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Translocation and plant availability of radio caesium in an organic soil

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Abstract

^{134}Caesium chloride solution was injected into the surface of a peaty podzol at 3 cm depth with 5 cm spacing over a M^2 at two upland sites, one of which had been fertilized and reseeded. The movement of radio caesium in the soil was subsequently followed by coring and/or taking out 10-cm square blocks at monthly intervals over a period of 2 years. There was very little movement of the caesium down the profile, with more than 95% remaining at the point of application. Lateral movement was also minimal. The caesium did not concentrate in the roots. Samples of herbage collected from the surface during the growing season showed a progressively decreasing concentration of caesium over the period. Total caesium removed in herbage amounted to 3.5 and 0.6% of the original present for the improved and unimproved soils, respectively.

The extractability of the caesium from the ^{134}Cs-amended peaty soil was compared with that from a low-ash peat which had been treated with the radionuclide for 2 months. Initially 1 *M* ammonium acetate (pH 7.2) extracted 30 and 56% of the Cs from 0–3 and 3–6 horizons of the improved peaty podzol, respectively. After 5 months only about 12 and 7% of the Cs was extracted by 1 *M* ammonium acetate and 4 and 1% by 0.1 *M* sodium pyrophosphate from the 0–3 and 3–6 cm horizons, respectively. After 24 months 1 *M* ammonium acetate extracted 1.6 and 3.3% of the Cs from the 0–3 and 3–6 cm horizons, respectively. In contrast, the values for the peat after 2 months were 100 and 80% for 1 *M* ammonium acetate and 0.1 *M* pyrophosphate, respectively.

Introduction

Contamination of upland soils by radio caesium as a result of the Chernobyl disaster in 1986 is still a problem because of the continuing availability of the Cs to plants and thence, via sheep and other animals, its subsequent movement into the human food chain. Models which describe the behaviour of caesium in lowland pastures have been found to be inappropriate for hill and upland agricultural systems, mainly because the cycling of caesium in organic soils is different from that in mineral ones: caesium in organic soils is not fixed, but neither is it rapidly leached (Livens and Loveland, 1988). Cawse and Horrill (1986) observed that about 90% of the caesium derived from bomb fallout deposited over the previous years remained in the top 15-cm of two peats. Despite this, peaty soils are considered to be in a low immobilisation class, and peat soils in a very low one (Livens and Loveland, 1988). Soil maps in conjunction with physico-chemical factors have been used to predict Cs behaviour in soils, but really quantitative data are lacking. This study seeks to provide such data for a peaty podzolic soil.

Materials and methods

Caesium

Virtually carrier-free radioactive ^{134}caesium as CsCl was obtained from Amersham International. This source contained less than 0.6 μg ^{133}Cs/Mbq ^{134}Cs and enabled quantities of Cs to be used as small as those occurring in polluted soils. In this way, differences in behaviour, when large quantities are present, were avoided. ^{134}Cs was used because of its relatively short half-life (2.05 y). The activity of all samples was determined using 5 ml-capacity polythene vials and a three-inch NaI (Th activated) crystal in a Packard automated 500 instrument. All results are corrected for decay and inefficiencies of counting.

Field sites

Two experimental sites were established on a peaty podzol of the Gaerlie series, Strichen Association (Glentworth and Muir, 1963), described as a Placic Podzol by FAO nomenclature. One had been limed (pH 5.0 in water), fertilized and reseeded with ryegrass and clover (grid reference NO 660 803) and was receiving N, P and K at a rate of 116, 22 and 46 kg ha^{-1} y^{-1}. The other, which had a shallower peaty horizon (pH 3.8 in water), was unimproved (grid reference NO 663 795), but the old heather had been burned 6–8 years previously and the area subsequently managed by sheep grazing. The vegetation was primarily grasses (*Agrostis*/*Festuca* species) with small areas of regenerating heather (*Calluna vulgaris*). Both were sited at the Glensaugh Research Station, North East Scotland, on sloping ground. The ash contents were 14–16% in the surface horizons.

Laboratory experiments were also carried out using a low-ash peat of the Hatton series from the Hill of Fisherie (NJ 820 585). The ash content was 1.5%.

Experimental procedures

In July, 1988, 6105 K Bq ^{134}CsCl were applied to a 1-m^2 area by the injection of four hundred 5-mL aliquots of solution evenly spaced using a specially constructed perspex grid which bridged the plot. The injection was made at a depth of 3 cm into the peat surface using a specially constructed syringe system which allowed outflow from the side of the needle rather than the tip (C A Salt and R W Mayes, personal communication).

