

Technical University of Denmark



An intercomparison of sampling techniques among five European laboratories for measurements of radiocaesium in upland pasture and soil

Nielsen, Sven Poul; Aarkrog, A.; Colgan, P.A.; McGee, E.; Synnott, H.J.; Johansson, K.J.; Horrill, A.D.; Kennedy, V.H.; Barbayiannis, N.

Publication date:
1992

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Nielsen, S. P., Aarkrog, A., Colgan, P. A., McGee, E., Synnott, H. J., Johansson, K. J., ... Barbayiannis, N. (1992). An intercomparison of sampling techniques among five European laboratories for measurements of radiocaesium in upland pasture and soil. (Denmark. Forskningscenter Risoe. Risoe-R; No. 620(EN)).

DTU Library
Technical Information Center of Denmark

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

An Intercomparison of Sampling Techniques Among Five European Laboratories for Measurements of Radiocaesium in Upland Pasture and Soil

**S.P. Nielsen, A. Aarkrog, P.A. Colgan, E. McGee, H.J. Synnott, K.J.
Johansson, A.D. Horrill, V.H. Kennedy and N. Barbayiannis**

An Intercomparison of Sampling Techniques Among Five European Laboratories for Measurements of Radiocaesium in Upland Pasture and Soil

Riso-R-620(EN)

**S.P. Nielsen¹, A. Aarkrog¹, P.A. Colgan², E. McGee², H.J. Synnott²,
K.J. Johansson³, A.D. Horrill⁴, V.H. Kennedy⁴ and N. Barbayiannis⁵**

¹ **Risø National Laboratory, Roskilde, Denmark**

² **Radiation Protection Institute, Dublin, Ireland**

³ **Swedish University of Agricultural Sciences, Uppsala, Sweden**

⁴ **Institute of Terrestrial Ecology (ITE), Cumbria, United Kingdom**

⁵ **Aristotle University of Thessaloniki, Greece**

**Risø National Laboratory, Roskilde, Denmark
February 1992**

Abstract An intercomparison of sampling procedures used by five European laboratories for the determination of radiocaesium in vegetation and peaty soil was carried out at two locations in Cumbria. The soil sampling procedures included the collection of depth profiles in order to obtain information on the vertical distribution of radiocaesium in addition to the total deposition. The number of samples taken by each laboratory varied from one to five. The multiple sampling has given information on the homogeneity of the parameters studied at each location. The parameters comprise soil bulk densities, total deposition of ^{137}Cs , deposition of ^{137}Cs in three soil layers, biomass densities, concentrations of ^{137}Cs in pasture, and activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) in soil and vegetation.

The determination of total deposition of ^{137}Cs gave no indication of differences between the laboratories. The total depositions of ^{134}Cs and ^{137}Cs observed at one site were compared with

levels obtained from another study and the two sets of data were found to be in good agreement. The results from the soil profiles do indicate significant differences between laboratories. This covers the vertical distributions of ^{137}Cs deposition including the $^{134}\text{Cs}/^{137}\text{Cs}$ -activity ratios as well as the soil bulk densities. One laboratory using a coring technique observed difficulties during sampling due to compression of the soil. The coring technique should thus be avoided or applied with extreme care for the sampling of depth profiles in peaty soil. The results from the sampling of pasture show no indication of differences between the laboratories.

For the parameters studied the observed variabilities across soil depths and locations range from 10% to 81% in terms of relative standard deviations. A comparison across all results at the two locations indicate a 50% higher field variability at one of the sites relative to the other.

ISBN 87-550-1801-7
ISSN 0106-2840

Grafisk Service, Risø, 1992

Contents

| | |
|--|-----------|
| 1 Introduction | 5 |
| 2 Materials and Methods | 6 |
| 2.1 Sampling Sites | 6 |
| 2.2 Sampling Methods and Laboratory Procedures | 6 |
| 2.3 Statistical Procedures | 8 |
| 3 Results and Discussion | 8 |
| 3.1 Soil Bulk Densities | 8 |
| 3.2 Total Deposition | 10 |
| 3.3 Soil Depth Sections | 11 |
| 3.4 Vegetation | 16 |
| 3.5 Summary of ANOVA's and Site Variabilities | 18 |
| 4 Conclusions | 20 |
| Acknowledgement | 21 |
| References | 21 |
| Appendix | 22 |

1 Introduction

In connection with compilations of environmental data it is usually assumed that the data are unbiased and comparable. However, differences may exist between data from different investigators, and such differences should preferably be identified through an intercomparison programme and corrected for prior to data compilation. Systematic differences may be due to several reasons: eg. laboratory treatments, analytical techniques or sampling procedures. Depending on the type of data, the sampling procedures may have a significant influence on the quality of the results. In the case of soil data it is known (Federer et al, 1989) that properties such as pH, clay fraction and cation exchange capacity may require from a few to about 40 samples each of about 0.1 kg per 100 m² to achieve a confidence interval of 90%. Data on soil properties including levels of radioactive contamination may thus not be representative if the sampling procedures do not take this variability into account.

In order to investigate the influence of sampling procedures on data on radiocaesium in pasture grass and soil, the present sub-study was included in a CEC project on the Radioecology of Semi-Natural Ecosystems (B17-0044). The objective of this project is to study the levels of radiocaesium in upland vegetation and grazing animals, and to compare the transfer of radiocaesium in the soil-grass-animal foodchain for different upland ecosystems represented in the countries of the participants. The participants are the Institute of Terrestrial Ecology (ITE), Cumbria, United Kingdom; Radiation Protection Institute, Dublin, Ireland; Aristotle University of Thessaloniki, Greece; Swedish University of Agricultural Sciences, Uppsala, Sweden; and Risø National Laboratory, Roskilde, Denmark. The concentrations of radiocaesium in grass and soil are thus basic points of reference for these

investigations. During a project meeting at the ITE, Merlewood Research Station, a sampling intercomparison of vegetation and soil was carried out the 18 September 1990 at two locations in Cumbria. Each of the five participating laboratories used their own sampling technique to collect samples of vegetation and of soil segments at depths 0-5 cm, 5-10 cm and 10-20 cm at both locations. The participants brought their samples to their home laboratories where the samples were treated and analyzed using routine procedures.

