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Steep declines in radioactive caesium after 30 years of monitoring alpine plants in mountain areas of central Norway

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ABSTRACT

The Chernobyl accident exposed large areas of northern Europe to radiocaesium (^{137}Cs). We investigated temporal and spatial variation in concentrations of radiocaesium among five functional groups of alpine plants at two mountain areas in central Norway over a 31-year period from 1991 to 2022. Average concentrations of radiocaesium were initially high in lichens and bryophytes at around 4600–6400 Bq/kg dry weight during 1991–1994 but then decreased dramatically over three decades to current concentrations of <200 Bq/kg for all plant groups in 2019–2022. The effective half-life of radiocaesium was estimated to be 4–6 years in lichens and mosses, 7–13 years in herbaceous plants, and 22–30 years in woody plants, which were less than the physical half-life of 30.2 years. Concentrations of radiocaesium were greater at the nutrient-poor site than at the nutrient-rich site, probably due to greater deposition levels at higher elevations and the geographical pattern of the deposition. Functional groups of plants differed with higher concentrations among non-vascular than vascular plants. Common heather *Calluna vulgaris* was unusual among woody plants with high concentration of radiocaesium, especially in the new shoots. Our new estimates of concentrations and dynamics of radiocaesium for alpine plants in natural environments will be useful for modelling herbivore exposure and evaluating potential impacts on wildlife and human health.

1. Introduction

Norway received large amounts of radiocaesium (^{137}Cs) from atmospheric tests of nuclear weapons in the 1950–60's and with the accident at the Chernobyl Nuclear Power Plant in 1986 (Backe et al., 1986; Bretten et al., 1992; Varskog et al., 1994; De Cort et al., 1998). The Chernobyl accident alone led to a total deposition of up to 2300 ± 200 TBq from radioactive ^{137}Cs isotopes but with considerable geographic variation in levels of contamination (Backe et al., 1987). Mid and central regions of Norway experienced the highest deposition rates, resulting in detrimental exposure for both terrestrial and aquatic ecosystems. Radiocaesium is an element of environmental concern due to emissions of both β- and γ-radiation during decay. The physical half-life of ^{137}Cs is relatively long at 30.17 years and the isotope can have rapid and long-lasting incorporation into biological systems (White and Broadley 2000). It can also be highly mobile and transfer quickly through food chains (Anspaugh and Balonov 2006, De Medici et al., 2019). Due to the slow biological turnover rate in cold areas with a short growing season, radiocaesium and other fission products may persist longer in arctic and alpine ecosystems (Heinrich et al., 1999). Moreover, long-lasting availability of radionuclides have been shown to be the source of higher transfer in forest ecosystems as compared to agricultural areas.

Since 1986, environmental levels of radiocaesium have been monitored in different species of plants, wildlife, and livestock in Norway and elsewhere in northern Europe. In Norway, concerns were immediately raised after the Chernobyl accident in 1986 especially because the most heavily contaminated areas overlapped with key grazing areas for reindeer and domestic sheep, and hence fallout had consequences not only for alpine habitats in the mountains, but also for milk and meat production for human consumption (Skuterud et al., 2005). The maximum permitted concentrations for radiocaesium in food for sale in Norway are 370 Bq/kg for milk and dairy products, 3000 Bq/kg for freshwater fish, wild game, and reindeer, and 600 Bq/kg for other foods (DSA Norwegian Radiation and Nuclear Safety Authority, 2021). Exposure to radiocaesium remains an ongoing problem, and sheep herds in selected municipalities have been brought down to cultivated land and given clean feed for several weeks before slaughter (Liland and Skuterud 2013; Skuterud and Thørring 2021). Exposure to ^{137}Cs has also

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been reported in reindeer (*Rangifer tarandus*, Skuterud et al., 2005; Skuterud and Thørring 2021), large carnivores including Eurasian lynx (*Lynx*) and brown bear (*Ursus arctos*, Chaiko 2012; Gjelsvik et al., 2014), and traces have even been detected in marine mammals from Svalbard and the Barents Sea (Andersen et al., 2005).

Herbivores and carnivores are exposed to radioactive contaminants through their diet, and long-term uptake by plant species is therefore the main entry of radiocaesium into food webs (Clint and Dighton 1993; Broadley and Willey 1997; Zhu and Smolders 2000). Estimates of the rates of uptake, transport and persistence in different plants is necessary for a better understanding of how radioactive caesium can accumulate and be transported among trophic levels in terrestrial ecosystems (Åhman 2007).

Radiocaesium can enter plant tissues through the aboveground parts due to external surface contamination by air and water, or through root networks growing in contaminated water and soils (Nishita et al., 1961). However, the uptake depends on the bioavailability of the radiocaesium, which is dependent upon soil structure and composition, physical factors such as pH and temperature, as well as environmental factors such as soil microorganisms (Burger and Lichtscheidl 2018). In some areas, the ^{137}Cs will be bound to clay minerals, and hence not directly available for plant root uptake (Salt and Mayes, 1993). Radiocaesium can also, at some localities, be easily available for uptake in plant roots due to its chemical properties which resembles that of potassium ions (K^+) (White and Broadley 2000; Zhu and Smolders 2000). Uptake from roots allows for radiocaesium to be transported internally to aboveground plant parts, and low levels of potassium can also facilitate caesium uptake by plants at some locations (Zhu and Smolders 2000). The ^{137}Cs isotope can remain in the upper layers of the soil for a long time, depending on the bioavailability. The highest concentrations of ^{137}Cs are typically in the upper 5 cm of the soil profile (Fawaris and Johanson, 1994; Matsuda et al., 2015), and several studies have confirmed that the isotope remains within the top layers of the soil (Bretten et al., 1992; Konopleva et al., 2009; De Medici et al., 2019). Plants with a flat growth form and a high surface to volume ratio can be therefore be more likely to intercept and uptake radiocaesium. Exposure of aboveground plant parts is most relevant from a short-term perspective and especially immediately after a fallout event, whereas take-up by the root systems is more relevant from a long-term perspective, which was the case for our study.