The profile was sampled periodically by coring with a graduated copper tube (i.d. 20 mm) taking 3-cm-long cores from increasing depths in succession to avoid the effects of compaction. In addition, blocks of surface area 10 × 10 cm of the whole profile were taken out to obtain similar horizon material over a larger area. The holes were refilled by blocks of unlabelled material from an adjacent site. Cores were taken from soil 20 and 40 cm distant from the side of the treated area. Control samples were taken from profiles 2.5 m from the treated area.

Herbage was harvested from the site by cutting the entire sward to a height of about 2 cm to avoid contamination by soil. Plant materials were dried at 37°C in a ventilated drying unit. Some separation of plant material into heather, clover and grass species was made in the laboratory, usually after drying. One-gram amounts of plant material were packed in 5-mL polythene vials for counting. Soil cores were also dried at 37°C and lightly crushed, and samples of known weight placed in similar vials.

Soil blocks were carefully trimmed to size to remove material displaced by abstraction and divided into 3-cm-depth horizons using a saw blade and palette knife. The material from the layers was partially dried at 20°C and peat from the rooting zone was separated by sieving using a 2-mm screen. Other horizons were homogenized by manual fragmentation through a polythene bag. Surface plant material and the separated root matter from the 0–3 cm horizon were combined. One-gram samples of peat and plant matter were placed in vials for determination of ^{134}Cs activity.

Resin bags were prepared by enclosing 3.00 g Duolite C255H, 14–30 mesh ion exchange resin in 50 × 60 mm bags made from nylon fabric with 200 micron openings ('Nytal 7-200', supplied by A J Polak, Willesden, London, UK). They were washed with 0.5 *M* HCl and subsequently with water until the washings were chloride-free. They were kept moist until used.

Resin bags were inserted horizontally at different depths in the soil profile by removing a 20 × 20 cm block of soil on the down-slope side of the plot and cutting small slots with a knife into which the bags are inserted. The intact block of soil was replaced and the area firmed. Bags were replaced at monthly intervals into the same slot. During 1990 a second site adjacent to the first was also used for resin burial. The resin bags were washed free of soil particles, dried and placed in vials for ^{134}Cs counting.

A sample of Hatton peat held in an 18-cm diameter pot was injected with 14.8 G Bq ^{134}Cs at a depth of 3 cm and left for two months in an open-sided enclosure with a glass roof. The peat was watered regularly from the surface.

Extractions

Extractions were carried out on soil which was sampled from the reseeded plot at different times after application of ^{134}Cs. The soil was fresh or partially dried and was sieved to pass a 7-mm or 2-mm screen, respectively.

Extractions were made with 1 *M* ammonium acetate, pH 7.2, 0.1 *M* sodium pyrophosphate, pH 9.4, 5.3 *M* HCl or HF in centrifuge bottles using various ratios of solution to soil. The mixtures were shaken for 16 h on an end-over-end shaker. Soil was separated from solution by decanting the supernatant after centrifugation at 1200 *g*. Supernatants were further centrifuged at 2000 *g* to remove traces of sediment remaining. When multiple extractants were used where much solution was occluded by the soil residue, corrections were made by calculation based on the additional weight. Supernatant solution and residues were analysed for ^{134}Cs.

Extractions with resins were made using bags prepared in the same way as for field use with 1 bag (3 g resin) to 20 g soil or peat and 200 mL water. After 16 h on an end-over-end shaker the bags were recovered and washed free of soil or peat particles, dried and placed in vials for counting.

Soil solution was obtained by centrifuging field-moist soil at 2000 g.

Results and discussion

Distribution of ^{134}Cs at the field sites

Analysis of the cores showed that most of the ^{134}Cs was present within the upper 0–6 cm horizon close to the point of application, but there was considerable variation in the total ^{134}Cs content of the cores indicating a non-uniform distribution. This shows that vertical and lateral movement of the injected ^{134}Cs had been minimal. The distribution of ^{134}Cs within different horizons of the profiles as found by core sampling over a period of 18 months is shown in Fig. 1a and b.