This report gives the results of this investigation which focuses on possible systematic inter-laboratory differences. Such differences may, as stated, have other causes than the sampling procedures, and in order to identify possible differences between laboratories due to the gamma-spectrometric analyses, an additional intercalibration exercise comprising analyses of prehomogenised samples of vegetation and soil was carried out. The results of this intercalibration, which included only four of the five laboratories mentioned above, are reported elsewhere (McGee et al, 1992). The results in the present report have *not* been corrected in view of the findings from this intercalibration of gamma-spectrometric analyses. The latter intercalibration found statistically significant, but relatively small differences between laboratory determinations of ¹³⁷Cs; the mean percentage bias for each of the four laboratories was below 5%. The variation in the present investigation between multiple samples taken by the individual laboratories are in most cases much larger than 5%. The possible inter-laboratory bias for the gamma-spectrometric results for ¹³⁷Cs will thus have only a marginal influence on the conclusions for this radionuclide drawn from the present set of results.

2 Materials and Methods

2.1 Sampling Sites

Two sampling sites in areas of known Chernobyl deposition were chosen in Cumbria: Ennerdale (National Grid Reference NY 065128, altitude 275 m) and Corney Fell (National Grid Reference SD 150897, altitude 400 m). Both locations are upland acidic grassland sites grazed by sheep. The soil is characterized as mainly peat. Apart from manipulation of grazing pressures no known management is carried out in these two areas.

The meteorological conditions in Cumbria during the sampling on the 18 September 1990 were characterized by moderate to heavy rainfall.

2.2 Sampling Methods and Laboratory Procedures

In the following the five participating laboratories are denoted by the letters A through E. The sample numbers refer to the tables listed in the appendix.

Laboratory A

Laboratory A collected vegetation samples by clipping with garden shears. Soil samples were taken using two procedures. One procedure (samples no. 1 and 2) comprised the cutting of soil blocks of dimensions $10 \times 10 \text{ cm}^2$ or $15 \times 15 \text{ cm}^2$ with a knife as deep as possible. The edges of the blocks were trimmed to give as good a definition of the size as possible. In the field the blocks were divided in sections of 0-5 cm, 5-10 cm, 10-15 cm etc. by cutting with a pair of scissors.

The second procedure (sample no. 3) consisted of taking ten samples using a cylindrical metal corer with a diameter of 5.6 cm and a depth of 10 cm. The samples were taken randomly within an area of approx. 100 m^2 and composited to give one representative sample of the total deposition of the upper soil layer thus giving no information on the vertical distribution.

The soil samples were placed in plastic bags after sampling. In the laboratory the samples were dried at 108°C for a few days. The vegetation samples were milled and transferred to vials with volumes in the range of 35 mL to 180 mL. The soil samples were crushed and homogenized

manually. Plastic vials of 180 mL or 320 mL were filled with soil. The radiocaesium contents were determined using hyper-pure germanium detectors.

Laboratory B

Five vegetation samples and five soil samples were taken at each site (samples nos. 1-5). The vegetation samples were collected by clipping a $50 \times 50 \text{ cm}^2$ quadrat to approximately 1 cm above the soil surface. This facilitated the estimation of biomass and produced samples of sufficient bulk for radiocaesium analysis. The soil samples were collected by marking a $30 \times 30 \text{ cm}^2$ quadrat, digging a trench around this quadrat and extracting the soil monolith. The soil profiles represent the surface to sub-soil or bedrock interface. The extracted monoliths were sectioned into 0-5 cm, 5-10 cm, 10-20 cm and in further 10-cm sections to the base of the section. The large volume of the sections and the method of extraction is laborious, but avoids the problems of compression associated with the use of coring devices.

The entire vegetation samples from each quadrat were weighed both fresh and dry in order to allow calculation of biomass on both a wet and dry basis in kg m^{-2} . The vegetation samples were dried at 108°C for 24 hours and milled to less than 1-mm particle size in a Tecator Cyclotec plant mill and counted in geometries of 50 to 200 mL. All soil samples were sub-sampled taking known volumes of fresh material and drying at the same temperature and duration as for counted samples, bulk densities were then calculated by dividing dry weight by fresh volume. The remaining soil was dried for 72 hours at 108°C , ground to less than 1-mm particle size and counted in 500-mL Marinelli containers.

The soil samples were analyzed for radiocaesium by high-resolution gamma spectrometry using an Ortec GMX-series Ge detector (1.70 keV resolution at 1.33 MeV; 28% relative efficiency) linked to a Nuclear Data ND 9900 data acquisition system. Radioactivity concentrations were determined by automatic peak fitting using the Nuclear Data VAX/VMS spectroscopy applications software package. Efficiency calibration was carried out using a reference standard (QCY 44) supplied by Amersham International plc.

