashore from each sampling site. In Adventdalen, this distance was measured to the nearest point of Adventfjorden, while at Nordenskiöld coast, the distance was measured to the nearest open water of Isfjordbanken (see Fig. A1).

A principal component analysis (PCA) was conducted to explore the relationships among the elements and the spatial variation in the data set. The variable ocean distance was excluded from the PCA due to considerable differences between the sampling sites, which contributed significantly to the PCs and thus masked the contributions of the elements. All numerical data were normalised by centring and scaling it (using the mean and standard deviation) before performing the PCA. All statistical analysis was done using R Statistical Software (V4.2.2, R Core Team, 2022).

3. Results

As presented in Table 1, the faecal elemental composition significantly differed between Adventdalen and the Nordenskiöld coast. Concentrations of Cu, Zn, Al, and Fe were significantly higher in faeces from Adventdalen than at Nordenskiöld coast ($p < 0.01$). In contrast, higher Cd, Se, Si, S, Ca, As and Pb concentrations were found in the Nordenskiöld coast faecal samples ($p < 0.01$ for all elements but $p < 0.05$ for As and Pb). The levels of Hg did not significantly differ between the two locations in the faeces. There were, however, significantly higher soil concentrations of Hg at the Nordenskiöld coast and higher levels of Cu, As and Zn in Adventdalen soil ($p < 0.05$). Total organic content and pH did not differ significantly between the soil samples from the two areas, where the samples from Nordenskiöld coast had an average TOC of 13.1 ± 6.9 % and an average pH of 5.3 ± 0.6, compared to the samples from Adventdalen which had an average TOC of 10.0 ± 5.1 % and an average pH of 5.0 ± 0.3.

The PCA for the faecal elemental composition resulted in three significant principal components (PCs), explaining 69.6 % of the total variation in the dataset. The observed spatial difference in reindeer faecal elemental composition (Table 1) is also indicated in the PCA score plot (Fig. 2A), which shows a slight separation between the two sampled reindeer populations along PC1. The clustering of elements in the loading plot (Fig. 2B) indicates that elements such as Al, Ni, Fe and Cu are strongly positively related. Many other elements are spread out along PC1 and PC2 without clustering. However, some elements are suggested to correlate positively, such as P and Ca or Cd and Si. The elements Hg, Se and Pb are somewhat grouped along PC2, which may suggest a positive relationship between these variables. PC3 (visualised in Fig. A2) likewise demonstrates similar loadings for Hg, Pb, Cd and Se, which supports a relationship between these elements. When comparing the score and loading plots, it is indicated that samples collected from Adventdalen had the highest levels of Al, Ni, Fe, and Cu. In contrast, faeces from the Nordenskiöld coast were characterised by higher

Table 1

Element concentrations in Svalbard reindeer (*R. tarandus platyrhynchus*) faeces and soil from Adventdalen and Nordenskiöld coast. Average elemental concentrations ± SD are provided in regular font. Concentrations given in italic font provide the median (min-max) values. All concentrations are given in μg/g (dry weight). Significantly different concentrations between the two sampling areas are indicated as (*) $p < 0.05$; (**) $p < 0.01$.