Further evidence of the absence of lateral movement within the peat was obtained by analysis of cores taken to the sides of the plots at distances of 20 and 40 cm. These showed levels of ^{134}Cs activity comparable with those from a control area 2–3 metres away. However, there were higher levels of activity in the 0–3 cm horizons values from cores taken downslope from the plots at each site (Table 1). Statistical analysis was performed on samples collected over a period of 6 months since there appeared to be little change with time within that period. The mean levels of activity found in the surface horizons, of 14.4 and 37.7 Bq represent 0.75 and 2.0% of the activity present in cores sampled from within the plots in the improved and rough grazing areas, respectively.

Although the results are very variable it does appear that a substantial amount of Cs has moved down the slope at the surface. The most probable explanation for the movement of ^{134}Cs is a surface-hydrological effect, caesium being released from the herbage. Almost all the Cs in freeze-dried green grass plants is rapidly released by suspension in water.

The vertical distribution of ^{134}Cs within different horizons of the soil profile was best measured by analysis of the soil blocks because of their large area. Table 2 shows values for blocks sampled in October 1989 and July 1990, 15 and 24 months after application in two sites. They confirm the distribution data obtained by coring and show that there is very little downward movement of the ^{134}Cs.

Amounts of ^{134}Cs present in the vegetation

Table 1. ^{134}Cs activity (Bq) in 3 × 2 cm diameter soil cores taken from outside the treated plots

	Depth(cm)	Distance from plot edge(cm)				
		20	40	20	40	2500
		Side		Downslope		Reference
Improved pasture	0–3	2.533**	1.838	14.423*	8.384**	1.497
	3–6	1.545*	0.870	2.038*	2.878*	0.753
	6–9	1.588*	0.660	1.600	1.760*	0.875
	9–12	1.118	0.752	1.655	1.660	0.902
Rough grazing	0–3	2.264	1.370	37.68*	11.96	3.408
	3–6	1.808	1.563	11.03	3.408	1.800
	6–9	1.562	2.017	3.228	2.020	4.307
	9–12	1.480	1.400	NS	NS	2.615

Calculated 't' value significant at $p = 0.05$ (*).
$p = 0.01$ (**) in relation to reference values.
NS = no sample.