Laboratory C

Three replicate samples (samples nos. 1-3) were taken from each site within an area of approximately $100 \times 100 \text{ m}^2$. At each site an area of $1 \times 1 \text{ m}^2$ was marked out and the vegetation was clipped to within 2.5 cm of the soil surface with garden shears. In the centre of this clipped area a soil pit was excavated $25 \times 25 \text{ cm}^2$. A pit was actually excavated slightly to the side of the area needed in order that the soil could be sliced horizontally in the sampling area. Where possible 5-cm slices were taken until subsoil or bedrock prevented further work. Problems encountered in the field were mainly due to the occurrence of large stones and rocks in the soil. As far as possible rocks were assigned to their correct soil layers and taken to the laboratory for assessment.

Until treatment all samples were stored in a cold room at 2°C . Large rocks $> 2.5 \text{ cm}$ were removed by hand and weighed. The soil samples were then divided into 2 portions. The first was oven dried at 85°C for subsequent radiochemical analysis and the second reserved in the fresh condition for chemical analysis.

The dried soils were put through a rotary sieve and the $< 2\text{-mm}$ fraction placed in 150 mL counting containers for radiochemical analysis. Stones in the range 0.2 cm - 2.5 cm remaining in the sieve were weighed and discarded. In the case of the 0-5 cm layer the sieve contained an amount of organic material which would not pass the sieve and represented the tufted bases of graminoid species. This material was weighed and analyzed separately. No significant amount of roots was found for any of the other soil layers although the soil samples analyzed must have contained fragmented fibrous root material. Vegetation was dried to constant weight at 85°C . It was ground to pass a 0.7-mm sieve in a rotary-hammer mill.

All samples were counted on hyper-pure Ge detectors for about 22 hours. Spectral analysis was carried out using Canberra Apogee software. Counting errors vary with sample activity but, at the 95% confidence level, they were between 3 and 8% for the caesium isotopes.

Laboratory D

Laboratory D used a 6.5 cm diameter stainless steel corer for the soil sampling at each site. A total of 13 cores was taken from a $4 \times 4 \text{ m}^2$ grid as shown in Figure 1.

The corer was used to collect the desired depth segments in each of the 13 locations. The 13 subsamples thus obtained were composited to yield one sample for each depth segment. Grass and roots were kept in the samples.

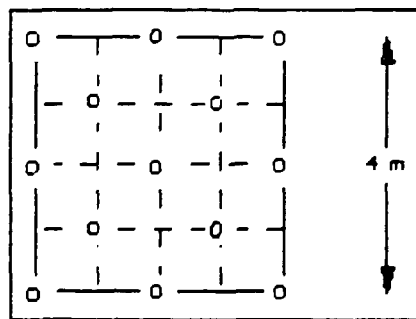
During sampling it was noted by the participants from laboratory D that the application of this coring technique to wet peaty soil proved to be rather difficult for the extraction of well-defined depth segments due to soil compression.

In the laboratory the soil samples were comminuted, dried, crushed, sieved, blended, and aliquots were taken for gamma-spectrometric analysis.

Vegetation samples were collected at each site from a quadrat of $1 \times 1 \text{ m}^2$ by cutting approximately 2 cm from the soil surface using a pair of garden shears. The samples were dried for 48 hours at 100°C .

The vegetation samples were measured in 200-mL cylindrical containers and the soil samples in 1-L Marinelli containers on Ge detectors connected to standard electronics and placed in 10-cm lead shields. The detectors are calibrated with water solutions of radionuclide standards, and the results are corrected for coincidence losses and self absorption in the sample. The recorded gamma spectra were analyzed by non-commercial software.

Figure 1. Scheme of soil sampling used by laboratory D.



Laboratory E

Laboratory E collected a vegetation sample from 1 m² at each site using a pair of scissors. Plastic cylinders with a diameter of 5.6 cm and 25-cm length were used for the soil samples which were taken from the area sampled for the vegetation. The cylinders were pushed into the soil slowly by hand when possible, or by means of a hammer; in the latter case a block of wood was used to avoid damage to the cylinder. The soil samples were pushed out of the cylinders with a wooden piston. When that was not possible eg. due to possible compaction in case of wet samples, the cylinders were brought to the laboratory, frozen and sectioned in 5-cm slices. The slices were composited to yield one sample for each depth segment of 0-5 cm, 5-10 cm and 10-20 cm.

In the laboratory the vegetation samples were dried for 48 hours at 80°C. The soil samples were air dried, crushed, sieved to less than 2-mm size, and sub-samples taken for radiometric analysis.

2.3 Statistical Procedures

The results have been subject to statistical analyses to test for differences between the labora-

tories. Two-way analyses of variance (ANOVA's) have been used to test the hypothesis that there are no systematic differences between the results from the different laboratories at the same time as testing for differences between either depths or locations. Differences found at the 5% level are indicated with a single asterisk (*) and called probably significant, differences found at the 1% level are indicated with a double asterisk (**) and called significant, and differences found at the 0.1% level are indicated with a triple asterisk (***) and called highly significant.

The results of the ANOVA's are shown in tables which list the sources of variation, the sums of squares, the degrees of freedom, the mean squares, the F-ratios and the significance levels. The within-laboratory variability is labelled 'Residual' and the between-laboratory variability appears under the main effects. In case that the differences between the laboratories are not insignificant, the laboratory means across the other factor (depths or locations) with 95% confidence intervals are illustrated in figures in order to give a graphical representation of the differences.

3 Results and Discussion

The individual results of the laboratory determinations of radiocaesium in soil and vegetation samples are shown in the appendix (Tables A-I to A-X). The results are given in terms of kilobecquerels per square metre (kBq m⁻²) and kilobecquerels per kilogramme dry weight (kBq kg⁻¹). All results given with reference to weight refer to dry weight conditions.