	Faecal samples					Soil samples		
(*n*)	Adventdalen (63)		Sig.	Nordenskiöld coast (33)		Adventdalen (5)	Sig.	Nordenskiöld coast (4)
	Mean ± SD	*Median (min-max)*		Mean ± SD	*Median (min-max)*	Mean ± SD		Mean ± SD
Hg	0.0777 ± 0.0193	*0.0759 (0.0467-0.165)*		0.0869 ± 0.0429	*0.0853 (0.0200-0.178)*	0.112 ± 0.0426	(*)	0.178 ± 0.0291
Se	0.354 ± 0.104	*0.322 (0.184-0.655)*	(**)	0.560 ± 0.248	*0.516 (0.154-1.06)*	0.809 ± 0.385		0.725 ± 0.255
Cd	0.366 ± 0.288	*0.271 (0.0662-1.19)*	(**)	0.552 ± 0.297	*0.482 (0.203-1.72)*	0.374 ± 0.259		0.252 ± 0.123
As	3.22 ± 1.92	*3.05 (0.411-11.9)*	(*)	3.44 ± 3.97	*1.24 (0.322-13.4)*	11.3 ± 4.78	(*)	4.57 ± 1.11
Pb	3.81 ± 1.63	*3.62 (0.887-9.21)*	(*)	5.36 ± 2.91	*5.19 (1.51-13.9)*	12.5 ± 3.72		14.5 ± 1.91
Cu	8.41 ± 2.00	*8.22 (4.49-14.8)*	(**)	5.71 ± 2.01	*5.32 (2.50-13.3)*	16.2 ± 2.80	(*)	10.5 ± 3.8
Ni	10.6 ± 3.73	*10.5 (3.11-21.7)*	(**)	4.50 ± 2.59	*4.22 (1.85-17.5)*	24.3 ± 4.40		16.9 ± 3.91
Zn	78.7 ± 25.9	*75.8 (39.2-178)*	(**)	57.4 ± 16.6	*55.3 (28.9-87.1)*	80.0 ± 15.4	(*)	50.2 ± 14.1
Na	636 ± 360	*600 (110-2990)*		723 ± 644	*538 (187-3430)*	432 ± 78.8		1030 ± 682
Si	1390 ± 330	*1380 (780-2290)*	(**)	1960 ± 362	*2000 (1330-2530)*	1390 ± 707		1670 ± 458
P	1520 ± 459	*1360 (893-2930)*		1583 ± 482	*1550 (621-2980)*	802 ± 124		1050 ± 382
S	1780 ± 208	*1750 (1370-2270)*	(**)	3160 ± 2490	*2040 (679-10700)*	1270 ± 424		1290 ± 315
Mg	2600 ± 499	*2560 (1440-4110)*	(**)	4820 ± 3290	*3390 (1390-16500)*	4690 ± 563		5840 ± 981
Fe	7690 ± 3860	*6980 (1080-18000)*	(**)	3960 ± 1410	*3930 (1530-9120)*	25,700 ± 8890		20,900 ± 4080
Al	9950 ± 5260	*9950 (1340-28600)*	(**)	4220 ± 1480	*4260 (1550-9520)*	29,500 ± 7330		2570 ± 7030
Ca	16,000 ± 360	*15900 (8230-21500)*	(**)	30,700 ± 810	*30700 (11700-57000)*	6870 ± 3420		8560 ± 7130

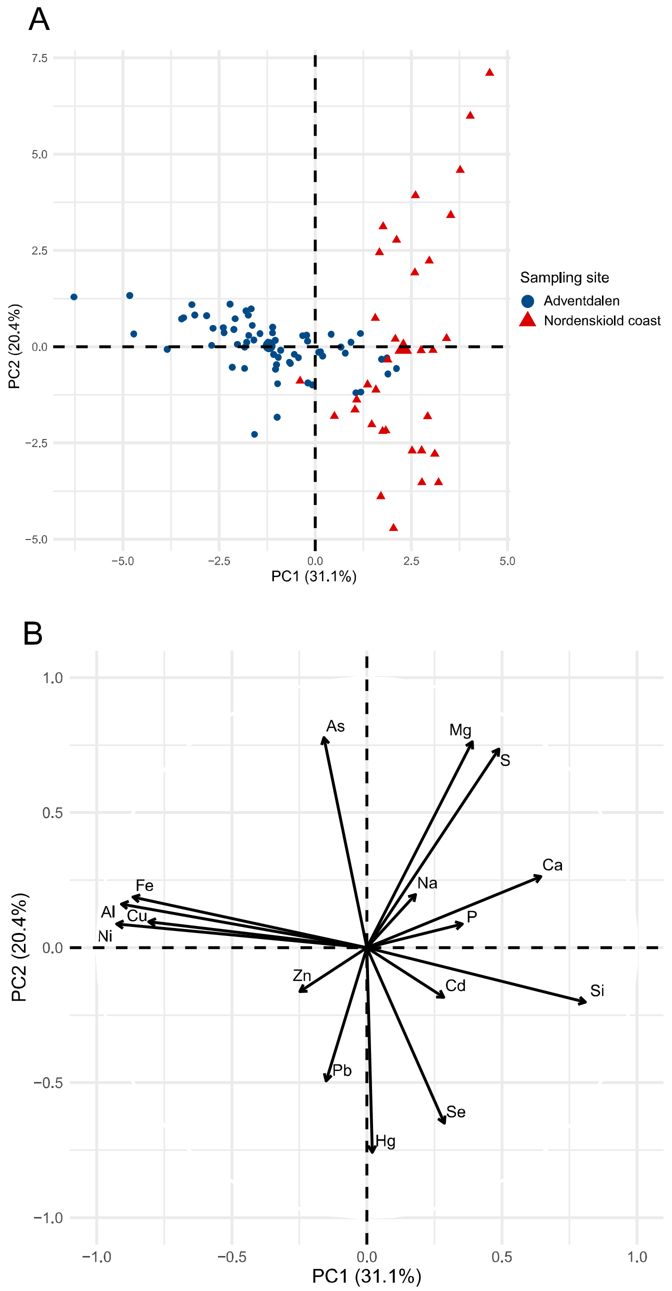
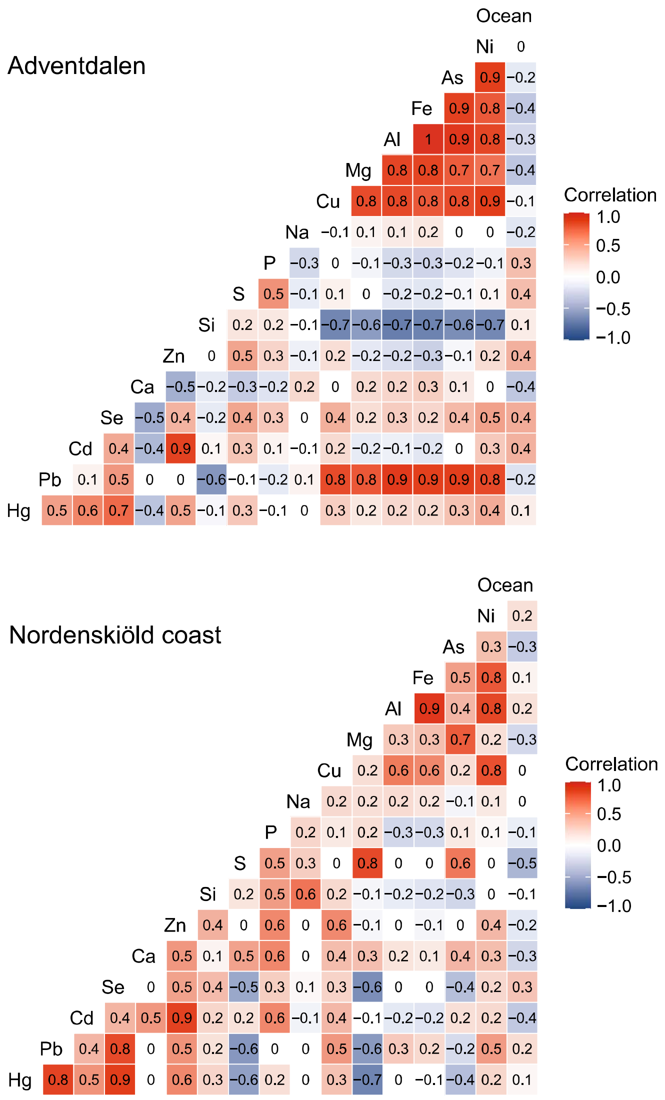