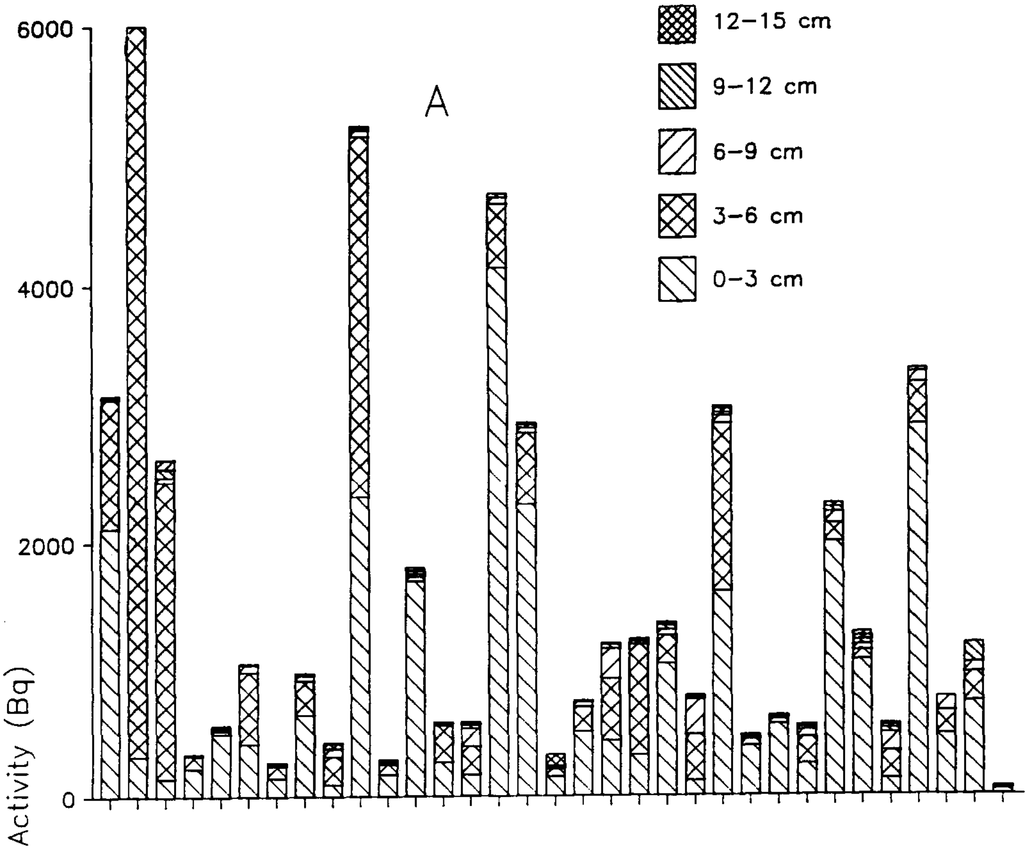

Fig. 1. ^{134}Cs activity in soil cores from **A**) improved pasture and **B**) rough grazing profiles. The total activity is sub-divided to show the activity in each consecutive horizon.

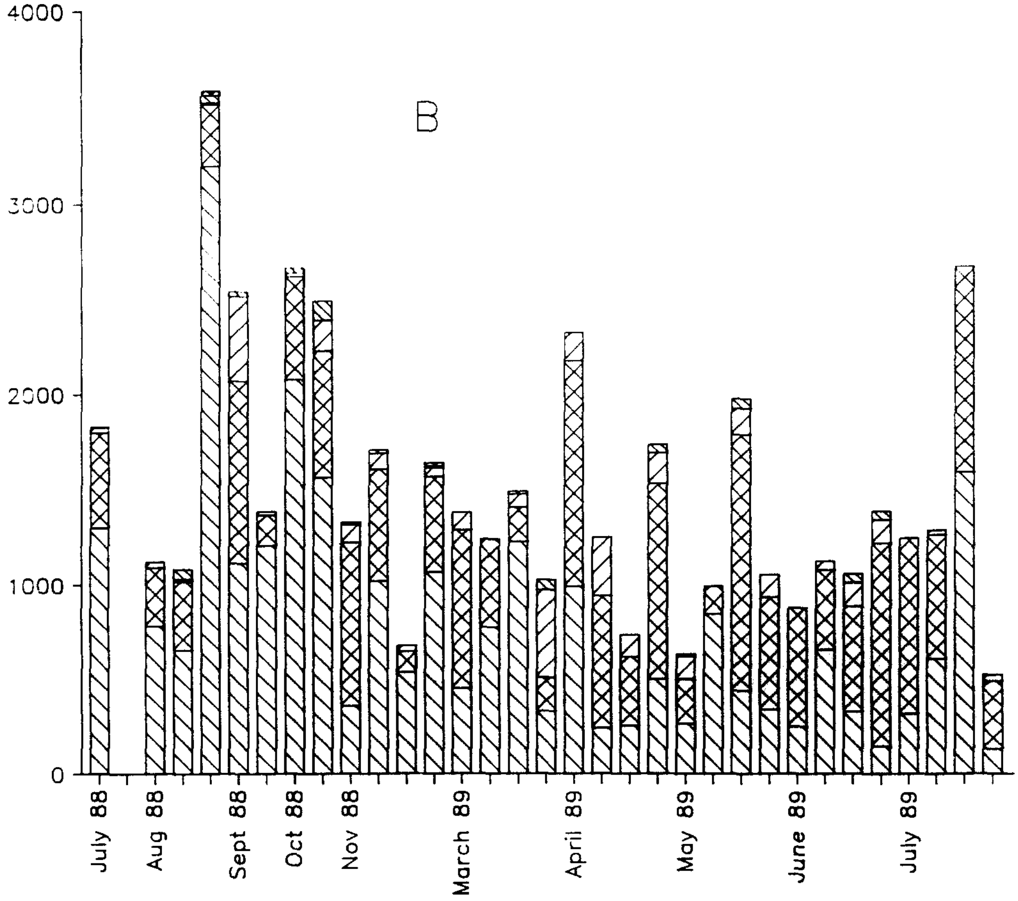

Fig. 1 (*continued*).

after various periods are shown in Table 3. In June 1989 there was no difference in the specific activity of the grass and clover on the improved pasture, whereas in July and subsequent months clover had a higher value. The values over the period of two years provide evidence of a progressive decrease in availability of the Cs to plants (Fig. 2). This is reflected in the progressive decrease in Cs released to buried resin (Fig. 3). Since the resin bags were being replaced in the same position in the soil there was the possibility of the decreasing uptake value being the result of localized depletion. This was shown not to be the case with bags placed in fresh sites showing similarly low release values. The uptake by the herbage was 1.53% of the total Cs in the first part-season, 1.58% in the second, and 0.4% in the third, a total of 3.5%.