Here, we present a long-term study of ^{137}Cs in selected alpine plant species including macrolichens, bryophytes and vascular plants in two mountain areas of central Norway. The objectives of our field study were threefold. First, we were interested in temporal variation in the concentrations of radiocaesium in alpine plants. Our time series data were collected over a 31-period from 1991 to 2022. The duration of our project was comparable to the physical half-life of ^{137}Cs at $t_{1/2} = 30.2$ years and we predicted that radiocaesium concentrations might drop at a faster rate than the rate predicted by a decay constant of $\lambda = -0.023$ due to biological processes in combination with physical decay. Second, we investigated spatial variation in radiocaesium concentrations. Our two study sites included a low and high productivity site and risk of exposure can depend on the deposition densities of the fallout, and also environmental conditions such as soil chemistry or precipitation (Staaland et al., 1995). We predicted that plants at the low productivity site would have higher concentrations of radiocaesium partly due to a higher deposition density, and partly because radiocaesium, with its resemblance to potassium ions, is likely to have a higher bioavailability in nutrient poor soils. In addition, each study site had a series of sampling stations along an elevational gradient from ca. 800 to 1600 masl. If soil particles with adsorbed ^{137}Cs are transported downslope by runoff and soil erosion, we predicted that concentrations of radiocaesium might be higher at lower elevations (Skuterud and Thørring 2021). Last, we investigated variation in ^{137}Cs within and among different functional groups of lichens, bryophytes, and vascular plants. Lichens and bryophytes are perennial species with a large surface area that can absorb both nutrients and contaminants directly from air and precipitation (Bretten et al., 1992; Lehto et al., 2008). A low-lying growth form make these plants vulnerable to radioactive fallouts, especially in the short-term. In contrast, graminoids, herbs and woody plants with a well-developed root system might have lower uptake of radiocaesium during the first period after a fallout event but may have a more persistent uptake over time (Skuterud and Thørring 2021). In a few cases, radiocaesium concentrations can also differ among plant parts with either lower or higher concentrations in newly grown shoots or flowers (Clint and Dighton 1993). Contamination with radiocaesium remains an environmental challenge in many parts of the world (Zhu and Smolders 2000). Knowledge on plant uptake and differences among plant species are important for devising effective strategies to minimize the transfer not only from soil to wild plants, but also to wildlife, livestock, and agricultural products, and potentially to humans.

2. Methods

2.1. Study areas

The two study areas were located in mountain areas of mid-central Norway; Dørålen in Rondane national park (Folldal municipality, Innlandet, 62.02N, 9.90E) and Knutshø beside Dovrefjell national park (Oppdal municipality, Trøndelag, 62.28M, 9.60E). Both study areas had mountainous terrain with elevation gradients from low-alpine to high alpine vegetation zones, ranging from 810 to 1460 masl at Dørålen and 975 to 1685 masl at Knutshø (Fig. 1). The two sites are ca. 35 km apart, but plant productivity differed between the environments because of differences in the underlying bedrock. The study area in Dørålen is characterized by nutrient poor soils over a sparagmite bedrock, whereas the area at Knutshø has nutrient rich soils with a bedrock of phyllite and mica schist (Jordhøy 2008; Rekdal and Angeloff 2015). Differences in soil quality had effects on vegetative composition with a lower diversity of vascular plants at Dørålen than at Knutshø. The Chernobyl accident occurred on April 26, 1986, and some of the highest deposition rates for the fission product ^{137}Cs (>50 kBq/m^2) were found afterwards in mountain areas of central Norway (Tingstad and Nybø 2019). Based on soil samples collected on May 15, 1986 or three weeks after the disaster, the average levels of activity of ^{137}Cs in surface soils were 23.93 kBq/m^2 at Folldal close to the nutrient-poor area and 9.28 kBq/m^2 at Oppdal close to the nutrient-rich area, which were 1.3–3.4 × higher than the national average of 7.1 kBq/m^2 for Norway (Backe et al., 1986).

2.2. Field sampling

Plant samples were collected during the growing season over a 31-year period from 1991 to 2022. Samples were collected annually from 1991 to 2011, and then at 2–4-year intervals afterwards (2013, 2017, 2019 and 2022). Sampling at Dørålen was conducted at 12 stations between 810 and 1460 masl, with 81.9% of the samples collected at the five mid-elevation stations between 925 and 1110 masl. In contrast, sampling at Knutshø was conducted at 14 stations between 975 and 1685 masl, but 77% of all samples were collected at the lowest station at Grønnbakken. Observers collected samples from five major functional groups: macrolichens (18 taxa of fruticose and foliose lichens), peat moss (13 taxa of *Sphagnum*), mosses (11 taxa), herbaceous plants (18 taxa) and woody plants (14 taxa, mostly ericaceous shrubs, Table 1). Sampled species were important for grazing by herbivorous species, including ptarmigan (*Lagopus* spp.), wild reindeer (*Rangifer tarandus*) and domestic sheep (*Ovis aries*). Plants were collected by hand or by clipping with scissors.

2.3. Lab analyses

Plant samples were sorted and dried at 70 °C in a drying cabinet for >24 h (or until a constant weight) and then analysed for concentrations of ^{137}Cs. For lichens and bryophytes, samples were separated into old