Fig. 2. A. PCA score plot for Svalbard reindeer (*R. tarandus platyrhynchus*) faecal elemental composition from Adventdalen and Nordenskiöld coast. PC1 and PC2 explain 51.5% of the variance in the data set.
B. PCA loading plot for selected elements in Svalbard reindeer (*R. tarandus platyrhynchus*) faeces. PC1 and PC2 explain 51.5% of the variance in the data set. Elements with high loadings have long arrows, while elements with lower loading have shorter arrows. See Table A5 for the component matrix, presenting the loading of each element on the PCs.



Adventdalen

	Pb	Cd	Se	Ca	Zn	Si	S	P	Na	Cu	Mg	Al	Fe	As	Ni	Ocean
Ni																0
As															0.9	−0.2
Fe														0.9	0.8	−0.4
Al													1	0.9	0.8	−0.3
Mg												0.8	0.8	0.7	0.7	−0.4
Cu											0.8	0.8	0.8	0.8	0.9	−0.1
Na										−0.1	0.1	0.1	0.2	0	0	−0.2
P									−0.3	0	−0.1	−0.3	−0.3	−0.2	−0.1	0.3
S								0.5	−0.1	0.1	0	−0.2	−0.2	−0.1	0.1	0.4
Si							0.2	0.2	−0.1	−0.7	−0.6	−0.7	−0.7	−0.6	−0.7	0.1
Zn						0	0.5	0.3	−0.1	0.2	−0.2	−0.2	−0.3	−0.1	0.2	0.4
Ca					−0.5	−0.2	−0.3	−0.2	0.2	0	0.2	0.2	0.3	0.1	0	−0.4
Se				−0.5	0.4	−0.2	0.4	0.3	0	0.4	0.2	0.3	0.2	0.4	0.5	0.4
Cd			0.4	−0.4	0.9	0.1	0.3	0.1	−0.1	0.2	−0.2	−0.1	−0.2	0	0.3	0.4
Pb		0.1	0.5	0	0	−0.6	−0.1	−0.2	0.1	0.8	0.8	0.9	0.9	0.9	0.8	−0.2
Hg	0.5	0.6	0.7	−0.4	0.5	−0.1	0.3	−0.1	0	0.3	0.2	0.2	0.2	0.3	0.4	0.1