Table A-I gives the total deposition of the two caesium isotopes at the two locations. The total deposition comprises that of the vegetation and the soil layer. The Tables A-II and A-III give the results for the 0-5 cm soil layer at the two locations in terms of depositions (kBq m⁻²) and concentrations (kBq kg⁻¹), and the Tables A-IV to A-VII give the corresponding results for the soil layers 5-10 cm and 10-20 cm.

Data on soil bulk densities have been calculated from the collected soil samples in units of kg m⁻³. These data are listed in Table A-VIII. Table A-IX shows the ratios of the contents of the two caesium isotopes (¹³⁴Cs/¹³⁷Cs) in the collected soil

samples at the two locations. Table A-X summarises the results of the grass samples for the two locations, giving biomass densities (kg m⁻²) and concentrations (Bq kg⁻¹) for the two isotopes.

3.1 Soil Bulk Densities

The results of the soil bulk densities for the two locations were analyzed for differences between the four laboratories that provided these results and between the three depth sections at each site. Furthermore, since laboratory E did not provide information on these densities, average values were used to convert the concentrations (Bq kg⁻¹) given by this laboratory to depositions (Bq m⁻²).

The ANOVA of the densities from the Ennerdale location is given in Table I and shows significant differences between the laboratories and highly significant differences between the depths. The factor means for the laboratories are shown in Figure 2 which demonstrates that the results from laboratory D are significantly lower

Table I. ANOVA of soil bulk densities (kg m^{-3}) for the Ennerdale location, cf. Table A-VIII.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign level |
|---------------------|----------------|-----------|--------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 246194 | 3 | 82065 | 5.374 | 0.008 (**) |
| Depths | 460415 | 2 | 230208 | 15.08 | 0.000 (***) |
| Interactions | | | | | |
| Lab \times Depth | 119516 | 6 | 19919 | 1.304 | 0.305 |
| Residual | 274860 | 18 | 15270 | | |

than those from the others. An ANOVA performed on the data without those from laboratory D still gives some indication of difference between the laboratories (results from laboratory C are lower than those from laboratories A and B), but this difference is only probably significant. The mean bulk densities in this latter case are 182 ± 13 ($n=10$) kg m^{-3} , 506 ± 37 ($n=10$) kg m^{-3} and 296 ± 85 ($n=7$) kg m^{-3} for the depths 0-5 cm, 5-10 cm and 10-20 cm, respectively. The uncertainties given here and in the following are, unless stated otherwise, one standard error of the mean; n is the number of determinations.

The soil bulk densities from Corney Fell show highly significant differences between depths and probably significant differences between the laboratories (Table II). However, the interaction between laboratories and depths is probably significant due to an atypical variation of the results at the three depths for laboratory D compared with the other laboratories. The factor means are

depicted in Figure 3 which shows results from laboratory D to be higher than those of the other laboratories. An ANOVA performed on the data without those from laboratory D shows no difference between the laboratories and the probably significant difference between the interactions vanishes. The mean bulk densities for the latter sub-set of the data are 160 ± 10 ($n=10$) kg m^{-3} , 225 ± 9 ($n=10$) kg m^{-3} and 254 ± 31 ($n=10$) kg m^{-3} for the depths 0-5 cm, 5-10 cm and 10-20 cm, respectively.

The results of the ANOVA's of the soil bulk densities thus demonstrate that the sampling technique applied by laboratory D for the present soil types provide bulk densities of the soil layers that differ from those obtained by the other laboratories. Laboratory D did in fact experience difficulties with separating the depth segments during the sampling due to compression of the wet peaty soil.

Figure 2. Soil bulk densities of samples from Ennerdale showing 95% confidence intervals for factor means.

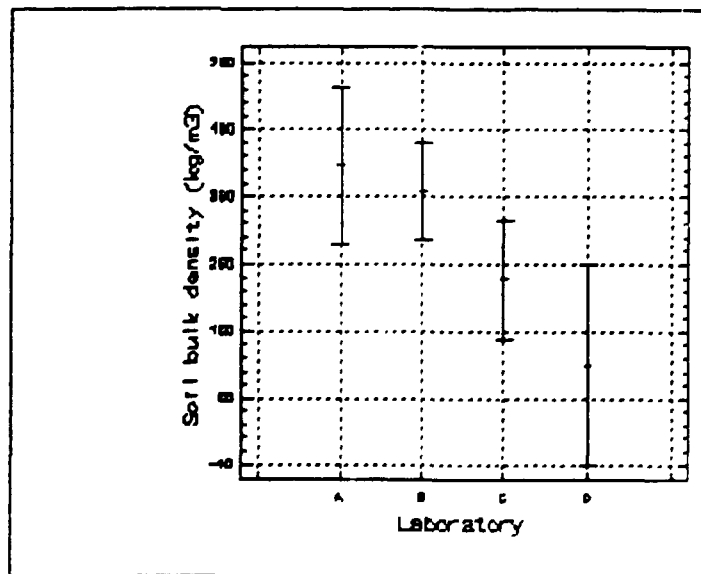
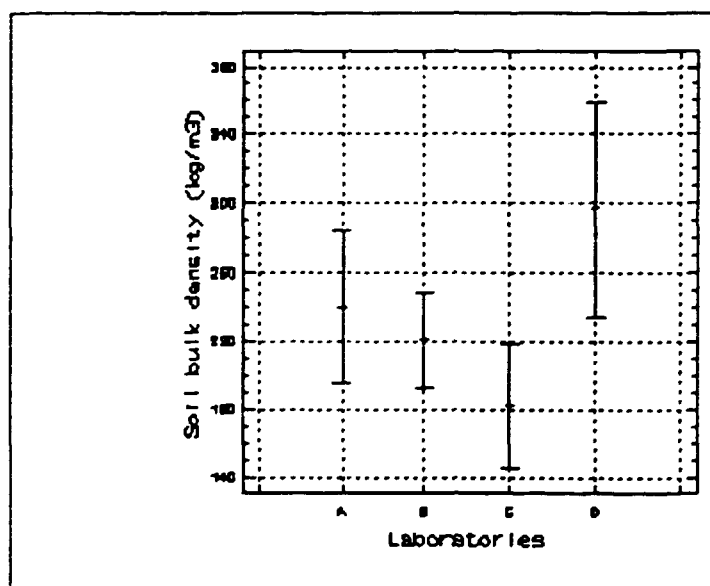