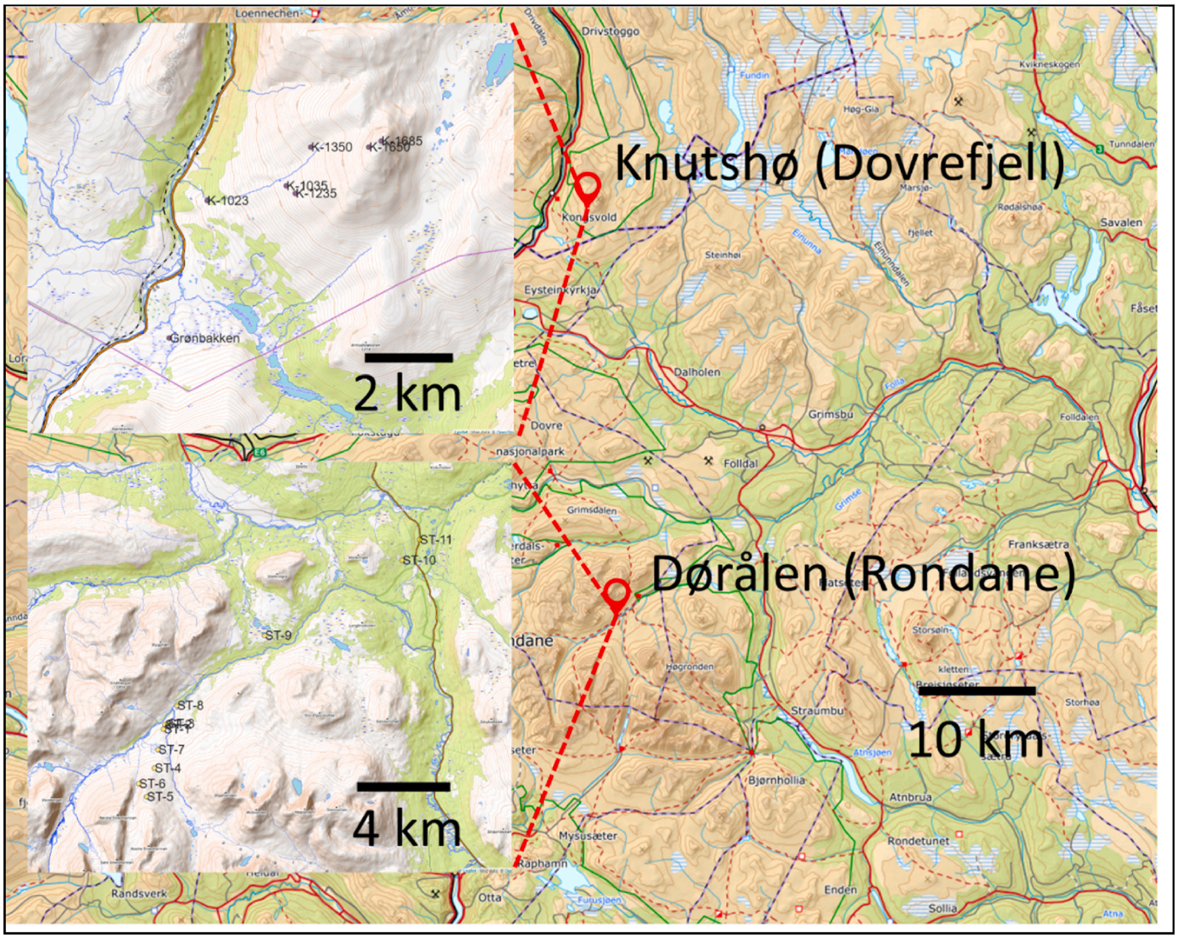

Fig. 1. Maps of the two study areas and sampling sites in mountain areas of central Norway. Dørålen is a nutrient poor area whereas Knutshø is a nutrient rich area due to geological differences in the underlying bedrock.

(dead) and new (living) plant parts when available, and these were analysed separately. However, in cases of low mass or where different plant parts could not be easily separated, this distinguishment was not done, and the material was analysed as a “total plant” sample. For vascular plants, samples were analysed from current year production including both leaves and stems. The only exception was heather *Calluna vulgaris* where leaves and stems (old parts of the plant) and new shoots were analysed separately.

From 1991 to 2010, laboratory analyses of radioactivity in the plant samples were conducted with a CompuGamma 1282 γ-spectrometer with a well detector (Kålås et al., 1994). From 2010 to 2022, all remaining analyses were performed with a Wizard 2480 γ-counter (PerkinElmer) at laboratory facilities of the Norwegian Institute for Nature Research (NINA). Concentrations of radioactivity for plant samples are reported in units of becquerel per kilo dry weight (Bq/kg dw). If measurements were below the detection limit of 20 Bq/kg, then the values were set to 20 Bq/kg for statistical analyses (Kålås et al., 1994).

2.4. Data preparation and statistical analysis

Our sampling design was unbalanced because field sampling was not conducted systematically. Observers collected different species for each functional group and sometimes multiple samples from individuals of the same plant species. Following our laboratory procedures, separate estimates of radioactivity concentration were also available for dead, live, or total plants depending on the species. In a first analysis, we compared differences in caesium concentrations among different plant parts in the five functional groups of plants. We then pooled samples from different plant parts to use all of our samples to investigate spatial and temporal variation in caesium levels. We observed that some plant samples had unusually high concentrations of ^{137}Cs up to 39.4 kBq/kg, which was consistent with precipitation of radioactive caesium as ‘hot particles’ (Nakamura et al., 2018). Thus, we opted to use the maximum value recorded in any of the plant tissues for each plant species per site and year as our response variable. We did not have sufficient samples to conduct separate analyses of long-term changes in caesium for multiple species of plants. Thus, we pooled species to examine broad patterns within the five different functional groups, and then conducted separate analyses for a representative species within each functional group. Representative species for the different functional groups included star-tipped cup lichen (*Cladonia stellaris*), stairstep moss (*Hylocomium splendens*), wavy-hair grass (*Avenella flexuosa*), and common heather (*Calluna vulgaris*). We were also interested in the potential effects of elevation above sea level on caesium concentrations in plants because alpine areas might experience greater rates of deposition. Here, we restricted our analyses to Dørålen because sampling effort was distributed among multiple sites across the elevational gradient, whereas a majority of sampling at Knutshø was conducted at one low elevation site.

All statistical analyses were conducted in an R Environment (ver. 4.0.3, R Core Team, 2020). We used a natural log transformation to normalize the distributions for measurements of ^{137}Cs concentrations in plant tissues and so that rates of decay could be estimated with linear models. Due to known differences in plant structure and physiology, we conducted separate analyses for each of the five different functional groups of plants. We analysed variation in radiocaesium concentrations

Table 1
Levels of radiocaesium in lichens, bryophytes and vascular plants sampled at Dørålen and Knutshø, Trøndelag in the 31-year period of 1991–2022. Radiocaesium levels (Bq/kg dw) in each species are given as median (range) with number of samples. Representative species for each group are indicated in boldface.