Nordenskiöld coast

	Pb	Cd	Se	Ca	Zn	Si	S	P	Na	Cu	Mg	Al	Fe	As	Ni	Ocean
Ni																0.2
As															0.3	−0.3
Fe														0.5	0.8	0.1
Al													0.9	0.4	0.8	0.2
Mg												0.3	0.3	0.7	0.2	−0.3
Cu											0.2	0.6	0.6	0.2	0.8	0
Na										0.2	0.2	0.2	0.2	−0.1	0.1	0
P									0.2	0.1	0.2	−0.3	−0.3	0.1	0.1	−0.1
S								0.5	0.3	0	0.8	0	0	0.6	0	−0.5
Si							0.2	0.5	0.6	0.2	−0.1	−0.2	−0.2	−0.3	0	−0.1
Zn						0.4	0	0.6	0	0.6	−0.1	0	−0.1	0	0.4	−0.2
Ca					0.5	0.1	0.5	0.6	0	0.4	0.3	0.2	0.1	0.4	0.3	−0.3
Se				0	0.5	0.4	−0.5	0.3	0.1	0.3	−0.6	0	0	−0.4	0.2	0.3
Cd			0.4	0.5	0.9	0.2	0.2	0.6	−0.1	0.4	−0.1	−0.2	−0.2	0.2	0.2	−0.4
Pb		0.4	0.8	0	0.5	0.2	−0.6	0	0	0.5	−0.6	0.3	0.2	−0.2	0.5	0.2
Hg	0.8	0.5	0.9	0	0.6	0.3	−0.6	0.2	0	0.3	−0.7	0	−0.1	−0.4	0.2	0.1

Fig. 3. Spearman correlation heat maps demonstrating the relationship between faecal elemental concentrations and distance to the ocean (Ocean) in samples from Adventdalen ($n = 63$) and Nordenskiöld coast ($n = 33$). Values in each box represent the Spearman correlation coefficient (r), where $|r| > 0.2 = p < 0.05$, $|r| > \pm 0.3 = p < 0.01$ and $|r| > \pm 0.4 = p < 0.001$.

concentrations of Mg, S, Ca, Si, Se and Cd, P, and Na to a lesser extent.

The clustering in the PCA loading plot indicated positive relationships between many elements in the faecal samples. Site-specific correlations between variables are shown in Fig. 3 (Spearman correlation). Strong positive correlations between Hg and Se ($p < 0.001$) and between Cd and Zn ($p < 0.001$) were found in the faecal samples from both locations (see Fig. 4). The molar ratio between Se:Hg was, on average, 13.6. Elements Al, As, Cu, Fe and Mg were positively associated. The relationships between these elements were, however, stronger in the faecal samples from Adventdalen ($p > 0.001$) compared to Nordenskiöld coast (significant correlation between Fe, Al and Ni, $p > 0.001$). Pb was strongly correlated to Hg in Nordenskiöld coast ($p < 0.001$) but had a stronger relationship to elements such as Al, Cu, Fe and Ni ($p < 0.001$) in Adventdalen. There were no strong correlations between the elements and ocean distance (Fig. 3). Overall, no indications of any relationship between faecal elemental concentrations and variables such as age, sex or year of sampling were identified (using Mann-Whitney *U* test).

4. Discussion

The faecal elemental concentrations of the toxic elements Hg, Pb and Cd are overall comparable to concentrations reported in a previous study, where Pacyna et al. (2019) detected similar ranges of Hg (0.069 ± 0.016 μg/g), Pb (5.23 ± 3.05 μg/g) and Cd (0.866 ± 0.411 μg/g) in fresh Svalbard reindeer faeces from Spitsbergen. There were, however, significant site-specific differences between the faecal elemental composition in samples from Adventdalen and Nordenskiöld coast. This suggests that, despite the two investigated sites being relatively close, there is a spatial influence on elemental exposure, uptake, or excretion of specific elements in the reindeer.