Table II. ANOVA of soil bulk densities (kg m^{-3}) for the Corney Fell location, cf. Table A-VIII.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 33335 | 3 | 11112 | 4.089 | 0.020 (*) |
| Depths | 64597 | 2 | 32298 | 11.88 | 0.000 (***) |
| Interactions | | | | | |
| Lab × Depth | 45479 | 6 | 7580 | 2.789 | 0.037 (*) |
| Residual | 57073 | 21 | 2718 | | |

Figure 3. Soil bulk densities of samples from Corney Fell showing 95% confidence intervals for factor means for the laboratories.



3.2 Total Deposition

The results of total deposition of ^{137}Cs in terms of kBq m^{-2} including vegetation and soil (Table A-I) show no differences between the laboratories whereas the differences between the locations are probably significant. The ANOVA is given in Table III. The mean total depositions of ^{137}Cs are 14.2 ± 1.3 ($n=13$) kBq m^{-2} for Ennerdale and 18.4 ± 1.3 ($n=13$) kBq m^{-2} for Corney Fell. For ^{134}Cs the mean total depositions are 1.13 ± 0.07 ($n=12$) kBq m^{-2} for Ennerdale and 1.63 ± 0.11 ($n=13$) kBq m^{-2} for Corney Fell.

The observations of total deposition of radio-caesium at Ennerdale may be compared with

post-Chernobyl levels of $19.3 \text{ kBq } ^{137}\text{Cs m}^{-2}$ and $3.1 \text{ kBq } ^{134}\text{Cs m}^{-2}$ which were obtained in winter 1988 at the same location by Baker and Cawse (1990). The figure for ^{137}Cs is within the observed range from the present investigation and the deviation from our mean value equals about one standard deviation. If we decay correct the above mentioned result for ^{134}Cs for the time period from January 1988 to September 1990 we get a value of $1.3 \text{ kBq } ^{134}\text{Cs m}^{-2}$. Again this value is within the range of our observed values, and the deviation from our mean value is within one standard deviation. The quoted figures from 1988 are thus in good agreement with the present results.

Table III. ANOVA of total deposition of ^{137}Cs (kBq m^{-2}), cf. Table A-I.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 103.42 | 4 | 25.85 | 1.295 | 0.314 |
| Locations | 111.37 | 1 | 111.37 | 5.578 | 0.031 (*) |
| Interactions | | | | | |
| Lab \times Loc | 101.59 | 4 | 25.40 | 1.272 | 0.323 |
| Residual | 319.42 | 16 | 19.96 | | |

3.3 Soil Depth Sections

The analyses of the results for the three soil depth sections have been made by carrying out ANOVA's for the data on deposition of ^{137}Cs . The data on ^{134}Cs have been considered by analyzing the isotopic ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) for consistencies.

The results of the ^{137}Cs depositions (kBq m^{-2}) in the upper soil layer of 0-5 cm (cf. Table A-II) give only an indication of differences between the laboratories. The ANOVA is shown in Table IV, where the difference between the laboratories is found to be probably significant. The factor means for the laboratories are shown in Figure 4. The mean values for Ennerdale are 10.5 ± 1.3 ($n=12$) $\text{kBq } ^{137}\text{Cs m}^{-2}$ and 1.00 ± 0.09 ($n=11$) $\text{kBq } ^{134}\text{Cs m}^{-2}$, and 11.7 ± 0.6 ($n=12$) $\text{kBq } ^{137}\text{Cs m}^{-2}$ and 1.22 ± 0.09 ($n=12$) $\text{kBq } ^{134}\text{Cs m}^{-2}$ for Corney Fell.

The ANOVA of the results for the ^{137}Cs deposition in the 5-10 cm soil layer (cf. Table A-IV) is shown in Table V where the difference between

the laboratories is found to be significant. The factor means are shown in Figure 5. However, the probably-significant interaction may be due to an atypical variation between the two locations for laboratory A compared with the other laboratories. Laboratory A finds a higher deposition of ^{137}Cs Cs in the 5-10 cm soil layer at Ennerdale than at Corney Fell in contrast to the other laboratories. When the ANOVA is repeated without the data of laboratory A, the probably significant interaction disappears and there remains probably significant differences between the laboratories and the locations. The differences between the laboratories are such that the results of laboratory C tend to be larger than those of the laboratories B, D and E. The factor means for the data without those from laboratory A are for Ennerdale 3.0 ± 0.6 ($n=10$) $\text{kBq } ^{137}\text{Cs m}^{-2}$ and 0.16 ± 0.03 ($n=7$) $\text{kBq } ^{134}\text{Cs m}^{-2}$, and 4.5 ± 0.5 ($n=10$) $\text{kBq } ^{137}\text{Cs m}^{-2}$ and 0.28 ± 0.04 ($n=10$) $\text{kBq } ^{134}\text{Cs m}^{-2}$ for Corney Fell.