The profile on the rough grazing site was much shallower and wetter. The distribution of ^{134}Cs within the profile determined by coring was again variable but indicated a slightly greater downward movement than on the reseeded peaty podzol (see Fig. 1b). This was not apparent from the analysis of the blocks (Table 2). The concentration within the grass was lower, and yields were smaller (Table 3). There was a significant difference in specific activity of grass and heather on this site, with heather being more active. The cumulative activity of ^{134}Cs removed by the herbage from this site represented only 0.64% of the activity applied.

Distribution of ^{134}Cs on untreated Hatton peat

The movement of ^{134}Cs in the peat during the two months was greater than in the peaty soils, with 12% reaching the 6–9 cm layer. The 9–12

Table 2. Distribution (%) of ^{134}Cs activity in 10 × 10 cm soil blocks sampled during October, 1989 and July, 1990, 15 and 24 months after application, respectively

Horizon	15 months		24 months	
	%	SE	%	SE
Peaty podzol, improved pasture				
Root mat[a]	5.99	0.364	7.32	0.264
0–3 cm	54.28	11.50	49.17	10.85
3–6 cm	36.78	11.01	36.72	11.73
6–9 cm	2.13	0.238	3.20	0.500
9–12 cm	0.45	0.081	1.13	0.140
12–15 cm	0.38	0.102	1.18	0.506
15–18 cm	–	–	0.88	0.205
Initial activity (%)[b]	97.21	5.73	93.5	4.24
Peaty podzol, rough grazing				
Root mat[a]	7.06	0.118	13.23	0.259
0–3 cm	40.84	1.65	32.13	5.70
3–6 cm	50.00	1.16	51.25	5.61
6–9 cm	1.90	0.707	3.06	1.09
9–12 cm	0.21	0.205	0.31	0.00957
>12 cm	0.00		–	–
Initial activity (%)[b]	72.42	5.53	92.05	3.36

[a] Surface root mat containing inseparable soil.

[b] Refers to total ^{134}Cs activity found in blocks compared with amount applied in July, 1988.