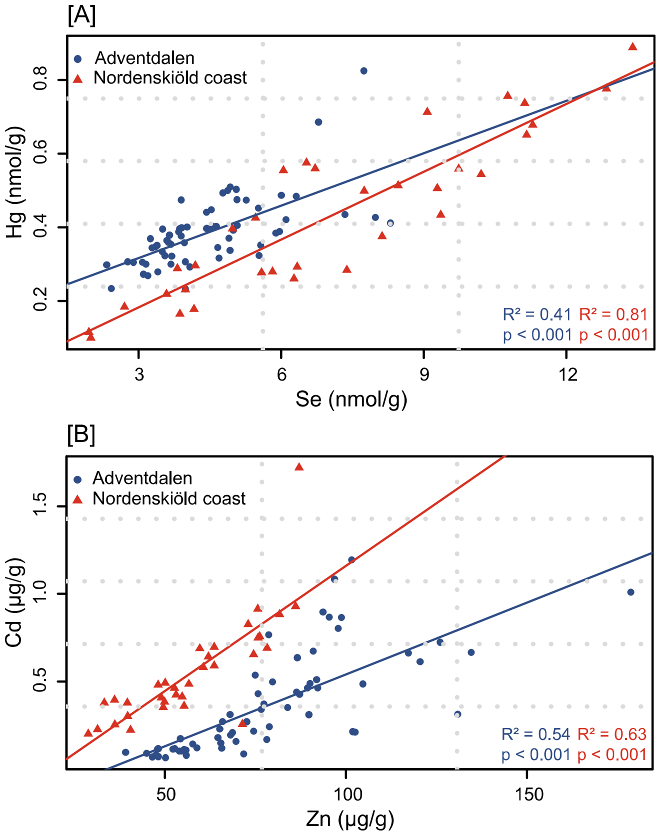


Fig. 4. Correlation plots between [A] Hg and Se and [B] between Cd and Zn in the faecal samples.

4.1. Dietary differences

Reindeer are exposed to elements through multiple pathways, but their exposure mainly derives from the vegetation on which they feed. The elemental composition of vegetation is affected by atmospheric deposition and soil composition, which further depends on the geochemical characteristics of the underlying bedrock (Ismael et al., 2019; Pourrut et al., 2011). For instance, Kłos et al. (2017) reported a species-dependent preference in metal absorption from the underlying soil when evaluating the metal accumulation capacity in different species of vascular plants on Svalbard. Mosses and lichen are known accumulators of atmospheric pollutants as they lack roots and depend on the ambient atmosphere for nutrients and water, making them susceptible to the absorption of pollutants from the air (Bargagli, 2016). Different species of plants will, therefore, differ in their elemental composition. When comparing the uptake of metals in lichen, bryophytes (i.e., mosses) and vascular vegetation, a study found the highest levels of Pb (2.1–2.4 µg/g dw) in lichen and mosses, Hg (0.033 µg/g dw) in mosses and Cd (0.52 µg/g dw) in vascular vegetation such as *Salix polaris* (Wojtuń et al., 2013).

The Svalbard reindeer diet consists of graminoids, forbs, bryophytes, and shrubs. Their diet undergoes seasonal shifts based on the availability of the different vegetation species and is in late winter dominated by *S. polaris* (Bjørkvoll et al., 2009). Similarly to other *Salix* spp., *S. polaris* is a phytoaccumulator (i.e., a plant with the capacity to absorb, accumulate and tolerate elevated levels of, for instance, heavy metals) (Pulford, 2003). A study conducted in Svalbard found levels of Cd up to sixty times higher in the stems of the plant than in the underlying soil (Wojtuń et al., 2019). *S. polaris* is widespread over the archipelago of Svalbard and has been identified in both Adventdalen and Nordenskiöld coast (Artsdatabanken, n.d.). The fraction of *S. polaris* in the diet of the reindeer depends on the presence of other species of vegetation, which may influence their exposure to elements such as Cd. In Svalbard reindeer, it has been reported that season-dependent (i.e., winter versus summer) dietary shifts can result in variations in faecal concentrations of, for instance, Cd and Fe (Węgrzyn et al., 2018). As a result, differences in faecal elemental composition between the two populations may reflect potential dietary differences. There is, however, a lack of studies on the presence of different species of vegetation at the two sampling sites, as well as the dietary differences among these two reindeer populations, creating difficulties in proving this theory.

4.2. Element correlations in faeces

The excretion of elements via faeces is one elimination pathway of toxic metals in exposed organisms (Pacyna et al., 2019; Roggeman et al., 2013). Faecal concentrations of essential elements (i.e., nutrients that are vital for homeostasis) mainly depend on the current physiological requirements of the individual. Hence, to what extent the faecal concentration reflects the intake of essential elements will depend on the uptake rate, potential physiological needs, and elimination rates via faecal excretion.