Group	Species	Dørålen	Knutshø
Macrolichens	*Alectoria ochroleuca*	534 (53, 7644), 24	573 (<20, 7253), 35
	Bryocaulon divergens	4504 (513, 21,068), 23	3690 (77, 12,874), 24
	Bryoria fuscescens	662 (75, 1377), 18	–
	Cetraria islandica	1028 (168, 10,459), 37	80 (80, 80), 1
	Cetrariella delisei	5321 (4,401, 6241), 2	–
	Cladonia arbuscula	1276 (<20, 12,845), 63	595 (<20, 6621), 33
	Cladonia rangiferina	405 (45, 6606), 29	200 (<20, 2703), 7
	Cladonia stellaris	786 (<20, 10,407), 135	1140 (62, 7766), 39
	Cladonia stygia	1191 (80, 1843), 12	1,539, 1
	Flavocetraria cucullata	2684 (145, 17,822), <20	490 (200, 756), 3
	Flavocetraria nivalis	1718 (166, 13,648), 55	1212 (<20, 9242), 30
	Lav sp.	4763 (4,113, 5412), 2	–
	Pseudephebe pubescens	9421 (5,287, 33,925), 15	–
	Ramalina sp.	158, 1	–
	Stereocaulon paschale	4858 (1,474, 39,408), 8	1417 (126, 6306), 24
	Umbilicaria sp.	2586 (2,372, 5795), 3	–
	Subtotal	1214 (<20, 39,408), 447	1221 (<20, 12,874), 197
Peat moss	*Sphagnum angustifolium*	2,420, 1	–
	Sphagnum capillifolium	415 (85, 1467), 8	714 (57, 8977), 20
	Sphagnum compactum	2418 (468, 2525), 3	–
	Sphagnum fuscum	3392 (218, 11,980), 13	1399 (489, 4398), 4
	Sphagnum girgensohnii	1792 (154, 3430), 2	–
	Sphagnum lindbergii	928, 1	–
	Sphagnum magellanicum	1423 (249, 2597), 2	–
	Sphagnum majus	1285 (549, 2020), 2	–
	Sphagnum rubellum	7528 (6,732, 8323), 2	<20, 1
	Sphagnum russowii	506 (358, 6906), 3	–
	Sphagnum sp.	527 (485, 568), 2	–
	Sphagnum teres	106, 1	–
	Sphagnum warnstorfii	–	84 (54, 114), 2
	Subtotal	1016 (85, 11,980), 40	674 (<20, 8977), 27
Mosses	*Barbilophozia floerkei*	2139 (<20, 7490), 14	–
	Hylocomium splendens	461 (<20, 2304), 29	287 (<20, 12,148), 32
	Pleurozium schreberi	841 (<20, 7372), 32	617 (101.97, 28,613), 15
	Subtotal	829 (<20, 7490), 75	411 (<20, 28,613), 47
Herbs and graminoids	*Alchemilla alpina*	404, 1	–
	Angelica archangelica	323 (214, 1175), 4	–

Table 1 (*continued*)

Group	Species	Dørålen	Knutshø
	Avenella flexuosa	248 (<20, 3943), 74	154 (<20, 658), 30
	Carex bigelowii	3157 (226, 18,699), 21	–
	Eriophorum angustifolium	389 (<20, 3032), 13	–
	Festuca ovina	–	<20, 1
	Oxyria digyna	–	178, 1
	Rumex acetosa	497 (485, 508), 2	–
	Rumex acetosella	616, 1	–
	Silene acaullis	–	439, 1
	Solidago virgaurea	237 (41, 3745), 25	514 (58, 2295), 25
	Subtotal	411 (<20, 18,699), 141	272 (<20, 2295), 58
Woody plants	*Betula nana*	89 (<20, 862), 61	109 (<20, 267), 35
	Betula pubescens	28 (22, 33), 3	193 (<20, 383), 21
	Betula tortuguosa	273 (<20, 441), 19	–
	Calluna vulgaris	1369 (54, 12,641), 61	670 (209, 1211), 16
	Empetrum hermaphroditicum	386 (<20, 2528), 38	96 (<20, 165), 5
	Empetrum nigrum	109 (<20, 498), 29	52 (<20, 189), 12
	Phyllodoce caerulea	818 (562, 1073), 2	–
	Pinus sylvestris	181 (<20, 466), 17	–
	Salix glauca	26 (<20, 81), 6	125 (<20, 1252), 14
	Salix herbacea	126 (<20, 850), 23	–
	Salix lapponum	152 (<20, 447), 19	130 (<20, 331), 18
	Salix myrsinifolia	–	20, 1
	Salix phylicifolia	29 (<20, 2506), 18	122 (<20, 507), 15
	Salix reticulata	–	128 (92, 163), 2
	Vaccinium myrtillus	546 (<20, 3193), 78	163 (64, 212), 6
	Subtotal	224 (<20, 12,641), 374	144 (<20, 1252), 145
Total		549 (<20, 39,408), 1077	304 (<20, 28,613), 474

with an analysis of covariance model (ANCOVA) with a categorical effect of area (Dørålen vs. Knutshø), a continuous effect of year (1991–2022), and an interaction between the two factors to test for a difference in slopes. The physical half-life of ^{137}Cs is $t_{1/2} = 30.17$ years and the expected decay constant (λ) was calculated as $\lambda = \ln(2)/t_{1/2} = -0.023$. Slopes from the regression models provided estimates of the rate of decay under field conditions, and we used a one-sample *t*-test for comparisons to the declines predicted from the decay constant ($\lambda = -0.023$). Similarly, we estimated the effective half-life from $t_{1/2} = \ln(2)/\lambda$, where λ was the slope coefficient from the log-linear plots. The effective half-life includes the effects of ecological (biological) processes and the physical decay of the radionuclide (IAEA, 2006). To illustrate the predicted rates of decay, we calculated the average concentrations of radiocaesium among the samples during the first year of the study (1991), and then used the decay constant to predict declines over the next three decades. In a last step, we used partial regression plots to model variation in concentrations of radiocaesium as a function of elevation at Dørålen. We controlled for the temporal changes by taking the residuals from a log-linear model of ^{137}Cs versus year, and then modelled the residuals as a function of elevation at the sampling site. To facilitate comparisons with annual rates of decay, we report changes in the $\log_e$ concentrations of ^{137}Cs per 100 m of elevation. In all models, coefficients of determination were taken as the adjusted r-squared value.

3. Results

3.1. Samples

Measurements of ^{137}Cs were recorded for a total of 1551 plant samples collected from 1991 to 2022 (Table 1). We measured caesium levels in both the live and dead parts of the plant for a subset of 683 samples. Mosses had significantly higher caesium levels in the dead plant parts (live minus dead ± 1SD: 906 ± 3040 Bq/kg, n = 89, $t_{88} = -2.81$, $P = 0.006$), and the pattern was similar in each of the three study species: *Barbilophozia floerkei* (−1060 ± 943 Bq/kg, n = 10, $t_9 = -3.55$, $P = 0.006$), *Hylocomium splendens* (−580 ± 1812 Bq/kg, n = 47, $t_{46} = -2.19$, $P = 0.033$), and *Pleurozium schreberi* (−1337 ± 4553 Bq/kg, n = 32, $t_{31} = -1.66$, $P = 0.11$). In contrast, caesium levels tended to be higher in live plant parts in the other four groups but the difference was not significantly different from zero in lichens (difference ± 1SD: 31 ± 1223 Bq/kg, n = 438, $t_{437} = 0.052$, $P = 0.60$), peat moss (408 ± 1696 Bq/kg, n = 57, $t_{56} = 1.82$, $P = 0.074$), herbaceous plants (146 ± 3166 Bq/kg, n = 42, $t_{41} = 0.30$, $P = 0.77$), or woody plants (318 ± 1663 Bq/kg, n = 57, $t_{56} = 1.44$, $P = 0.15$). The largest differences were observed in the lichen *Stereocaulon paschale* (1444 ± 2430 Bq/kg, n = 30, $t_{29} = 3.26$, $P = 0.003$), and in heather *Calluna vulgaris* (1042 ± 2853 Bq/kg, n = 18, $t_{17} = 1.55$, $P = 0.14$). Thus, we used measurements from the plant part with highest value or from the total sample to proceed with our next analyses of spatial and temporal variation in ^{137}Cs.