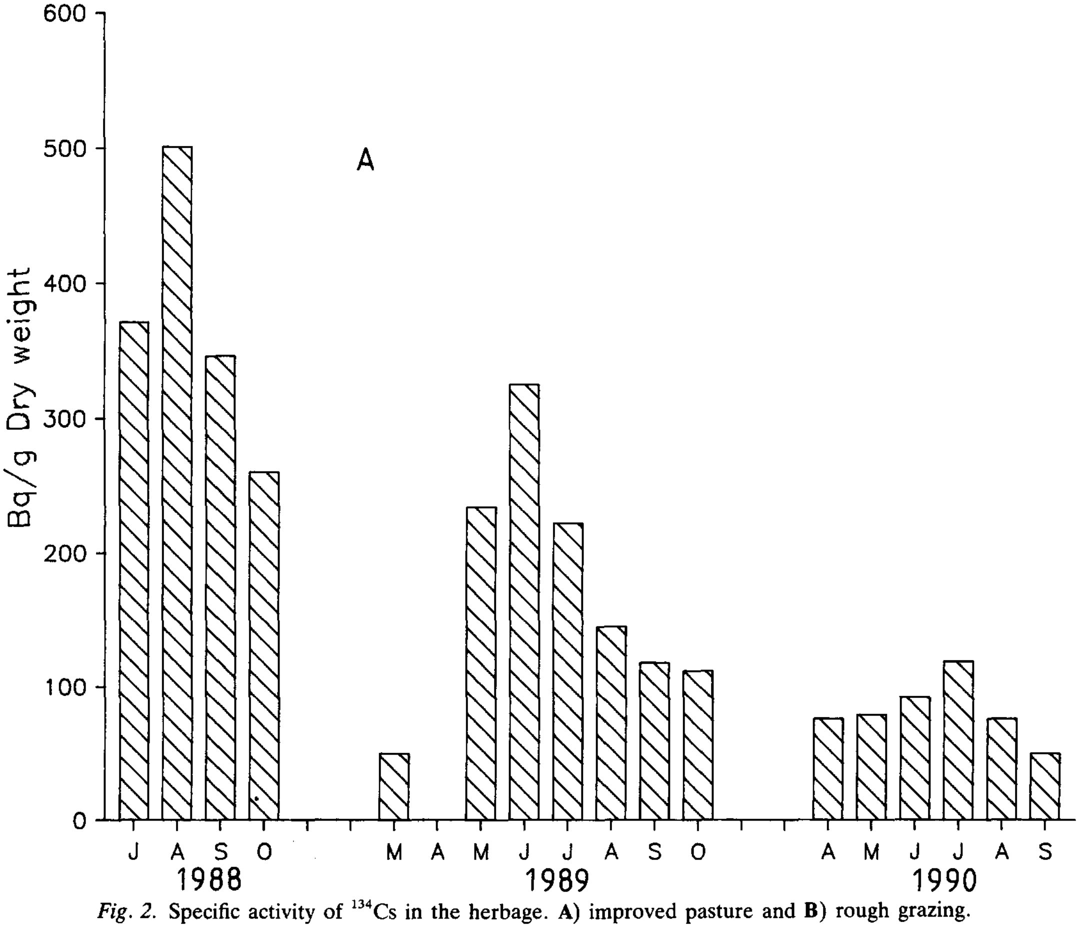

Fig. 2. Specific activity of ^{134}Cs in the herbage. **A**) improved pasture and **B**) rough grazing.

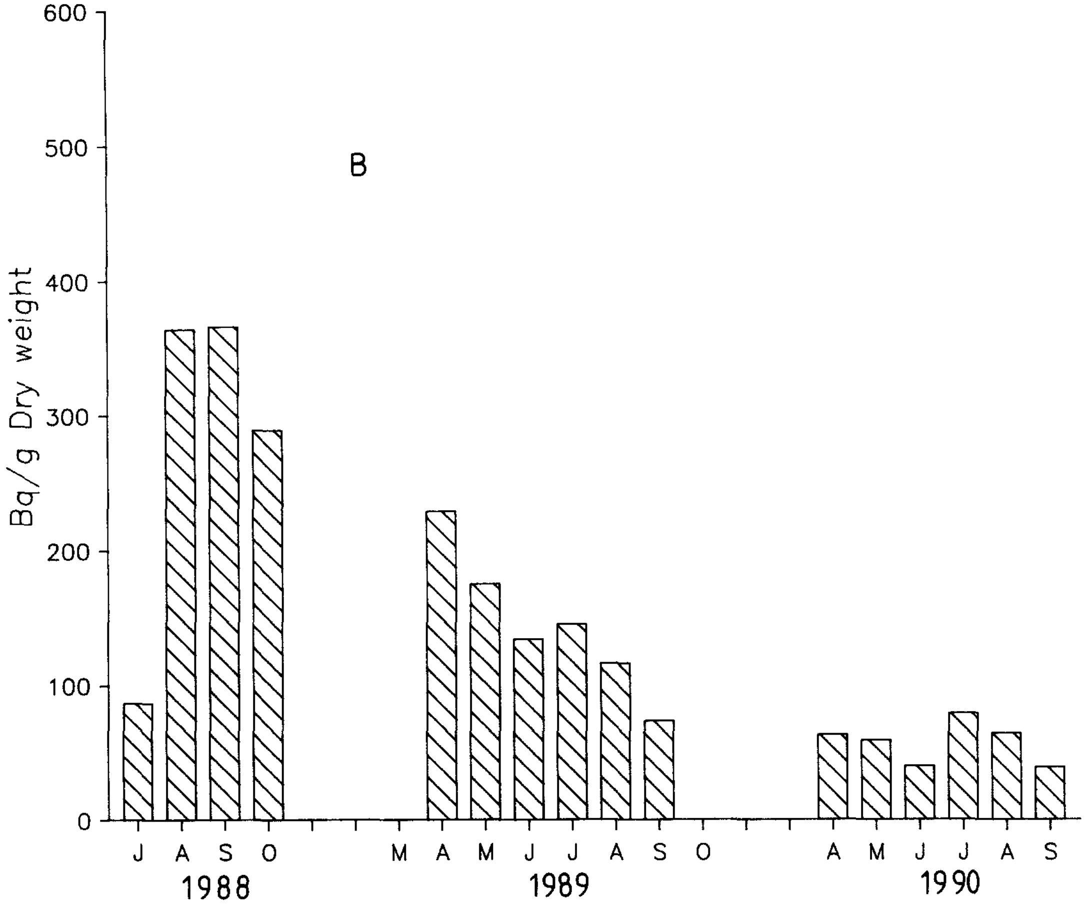

Fig. 2 (continued).

and 12–15 cm layers had 1.9 and 0.4%, respectively. The 3–6 cm layer of the peat, which contained 24% of the caesium, was used in extraction experiments.