Se is an essential element as it is a central part of the biological antioxidant defence system and, therefore, an active element in the protection against toxic metals. The protection mechanism includes reacting with metals such as Hg and forming an equimolar, inactive Se—Hg complex, often found in the liver (Ikemoto et al., 2004; Nève, 1991). High exposure to Hg and, thus, increased formation rates of these complexes can cause a deficiency of Se. However, Romero et al. (2016) and Sørmo et al. (2011) argue that a molar ratio of Se:Hg > 1 ensures a sufficient supply of Se to sustain its physiological function while, in addition, protecting against Hg. As the relationship between Se and Hg in this study far exceeded a molar ratio of 1 (minimum faecal Se:Hg = 7.8), Hg exposure does not seem to limit the Se availability for physiological functions or result in Hg toxicity. Although, considering that the analysed matrix is faeces, the elemental ratio in these samples provides us with little insight into the relationship between Hg and Se in internal organs such as the liver, where these Hg—Se complexes are stored, or whether there are sufficient internal concentrations of this essential element to sustain homeostasis.

As shown in Fig. 3, Cd concentrations strongly correlate to Zn in the faecal samples from both locations. To our knowledge, no previous studies have reported a correlation between these two elements in faeces. However, these elements are known to positively correlate in other matrices, such as in ore deposits or vegetation (Kabata-Pendias and Szteke, 2015). Multiple studies have evaluated the elemental composition of Svalbard vegetation. Some studies found Cd and Zn to positively correlate in species such as *S. polaris* or the moss *Dicranum angustum* (Ma et al., 2020; Wei et al., 2022; Wojtuń et al., 2019), while other studies did not (Krajcarová et al., 2016). Faecal levels of metals, including toxic metals, have been reported to correspond well with the dietary intake of these elements in grazers (*Bos taurus*) (Roggeman et al., 2013). This both suggests that increased exposure to metals does not necessarily result in higher intestinal uptake and that faecal metal levels depend on current exposure rather than representing previous assimilation that has been taken up, circulated, and subsequently eliminated via faeces. Consequently, the faecal elemental composition most likely depends on the elemental composition of the plants that comprise the diet. This may help explain the positive Cd—Zn relationship observed in the faeces, as these elements correlate in plant species consumed by reindeer (Ma et al., 2020; Wei et al., 2022; Wojtuń et al., 2019). Whether plant elemental composition also explains the significant correlation between faecal Hg and Se remains uncertain. To our knowledge, no studies have evaluated this correlation in Arctic vegetation or species included in the Svalbard reindeer diet. However, a relationship between Hg and Se has been observed in vegetation such as rice (*Oryza sativa*), suggesting a positive faecal Hg—Se correlation could derive from dietary exposure (Bai et al., 2019; Zhang et al., 2012).

4.3. Site-specific differences

Reindeer can consume both gravel and soil during grazing, further emphasising the influence of the elemental composition of the underlying bedrock on elemental exposure (Makarov et al., 2022; Orpin et al., 1985). The dominant bedrock material in Adventdalen is composed of different sandstone forms, which are dominated by Si, Al and Fe in descending order of concentrations (Middleton, 1960). In contrast, the Nordenskiöld coast has a more complex bedrock composition, which, in addition to sandstone, consists of chert and siliceous shale. These bedrocks contain high levels of Si, and limestone, which is composed of Ca (Cressman, 1967). However, despite the above-mentioned theoretical geochemical differences in parent bedrock material, only Zn, Cu, As and Hg concentrations differed significantly among all the analysed elements in the soil samples from the two sites. The differences in Zn, Cu and As in soil were reflected in the faeces, as these elements were significantly higher in faecal samples from Adventdalen, suggesting that these elements are of geogenic origin. There were, however, no differences in Hg faecal levels between the two locations.

Although the elemental distribution of the soil from the two locations may be similar, it is important to acknowledge that these elements might be incorporated into different types of minerals or be bound to different types of organic matter, which affects their bioavailability. Soil mobility of metals bound to organic matter is, for instance, higher than the mobility of metals bound to sulfide minerals, which have low solubility and are relatively stable under normal environmental conditions (John and Leventhal, 1995). Mobile elements are available to be taken up by vegetation and subsequently enter the food chain when the plants are grazed upon. Soil bioavailability may, therefore, partly explain the decoupling between the faecal and soil elemental composition at the two locations.

The organic content of Svalbard soils is, in general, far lower than in the soil from mainland Norway (Halbach et al., 2017). This results from slow rates of organic soil formation and cryoturbation, which allows for the mixing of the organic and mineral layers (Nygård et al., 2012). This can be observed in the low TOC content in the soil samples from both study sites, where the organic carbon percentage ranged between 3.3 % (in Adventdalen) and 21.6 % (at Nordenskiöld coast). Consequently, the sampled surface soils are highly characterised by the underlying mineral soils, which should reflect the differences in parent bedrock material between the two sampling areas better. The lack of significant difference in most analysed elements in the soil may be ascribed to poor statistical power, as few soil samples were included in the study ($n = 5$ and $n = 4$ for Adventdalen and Nordenskiöld coast, respectively).