Table IV. ANOVA of the deposition of ^{137}Cs (kBq m^{-2}) in the 0-5 cm soil layer, cf. Table A-II.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 110.13 | 4 | 27.53 | 3.274 | 0.043 (*) |
| Locations | 8.20 | 1 | 8.20 | 0.975 | 0.351 |
| Interactions | | | | | |
| Lab \times Loc | 56.27 | 4 | 14.07 | 1.673 | 0.212 |
| Residual | 117.73 | 14 | 8.41 | | |

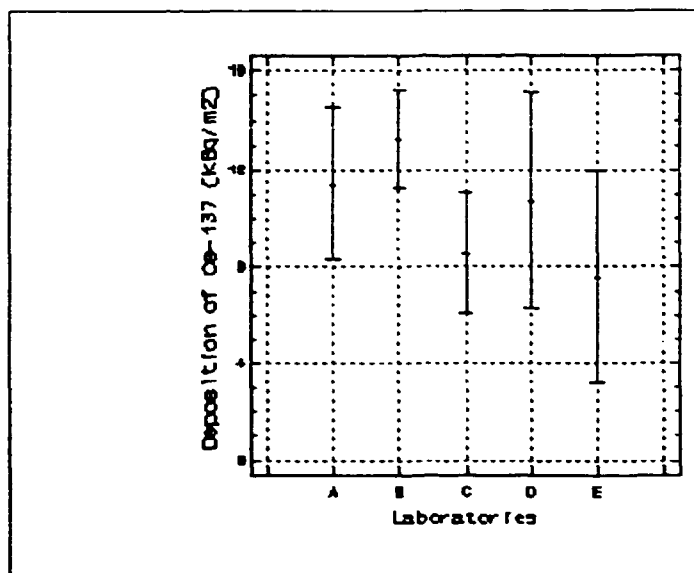
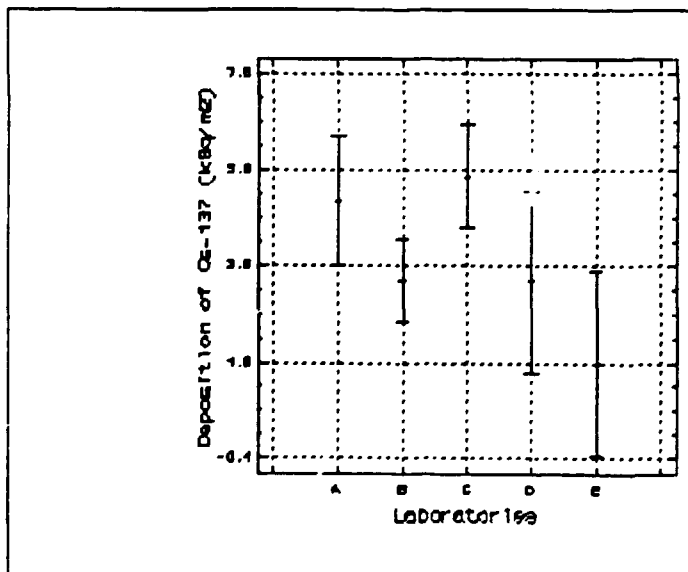


Figure 4. Depositions of ¹³⁷Cs (kBq m⁻²) in the 0.5 cm soil layers at the two locations showing 95% confidence intervals for factor means for the laboratories.

Table V. ANOVA of the deposition of ¹³⁷Cs (kBq m⁻²) in the 5-10 cm soil layer, cf. Table A-IV.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 34.394 | 4 | 8.599 | 5.557 | 0.007 (**) |
| Locations | 2.581 | 1 | 2.581 | 1.668 | 0.217 |
| Interactions | | | | | |
| Lab × Loc | 22.54 | 4 | 5.636 | 3.643 | 0.031 (*) |
| Residual | 21.66 | 14 | 1.547 | | |

Figure 5. Depositions of ¹³⁷Cs (kBq m⁻²) in the 5-10 cm soil layers at the two locations during 95% confidence intervals for factor means for the laboratories.



The ANOVA for the results of ^{137}Cs deposition in the 10-20 cm soil layer (cf. Table A-VI) is given in Table VI, which shows no difference between the laboratories and a probably significant difference between the locations. The mean values found at Ennerdale are 0.5 ± 0.1 ($n=9$) $\text{kBq } ^{137}\text{Cs m}^{-2}$ and 0.07 ± 0.04 ($n=3$) $\text{kBq } ^{134}\text{Cs m}^{-2}$, and 2.5 ± 0.6 ($n=12$) $\text{kBq } ^{137}\text{Cs m}^{-2}$ and 0.12 ± 0.07 ($n=11$) $\text{kBq } ^{134}\text{Cs m}^{-2}$ at Corney Fell.

The ANOVA of the ^{134}Cs results from the soil samples is based on the activity ratios, $^{134}\text{Cs}/^{137}\text{Cs}$, cf. Table A-IX. At Ennerdale the data are characterized by a number of missing values due to the absence of samples for the 10-20 cm soil layer as well as to concentrations of ^{134}Cs being below the limits of detection in several samples. In addition, laboratory A has an obvious outlier for the ratio (0.316) for the 10-20 cm soil layer. The ANOVA for the data from Ennerdale without this outlier is given in Table VII which shows significant differences between the laboratories, highly significant differences between the depths, and probably significant differences between the interactions. The factor means for the

laboratories are shown in Figure 6 where the results for laboratory D are seen to be higher than those of the other laboratories. When the ANOVA is repeated on the same data from which the results of laboratory D are removed, the probably significant interaction vanishes, but probably significant differences still remain between the laboratories (results of laboratory B lower than those of laboratories A and C) and highly significant differences between the depths. There thus seems to be some indication that the problems of compression of the soil samples by laboratory D are reflected in the results for the activity ratios. This is probably due to the fact that the slope of the vertical profile for ^{134}Cs is steeper than for ^{137}Cs since the latter includes older weapons fallout which shows greater vertical penetration than Chernobyl fallout. The activity ratio of a soil sample thus depends on sample depth. The factor means for the activity ratios without the outlier and those of laboratory D are 0.096 ± 0.006 ($n=10$) for the 0-5 cm soil layer, 0.041 ± 0.004 ($n=8$) for the 5-10 cm soil layer and 0.049 (single value) for the 10-20 cm soil layer.