Extraction experiments

Three days after the addition of Cs to the improved peaty soil ammonium acetate removed 30 and 56% of the Cs from the 0–3 and 3–6 horizons, respectively. After 5 months only a small proportion of the ^{134}Cs present in the 0–3 or 3–6 horizon of the improved pasture soil was released by extraction with water, ammonium acetate, sodium pyrophosphate or by treatment with resin (Table 4), and the ammonium acetate values became progressively smaller with time. The ammonium acetate and resin would be expected to release exchangeable forms, whereas the pyrophosphate should, in addition, release complexed ions or those associated with humified organic matter. In contrast, with the Hatton peat all ^{134}Cs was exchangeable, and almost 80% was released by sodium pyrophosphate. The difference between the peat and soils is probably related to the contrasting mineral contents. The Hatton peat has a very low ash content which has a composition commensurate with plant ash. The ash content of the peaty podzol indicated the presence of structured silicate mineral components and some of these may have forms capable of fixing Cs. Although treatment of the ^{134}Cs-rich horizon material of the improved peaty podzol with 20% HF (100 mL g^{-1}) for 5 days to destroy silicate released 58% of the ^{134}Cs, an equivalent treatment with HCl of similar concentration released almost the same amount (56%), suggesting that it was the acidity of the reagent that was effective. On the other hand, it is difficult to believe there