In addition to the geochemical background, atmospheric deposition may influence the elemental composition in the sampling areas. The scavenging of airborne pollutants and aerosols through wet deposition has been found to contribute to levels of Cd and Hg in Arctic regions (Macdonald et al., 2005; Outridge et al., 2002; Pearson et al., 2019). Annual precipitation at Nordenskiöld coast is higher than in Adventdalen (Førland et al., 2011). The Nordenskiöld coast is furthermore closer to the sea and the Arctic Ocean when compared to Adventdalen. The on-land deposition of atmospheric Cd, to some extent, depends on the formation of particles with aerosols and, therefore, increases close to sources of open water where sea salt and water droplets act as scavengers (AMAP, 2005). This may explain the significantly higher levels of Cd in the Nordenskiöld coast faecal samples. However, no differences in Cd concentration were found in the soil samples from the two locations. On the contrary, Hg concentrations do not differ in faeces from the two study sites, while concentrations of Hg were significantly higher in soil samples from the Nordenskiöld coast than in Adventdalen. Atmospheric mercury depletion events (AMDEs) are enhanced in areas closer to the sea as the ocean is a source of halogens, which initiate depletion by oxidising atmospheric elemental mercury (Lindberg et al., 2002; Steffen et al., 2008). Increased deposition levels at Nordenskiöld coast may explain the higher levels of Hg in soil. The dominant Hg uptake pathway in vegetation is, however, via the atmosphere, while root uptake from the soil is negligible (Zhou et al., 2021). Increased levels of Hg in the soil may, therefore, not translate into increased vegetation levels, which might explain the concentration decoupling between the soil and faecal Hg concentrations.

Another vector of pollution in Arctic terrestrial ecosystems is marine birds. Seabirds such as the little auk (*Alle alle*) feed off the marine ecosystem and breed in bird cliffs on the slopes of certain high land-based cliffs. Through processes such as the excretion of faeces or the dropping of food when flying, they transfer nutrients, elements and contaminants, such as toxic elements, from the marine to the terrestrial environment as they move between the ecosystems (Zwolicki et al., 2016). As a result, increased levels of soil nutrients such as Ca, Cu, Se, Zn and toxic elements such as Cd have been found in areas in which seabirds breed (Mallory et al., 2015; De La Peña-Lastra et al., 2022). This may help explain why levels of Ca, Se and Cd are significantly higher in the faecal samples from Nordenskiöld coast. There is moreover a strong correlation between Ca and P in Nordenskiöld coast, which is not observed in the samples from Adventdalen. P is a major component of bird guano (Duda et al., 2020) and would, therefore, correlate well with other guano-related elements such as Ca. There are, however, only marginal differences in P concentrations in the reindeer faeces from the two sampling sites. There is a bird reserve on the northernmost point of Nordenskiöld coast, but there are no seabird cliffs in the area. Therefore, it is not likely that the increased levels of Cd derive from bird guano.

Despite Cd levels being higher in Nordenskiöld coast faeces, the heat maps (Fig. 3) do not support a relationship between toxic metals and the distance to the ocean. It is, however, important to note that the reindeer's digestive retention time ranges from 21 to 69 h (Aagnes and Mathiesen, 1994). Hence, the location of excretion can differ from that of grazing, thus confounding the possible association between the distance from the ocean and the faecal elemental composition.

The heat maps, however, showed significant correlations between several elements in the faecal samples. At Nordenskiöld coast there was a significant positive relationship between Pb and Hg. In Adventdalen, the results instead showed significant positive relationships between Pb and several elements of mineral origin, such as Cu, Al, Fe, and As. The Pb levels were, moreover, higher in faeces from the Nordenskiöld coast, and we, therefore, suggest that the origin of Pb may differ at the two sampling sites. We propose that the Pb detected in the faecal samples from Adventdalen could be of mineral origin, while the positive relationship between Hg and Pb, together with increased levels of Pb at Nordenskiöld coast, may suggest anthropogenic input.