Table VI. ANOVA of the deposition of ^{137}Cs (kBq m^{-2}) in the 10-20 cm soil layer, cf. Table A-VI.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 3.606 | 4 | 0.901 | 0.247 | 0.906 |
| Locations | 22.390 | 1 | 6.131 | 6.131 | 0.031 (*) |
| Interactions | | | | | |
| Lab \times Loc | 2.430 | 4 | 0.607 | 0.166 | 0.951 |
| Residual | 21.66 | 14 | 1.547 | | |

Table VII. ANOVA of the activity ratios of the radiocaesium isotopes ($^{134}\text{Cs}/^{137}\text{Cs}$) in soil samples from Ennerdale, cf. Table A-IX.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 0.003158 | 3 | 0.00105 | 8.678 | 0.003 (**) |
| Depths | 0.015565 | 2 | 0.00778 | 64.15 | 0.000 (***) |
| Interactions | | | | | |
| Lab \times Depth | 0.00168 | 4 | 0.00042 | 3.455 | 0.042 (*) |
| Residual | 0.00146 | 12 | 0.00012 | | |

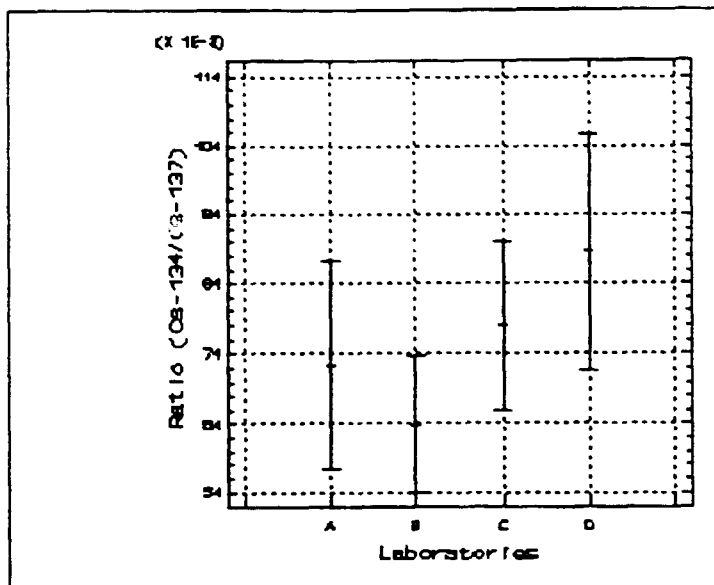


Figure 6. Activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) in soil samples from Ennerdale showing 95% confidence intervals for factor means for the laboratories.

The activity ratios from Corney Fell represent an almost complete set of observations; only a single value is missing. The data have been analyzed in an ANOVA which is shown in Table VIII; the factor means are shown in Figure 7. The ANOVA shows significant differences between the laboratories and highly significant differences between the depths. The results of laboratory A are higher than those of the other laboratories, and when the ANOVA is repeated on the data from which the results of laboratory A are

removed, the difference between the laboratories disappears. The factor means for the activity ratios in the latter case are 0.102 ± 0.004 ($n = 10$) for the 0-5 cm soil layer, 0.064 ± 0.005 ($n = 10$) for the 5-10 cm soil layer and 0.040 ± 0.006 ($n = 9$) for the 10-20 cm soil layer.

Since the major part of the ^{134}Cs activity is present in the upper soil layer, an ANOVA was carried out on the activity ratios for the 0-5 cm soil layers at the two locations. The results are given in Table IX which shows significant inter-

Figure 7. Activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) in soil samples from Corney Fell showing 95% confidence intervals for factor means for the laboratories.

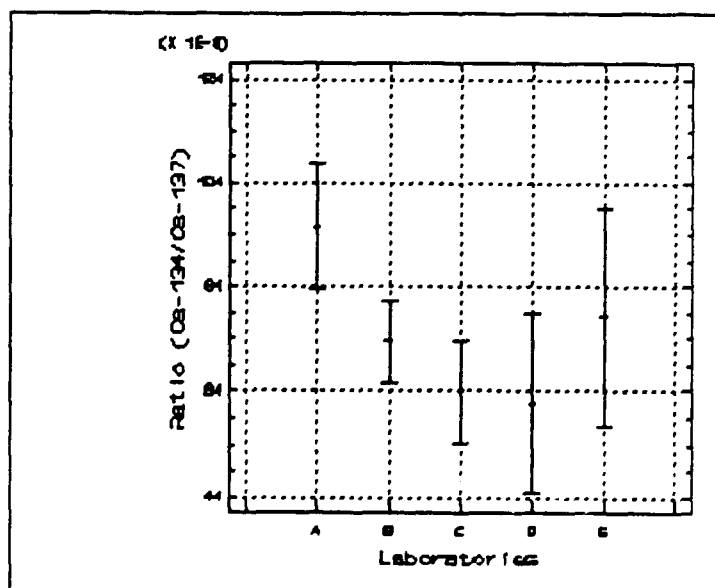


Table VIII. ANOVA of the activity ratios of the radiocaesium isotopes ($^{134}\text{Cs}/^{137}\text{Cs}$) in soil samples from Corney Fell, cf. Table A-IX.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 0.004263 | 4 | 0.00107 | 5.312 | 0.004 (**) |
| Depths | 0.019305 | 2 | 0.00965 | 48.107 | 0.000 (***) |
| Interactions | | | | | |
| Lab × Depth | 0.003272 | 7 | 0.00047 | 2.330 | 0.063 |
| Residual | 0.004214 | 21 | 0.00020 | | |