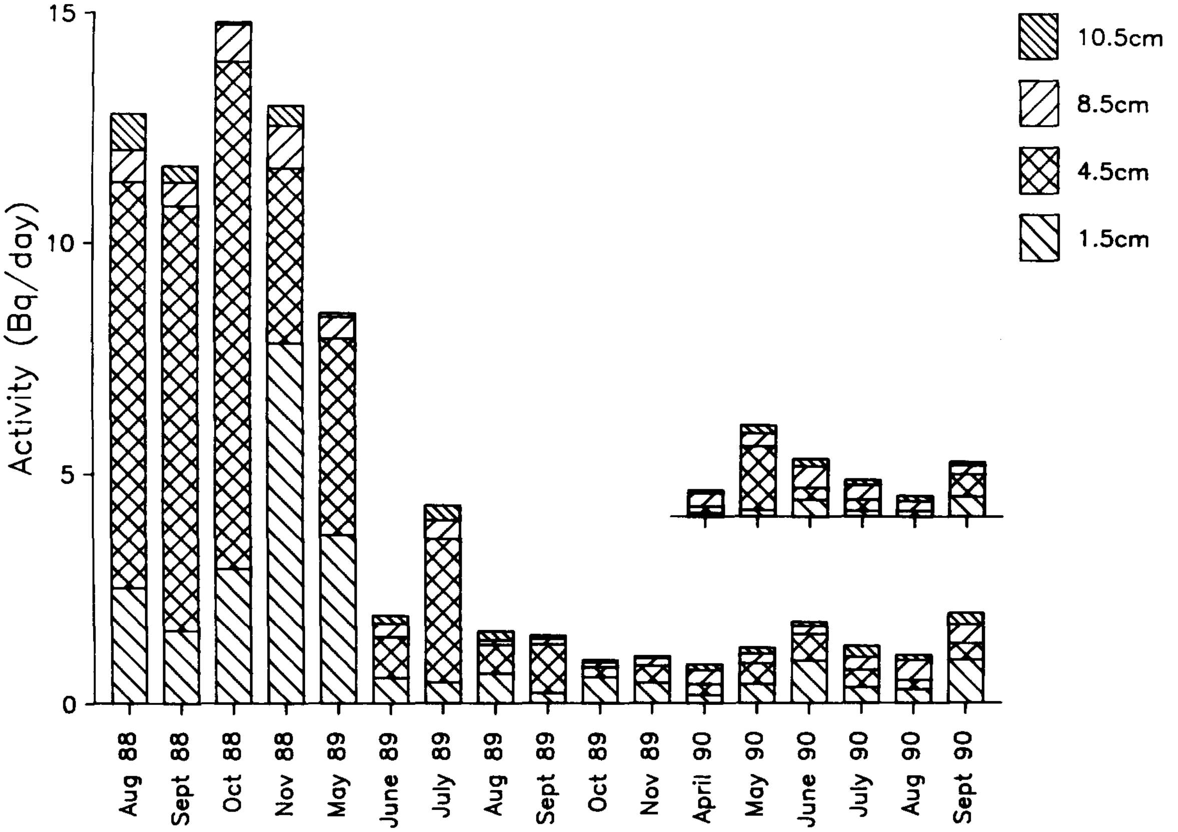

Fig. 3. ^{134}Cs activity (Bq day^{-1}) in resin bags buried horizontally at different depths for 1 month in improved soil. *Inset* shows activity in resin bags buried in a fresh, adjacent position from April '90.

are differences in the organic matter of the two types of soils sufficient to explain the very different extractabilities observed for Cs, because this element forms relatively few organic complexes.

Dunigan and Francis (1972) found that ^{137}Cs sorbed by a humic acid from a silt loam was strongly retained. Other studies have observed association between organic matter and ^{137}Cs content or retention (Cox and Frankhauser, 1984; de Jong et al., 1986; Sheppard et al., 1980), but interpretation of the results is usually equivocal because of the association of mineral and organic-matter fractions (Maulé and Dudas, 1989; Wilkins et al., 1985).

It is envisaged that most of the added Cs would initially occupy exchange sites on the soil organic matter from which it would be readily available to plants. With time, under thermodynamically controlled processes, the Cs would gradually move to less available sites which may include fixed forms on soil minerals and organic matter.

Conclusion

Very little movement of ^{134}Cs occurred down the profile of a peaty podzol over the period of 24 months after application. The proportion of the Cs which was exchangeable as judged by extractability with 1 *M* ammonium acetate and cation exchange resin decreased with time.

There was a progressive decrease in availability of Cs to plants with time implying a progressive fixation of Cs in the soil.

Acknowledgements

We would like to thank Harry Shepherd for

Table 3. Dry matter yield (g m^{-2}) and ^{134}Cs activity (Bq g^{-1}) of plant material at different harvesting times

Period	Improved pasture				Rough grazing			
	Yield		Activity		Yield		Activity	
	Grass	Clover	Grass	Clover	Grass	Heather	Grass	Heather
1988								
July	47		371		21.1		87	
August	119		501		6.6		364	
September	27.1		346		8.7	2.7	310	545
October	28		260		0.8		289	
1989								
March	66.4		50		0		–	
April	0		–		33.6		229	
May	133		234		22.0	3.0	133	483
June	62.1	3.3	324	346	12.9		134	
July	68.3	12.1	191	399	37.8	31.0	55	255
August	87.7		145		6.5	5.9	58	179
September	63.4		118		12.2		73	
October	21.9		112		0		–	
1990								
April	6.3		76		7.5	4.7	33	111
May	11.6		79		8.9		59	
June	69.7		92		27.1	1.5	36	107
July	72.9	10.7	116	137	0		–	
August	79.9	8.6	71	119	9.8	15.1	35	83
September	25.5	3.6	39	128	5.3	3.5	25	60

Table 4. Percentage of ^{134}Cs released by sequential extraction using a 10:1 ratio of extractant to soil or peat

Extractant	Extraction number	Peaty podzol improved pasture						Hatton peat
		3 days		5 months		24 months		2 months
		0–3 cm	3–6 cm	0–3 cm	3–6 cm	0–3 cm	3–6 cm	3–6 cm
Water[b]	1	–	–	0.098	0.017	–	–	
Ammonium acetate (1 *M*)	1	24.2	45.1	9.02	5.78	1.05	2.13	82.8
	2	3.2	7.09	2.24	1.25	0.33	0.56	15.2
	3	1.20	1.75	1.07	0.52	0.17	0.31	5.32
Sodium pyrophosphate (0.1 *M*)	1	–	–	3.29	0.69	–	–	46.0
	2	–	–	1.21	ND[a]	–	–	21.6
	3	–	–	ND[a]	ND[a]	–	–	12.3
Cation exchange resin[c]	1	–	–	1.13	0.64	–	–	78.9
	2	–	–	1.31	1.61	–	–	16.7
	3	–	–	1.33	2.59	–	–	4.12

[a] ND, Not determinable.

[b] Soil solution obtained by centrifuging field-moist soil at 2000 *g*.

[c] One bag (3 g resin) to 200 mL water and 20 g soil or peat.

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