4.4. Local sources of pollution

Most Hg in the Arctic originates from anthropogenic activities and long-range atmospheric transport from sub-Arctic latitudes (Dastoor et al., 2022). However, as mentioned, a few local sources also contribute to the overall levels of several toxic elements in Svalbard (Drotikova et al., 2020a). As shown, the levels of the toxic elements Cd and Pb were higher in faeces from the Nordenskiöld coast than from Adventdalen. Additionally, Hg levels were higher in the soil at Nordenskiöld coast than in Adventdalen. This potentially contradicts our presumption that the Nordenskiöld coast is less affected by anthropogenic influences. There are no significant local point sources of pollutants on the Nordenskiöld coast. The area is, however, relatively close to the nearby settlement of Barentsburg (15 km), where there is an active coal power plant (Dekhtyareva et al., 2016). The annual general wind direction in Barentsburg is northeasterly, which may transport a considerable amount of the power plant exhaust towards the Nordenskiöld coast (Åkerman, 1980; The Norwegian Meteorological Institute, n.d.). Depending on the chemical composition of the coal used in these power plants, combustion can be a source of fossil fuel-related elements, such as heavy metals (Lewińska-Preis et al., 2009). Anthropogenic activity in Barentsburg could, therefore, to some extent, be the common origin of both Hg and Pb, which may explain their relationship at this site. The same relationship between Hg and Pb was absent in faecal samples from Adventdalen despite its closeness to the coal power plant in Longyearbyen (3–18 km). The general annual wind direction in Adventdalen is, however, mostly southeasterly (Dekhtyareva et al., 2022), which is downwind from Longyearbyen and its associated point sources. The sampled reindeer in Adventdalen may, therefore, not be exposed to the potential anthropogenic contributions of toxic metals originating from the Longyearbyen powerplant.

The elemental composition of faeces is the final product of the exposure, intestinal uptake and excretion of surplus essential or toxic elements. Roggeman et al. (2013), for instance, reported that faecal concentration of elements corresponded well with dietary exposure. The levels of elements in faeces do, therefore, not necessarily reflect the levels in other tissues or organs of the organism. Although it has been argued that faecal samples can be used as a proxy for exposure to pollutants in terrestrial ecosystems (Pacyna et al., 2019), relationships between faecal concentrations of toxic metals and internal soft tissue and organ concentrations have been found in some Arctic species, but not yet been assessed in reindeer (Dietz et al., 2009). The biomonitoring of faeces should, therefore, instead be used as an indicator of exposure trends in Arctic terrestrial organisms rather than being used to derive the risk of exposure to, for instance, toxic metals. We furthermore suggest that faecal samples of reindeer can be applied to assess spatial and temporal differences and trends in patterns and levels of toxic elements (heavy metals) at the base of the food chain (i.e. in reindeer diet) in the Arctic.

5. Conclusions

In this study, we investigated the composition of toxic (e.g., Hg, Cd and Pb) and essential elements (e.g., Se) in Svalbard reindeer faeces with the key aim to I) assess the exposure of Svalbard reindeer to pollutants and II) evaluate whether reindeer inhabiting pristine areas have a different faecal elemental composition as compared to reindeer living in areas more influenced by anthropogenic activities. We found a significant difference in faecal elemental concentration and composition between Adventdalen and Nordenskiöld coast. Elements of geogenic origin (e.g., Al, Cu and Fe) were found at higher levels in faeces from Adventdalen. In comparison, levels of Ca, Se and the toxic elements Cd and Pb were higher in faecal samples from the Nordenskiöld coast. The significantly higher levels of Cd found in faeces from the Nordenskiöld coast may originate from the ocean or, considering the easterly wind patterns in late winter, from the coal combustion power plant in Barentsburg, located 15 km east of Nordenskiöld coast. The increased levels of Pb at Nordenskiöld coast faeces might also originate from the powerplant. These results indicate that reindeer living on the Nordenskiöld coast may be exposed to local anthropogenic pollution.

However, the same patterns in faecal elemental composition at the two study locations were not observed in the elemental composition of the soil samples from the same areas. This decoupling between matrices may be due to the reindeer diet, as plants have species-dependent uptake of elements from their surrounding environment. Potential dietary differences between the two sampled populations may also explain the differences in faecal elemental composition, but further studies on the elemental composition of the plants that comprise the reindeer diet are required to evaluate this.

We suggest that faecal samples of reindeer can be applied to assess spatial and temporal differences and trends in patterns and levels of toxic elements at the base of the terrestrial food chain in the Arctic.

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CRediT authorship contribution statement

Malin Andersson Stavridis: Formal analysis, Writing – original draft, Visualization. **Susanne Brix Røed:** Conceptualization, Investigation. **Brage Bremset Hansen:** Investigation, Writing – review & editing. **Øyvind Mikkelsen:** Investigation, Writing – review & editing. **Tomasz Maciej Ciesielski:** Conceptualization, Writing – review & editing. **Bjørn Munro Jenssen:** Conceptualization, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Soil and Faecal Elemental Concentrations (Original data) (Mendeley Data).

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