Plant samples with ^{137}Cs measurements below the detection limit of 20 Bq/kg in our γ−counters were more common in recent years but were a small proportion of our samples (4.6%, 71 of 1551 samples). On the

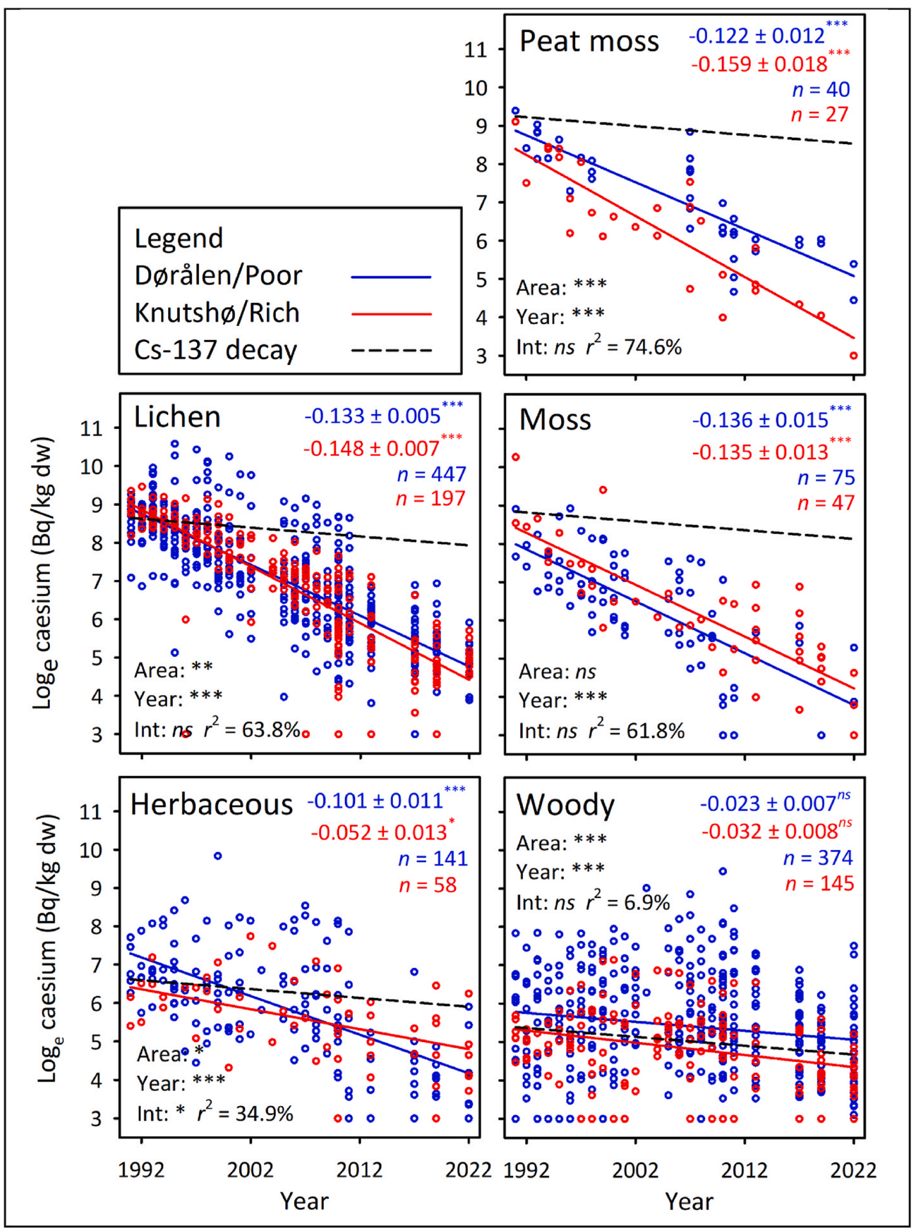

Fig. 2. Log-linear plots of declines in concentrations of radiocaesium in five functional groups of plants in a nutrient poor area (Døràlen, blue) and a nutrient rich area (Knutshø, red) in central Norway, 1991–2022. Analysis of covariance (ANCOVA) was used to test for effects of area, year, and their interaction (***$P < 0.001$, *$P < 0.05$, ns = $P > 0.05$, r^2 adjusted coefficient of determination). The Chernobyl disaster occurred in 1986 and the effective rate of decay in $\log_e$ caesium per year was estimated as regression slopes (±SE) and tested versus a physical decay rate of $\lambda = -0.023$ per year for ^{137}Cs (black dashed line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

other hand, the highest concentrations of ^{137}Cs recorded in the top 2.5% of samples ranged from 9.2 up to 39.4 kBq/kg dw. Most of the high concentrations were detected in samples from lichens (90.7%, 78 of 86 samples), especially two species of fruticose lichens (*Bryocaulan divergens* and *Pseudephebe pubescens*). In the first four years of the time series (1991–1994), radiocaesium concentrations were highest in lichens (mean ± 1SD: 6388 ± 3520 Bq/kg, n = 91), peat mosses (5918 ± 2981 Bq/kg, n = 11), and mosses (4616 ± 6662 Bq/kg, n = 16), but lower among herbaceous (1121 ± 939 Bq/kg, n = 28) and woody plants (379 ± 514 Bq/kg, n = 65, ANOVA: $F_{4,206} = 162.1$, $P < 0.001$). The differences among the functional groups persisted during the last four years of the time series (2019–2022) but average concentrations were <200 Bq/kg for all groups, including lichens (185 ± 157, n = 57), peat moss and mosses (123 ± 116, n = 19), herbaceous plants (138 ± 168, n = 29), and woody plants (161 ± 264, n = 81, ANOVA: $F_{3,182} = 4.14$, $P = 0.007$).

3.2. Plant functional groups

Analysis of covariance models with the effects of area and year explained more than two-thirds of the variation in radiocaesium concentrations in lichens and bryophytes ($r^2 = 61.8\%–74.6\%$) but less than a third of the variation for the vascular plants ($r^2 = 6.9\%–34.9\%$) (Fig. 2). The interaction term was significant only for herbaceous plants with a modest difference in slopes between the two areas. Concentrations of radiocaesium were significantly different between areas for 4 out of 5 functional groups but not for the mosses. In line with our predictions based on plant productivity, it was higher among plants at the nutrient-poor area at Døràlen versus the nutrient-rich area at Knutshø.

Concentrations of radiocaesium showed strong evidence of long-term declines during the study period from 1991 to 2022 among all functional groups of plants and areas. The rates of radioactive decay were greatest among the lichens (λ = −0.133 to −0.148) and the bryophytes (λ = −0.122 to −0.159), intermediate among the herbaceous plants (λ = −0.052 to −0.101), and lowest among woody plants (λ = −0.023 to −0.032). Apart from the woody plants, all the estimates for the rate of decay were significantly different from the rate of decay for ^{137}Cs predicted from a physical half-life of 30.17 years (λ = −0.023). The effective half-life of ^{137}Cs was estimated to be 4.4–5.7 years in lichens and bryophytes, 6.9–13.3 years in herbaceous plants, and 21.7–30.1 years in woody plants.

3.3. Representative plant species

We repeated the analyses with representative species of plants from each of four functional groups, with the exception of peat mosses where we did not have adequate samples to test an individual species of *Sphagnum* (Table 1). Comparisons of caesium concentrations of the selected representative species to other species in the same functional group showed that they were usually similar, with the exception of common heather *C. vulgaris* which had significantly higher concentrations of radiocaesium (1528 ± 1902 Bq/kg, $n = 77$) than the other species of woody plants (275 ± 381 Bq/kg, n = 442, two-sample *t*-test: $t_{109.5} = -13.97$, $P < 0.001$). Conditioning upon single species reduced some of the heterogeneity and the coefficients of determination were

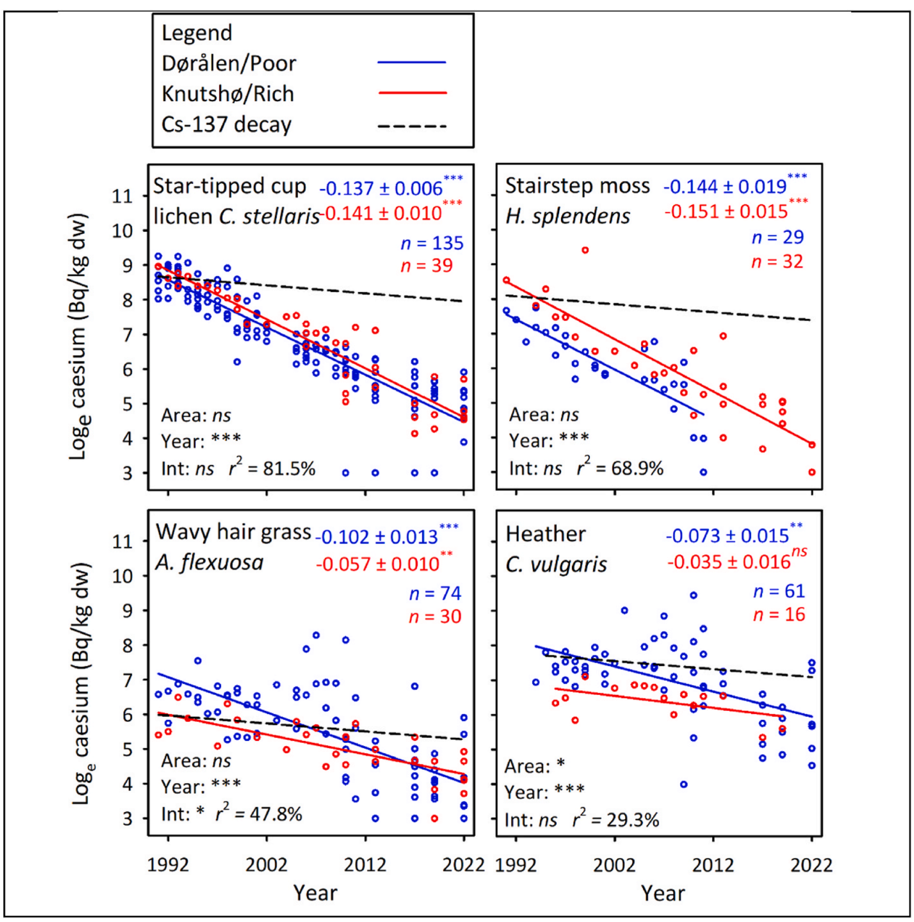

Fig. 3. Log-linear plots of declines in concentrations of radiocaesium in four representative species of alpine plants including star-tipped cup lichen (*Cladonia stellaris*), stairstep moss (*Hylocomnium splendens*), wavy-hair grass (*Avenella flexuosa*), and common heather (*Calluna vulgaris*) at a nutrient poor (Dørålen, blue) and a nutrient rich area (Knutshø, red) in central Norway, 1991–2022. See caption of Fig. 1 for description of statistical analyses. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

consistently higher than analyses based on the functional groups. Here, analysis of covariance models with the effects of area and year explained more than two-thirds of variation in radiocaesium in the lichen *C. stellaris* and the moss *H. splendens* ($r^2 = 68.9$–81.5%), and up to half of the variation in the two vascular plants *A. flexuosa* and *C. vulgaris* ($r^2 = 29.3$–47.8%, Fig. 3). Slopes differed between study areas for wavy hair grass but not for any of the other taxa because the interaction terms were nonsignificant. Concentrations of radiocaesium were significantly higher at Dørålen, the nutrient poor site, for *C. vulgaris* but not for the other three species. Rates of decay were highest among the lichen *C. stellaris* and the moss *H. splendens* ($\lambda = -0.137$ to -0.151), intermediate in *A. flexuosa* ($\lambda = -0.057$ to -0.102), and lowest in *C. vulgaris* ($\lambda = -0.035$ to -0.073). Overall, 7 of 8 estimates for the rate of decay were significantly different from the physical rate of decay of ^{137}Cs ($\lambda = -0.023$). The effective half-life of ^{137}Cs was ca. 5 years in the lichen and bryophyte, 6.9–12.2 years in the grass *A. flexuosa*, and 9.5–19.8 years in heather *C. vulgaris*.

3.4. Elevation

We used partial regression plots to investigate the role of elevation in explaining residual variation in radiocaesium concentrations after controlling for the effects of the long-term declines over the 31-year study period. The analysis of elevation was restricted to three functional groups at Dørålen where sampling effort was spread across the elevational transect. Elevation explained about a fifth of the residual variation in radiocaesium concentrations in lichens ($r^2 = 18.1\%$), but less than an eighth of the variation in the vascular plants ($r^2 \leq 12.6\%$, Fig. 4). The relationships between residual levels of radiocaesium and elevation were positive for all three functional groups, and the magnitude of the change in radiocaesium per 100 m of elevation ($\beta = +0.209$ to $+0.454$) were 2–9× greater than the annual rates of decay.

4. Discussion

Our project objectives were to investigate temporal, spatial, and taxonomic variation in ^{137}Cs concentrations in alpine plants in mountain areas of central Norway that were directly impacted by the Chernobyl accident in 1986. Our study design had good potential to detect long-term changes because the duration of our field project (31 years) was comparable to the physical half-life of ^{137}Cs (30.2 years). Our major results were three-fold. First, we found evidence for long-term declines in radiocaesium in alpine plants where the effective rates of decay were much faster than the rate predicted from the physical half-life. Nevertheless, detectable concentrations of radiocaesium were still present in alpine plants more than 35 years after the Chernobyl accident. Second, concentrations varied spatially with higher concentrations of radiocaesium at a low productivity site and in plants growing at higher elevations. Last, different functional groups of plants differed in concentrations with higher rates of contamination in lichens and bryophytes than vascular plants. Further, we also observed differences among various plant species important for grazing. Since plant herbivory is one of the main pathways for radioactive caesium to enter the food chain, new information on ^{137}Cs in alpine plants will help to understand the environmental impacts of radioactive contamination for other trophic levels.

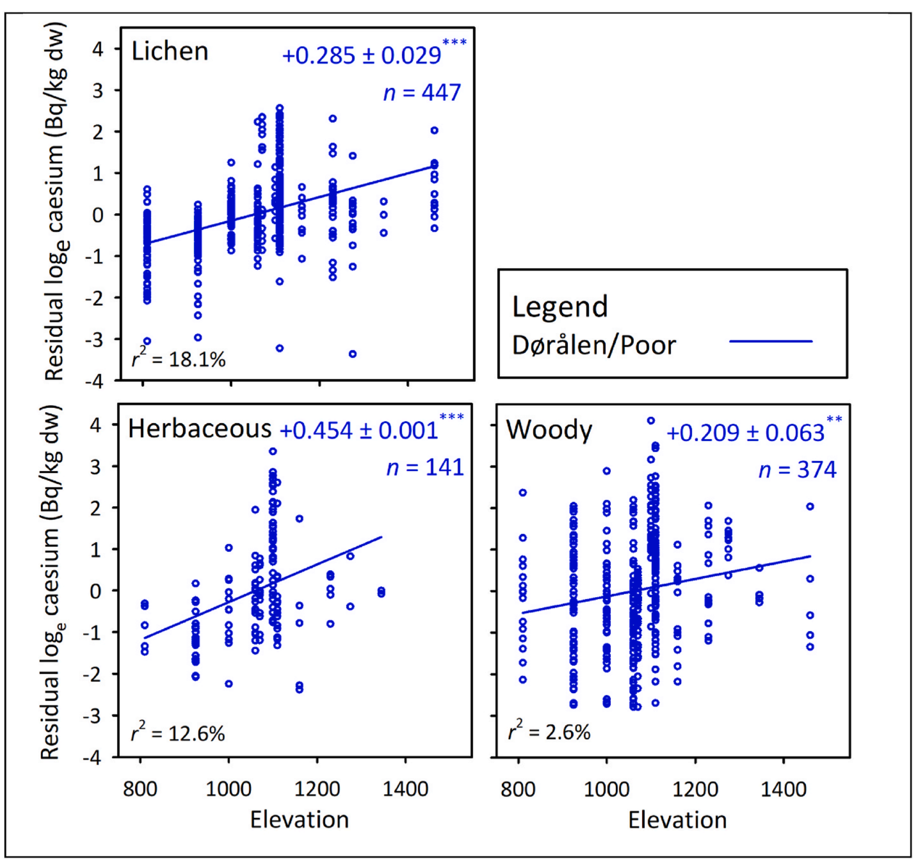

Fig. 4. Partial regression plots for concentrations of radiocaesium in lichens and plants across an elevational gradient at a nutrient poor area (Dørålen) in central Norway, 1991 to 2022. Residuals from the log-linear relationships of caesium and year were modelled as a function of elevation for three functional groups where samples were collected at sites between 810 and 1463 masl. Changes in $\log_e$ caesium per 100 m were estimated as the regression slopes for a log-linear plot of residual caesium versus elevation (±SE).

4.1. Long-term declines in concentrations of Cs-137 in alpine plants

Our long-term study revealed steep declines in the concentrations of ^{137}Cs over the last three decades, with similar patterns among different study areas and plant functional groups. The Chernobyl accident was the last major event releasing radiocaesium into northern environments and our measurements have tracked reductions in contamination since 1991. We found that lichens and bryophytes contained much higher concentrations of ^{137}Cs during the first years of sampling shortly after the Chernobyl accident. High values were likely related to fallout deposited directly on the bodies of lichens and bryophytes. Due to their structure and physiology, nutrients and other substances from air and precipitation can be absorbed by cells on the plant surfaces. Because of these characteristics, lichens and bryophytes are often used as bio-indicators for atmospheric pollutants, including radioactive fallout (Augusto et al., 2013). In our study, lichens and bryophytes must quickly have absorbed fallout after Chernobyl and the radioactive material remained within the cells of the slow-growing organisms. However, the concentrations of radiocaesium also showed rapid declines that were significantly faster than predictions based on the physical half-life time of 30.2 years. In fact, our measurements of ^{137}Cs in plant tissues showed that the effective half-life was around 4–6 years in lichens and bryophytes, comparable to estimates of 3–6 years for lichens at other sites in Europe (Cevik and Celik 2009, Machart et al., 2007). Rapid decay and a short effective half-life were likely due to replacement of plant parts with new growth of fresh uncontaminated material, consumption of plant material by herbivory, and also physiological responses to desiccation (Varskog et al., 1994). Lichens and bryophytes are poikilohydric organisms and changes in cell membrane permeability during drying and rewetting cycles can cause leakage of potassium and caesium ions (Green et al., 2011).

Vascular plants differ from lichens and bryophytes because they have different growth forms and obtain most of their nutrients from the roots. Hence, concentrations of radiocaesium in the first years after the Chernobyl accident were lower in vascular plants than lichens and bryophytes, but interestingly, the decay rate over time was also slower. Estimates of the effective half-life of 7–13 years for herbaceous plants in Norway were comparable to estimates of 8–17 years for pasture grasses in Europe (Corcho-Alvarado et al., 2016; Brimo et al., 2019). Furthermore, woody plants had a slower decay rate of radiocaesium with an effective half-life of 22–30 years – which was not much different from the physical half-life of ^{137}Cs at 30.2 years. Radiocaesium can remain in the soil for a long time and has been found mostly in organic soils and in the upper part of the soil profile (Varskog et al., 1994; Yoshida et al., 2004; Almgren and Isaksson 2006). Differences in root structure, physiology and mycorrhizal associations may make radiocaesium more bioavailable to woody than herbaceous plants, although some radiocaesium may be immobilized in the soil and hence not be available for plant growth (Corcho-Alvarado et al., 2016).

4.2. Spatial variation in levels of Cs-137 in alpine plants

Many factors influence bioavailability and plant uptake of nutrients at a given site, including soil properties, precipitation, and features of plant morphology and physiology (Broadley and Willey 1997). In our study, we compared two mountain sites in central Norway and found that concentrations of radiocaesium were generally higher among alpine plants at a nutrient poor site (Dørålen) than a nutrient rich site (Knutshø). The site difference was consistent among lichens, peat moss and vascular plants, but was not found in *Sphagnum* peat mosses. The variation among sites was likely linked to differences in soil type and nutrient availability but could also relate to the initial geographical pattern of deposition, and greater deposition rates at higher altitudes in general. The deposition was much higher near the nutrient poor site which explain some of the observed variation between our two sites. It could also be that bioavailability of radiocaesium, being directly influenced by soil properties such as K^+ status and clay content, also explain a part of the observed variation (Absalom et al., 2001). As a free ranging ion (Cs^+), radiocaesium has similar properties as potassium ions (K^+) and can be taken up by the plants through the same biochemical pathways (Zhu and Smolders 2000). Hence, the potassium levels of soils may influence the uptake of radiocaesium in plants. This mechanism could be an explanation for our findings that radiocaesium uptake was higher on nutrient-poor than nutrient-rich soils, as has been shown in previous studies (Varskog et al., 1994; Zhu and Shaw 2000). On the other hand, high levels of potassium can suppress radiocaesium uptake in forest (Rosén et al., 2011) and agriculture systems (Zhu and Shaw 2000), because plants generally discriminate against Cs in favour of K (Salt and Mayes 1993).

Radiocaesium concentrations increased across the elevational gradient from 800 to 1500 m at Dørålen, especially among lichens and herbaceous plants. Higher concentrations of radiocaesium at high elevations could be explained by higher deposition rates in alpine habitats, or a longer effective half-life time due to a colder climate and shorter growing season. For example, monitoring of air dose rates near Fukushima, Japan, showed higher deposition rates of radiocaesium on ridges and without significant downslope migration (Atarashi-Andoh et al., 2015). Also in Norway, a study by Bretten et al. (1992) found that plants on ridges and outcrops had higher concentrations of radiocaesium, which was consistent with the initial deposition pattern from Chernobyl. Other field studies in Europe have also found evidence for elevational gradients with higher concentrations of radiocaesium among lichens and plants on summits or ridgetops. During the first years after the Chernobyl accident, lichens and plants growing at higher altitudes also had higher concentrations in Poland (Seaward et al., 1988) and Austria (Heinrich et al., 1999).

4.3. Individual plant species

In addition to the study of the plant functional groups, we evaluated four representative species including star-tipped cup lichen (*Cladonia stellaris*), stairstep moss (*Hylocomnium splendens*), wavy-hair grass (*Avenella flexuosa*), and common heather (*Calluna vulgaris*). Results for individual species were similar to their respective groups, with the exception of *C. vulgaris* which showed higher concentrations of radiocaesium than other woody plants. Heather is important for browsing by ptarmigan, wild ungulates, reindeer, and domestic sheep, and is therefore a plant of considerable interest in studies of the long-term contamination of radiocaesium. The high concentrations of ^{137}Cs in heather in our study were consistent with past comparisons of radiocaesium among different vascular plants (Fawaris and Johanson, 1994). Contrary to expectations from dilution, we found that the new shoots of *C. vulgaris* had a higher concentration of radiocaesium than older plant parts, which was consistent with previous reports (Clint and Dighton 1993). Heather appears to be unusual among vascular plants because experimental studies in the UK have also demonstrated that this plant species has particularly high uptake rates for radiocaesium (Clint and Dighton 1993; Salt and Mayes 1993). The physiological mechanisms are not fully understood but are presumably related to the mycorrhizal fungus associated with the root systems of *C. vulgaris* and other ericaceous plants (Strandberg 1994).

5. Conclusion

Early investigations of radiocaesium in plants in Europe focused on grazed pastures (Corcho-Alvarado et al., 2016; Brimo et al., 2019) and agricultural crops on cultivated lands (Varskog et al., 1994; Zhu and Shaw 2000). Our field study contributes new estimates for the levels and dynamics of radiocaesium among wild plants in natural systems. Estimates of radiocaesium for plants in natural habitats have been a knowledge gap for models of exposure for wild and domestic animals (Åhman 2007). Our results are encouraging because current estimates of radiocaesium concentrations in alpine plants in central Norway (<200 Bq/kg) are less than national guidelines for acceptable concentrations of radiocaesium in wild game and reindeer (370–3000 Bq/kg, DSA Norwegian Radiation and Nuclear Safety Authority, 2021). Understanding the implications of current concentrations and trends in radiocaesium contamination will require better estimates of transfer factors among soils, lichens, plants and consumers (Skuterud and Thørring 2021).

CRediT authorship contribution statement

Lise Tingstad: Conceptualization, Data curation, Methodology, Project administration, Visualization, Writing – original draft, Writing – review & editing. **Brett Sandercock:** Conceptualization, Data curation, Formal analysis, Visualization, Writing – original draft, Writing – review & editing. **Signe Nybø:** Conceptualization, Supervision, Writing – original draft.

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

Data will be made available on request.

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