actions between laboratories and locations and probably significant differences between the locations. The factor means are shown in Figure 8. If the variabilities of the main effects are compared to the interactions rather than the residual, the probably significant difference between the locations disappears. The significant interactions are illustrated in Figure 9 which shows the levels of the activity ratios for the five laboratories by the locations. The figure shows the activity ratios for the two locations Corney Fell and Ennerdale for each of the five laboratories from A to E (laboratory E has no result from Ennerdale). It is noted that while the laboratories A and B show higher ratios at Corney Fell than at Ennerdale, the opposite is the case for laboratories C and D. If the ANOVA is repeated on the data without those of laboratory D due to the problems associated with the compression of the soil sections, the two-factor interaction remains significant and there are

no significant main effects when compared to this interaction. The reasons for the significant interactions are not clear from the present material. However, gamma-spectrometric analyses of ^{134}Cs are less straightforward than for ^{137}Cs because of the multi-gamma emissions from ^{134}Cs compared to the mono-energetic gamma rays from ^{137}Cs . The analysis of multi-gamma radionuclides require that the effects of coincidence losses are considered. It was indeed found from the intercalibration of gamma-spectrometric analyses of pre-homogenised samples mentioned in the introduction (McGee et al., 1992) that the bias errors and measurement errors were larger for ^{134}Cs than for ^{137}Cs , in particular for laboratory A. The factor means for the activity ratios of the 0-5 cm soil layer including all results are 0.104 ± 0.004 ($n=12$) for Corney Fell and 0.097 ± 0.005 ($n=11$) for Ennerdale.

Table IX. ANOVA of the activity ratios of the radiocaesium isotopes ($^{134}\text{Cs}/^{137}\text{Cs}$) in the 0-5 cm soil samples from Corney Fell and Ennerdale, cf. Table A-IX.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 0.001139 | 4 | 0.00028 | 2.754 | 0.070 |
| Locations | 0.000521 | 1 | 0.00052 | 5.043 | 0.041 (*) |
| Interactions | | | | | |
| Lab × Loc | 0.002806 | 3 | 0.00094 | 9.046 | 0.001 (**) |
| Residual | 0.001447 | 14 | 0.00010 | | |

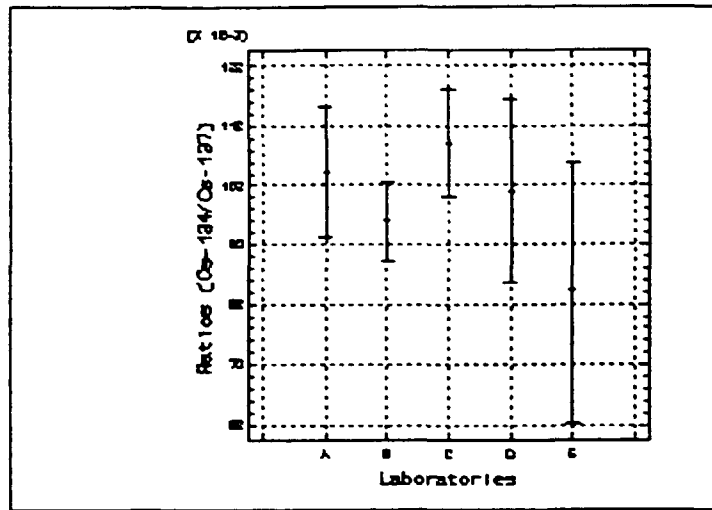
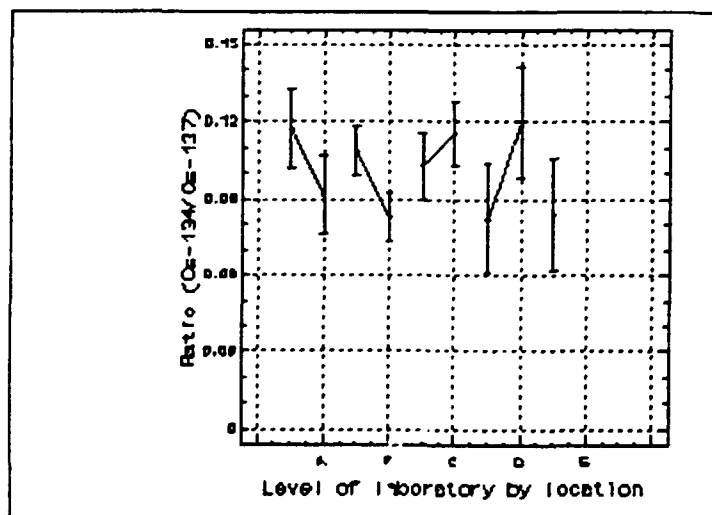


Figure 8. Activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) in the 0-5 cm soil layer samples from Corney Fell and Ennerdale showing 95% confidence intervals for factor means for the laboratories.

Figure 9. Activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) in the 0-5 cm soil layer samples from Corney Fell and Ennerdale showing 95% confidence intervals for factor means for the laboratories by the locations.



The results from the soil samples do not present a clear picture of inter-laboratory differences. The significant differences found from the ANOVA's indicate that laboratory A provides soil profiles of ^{137}Cs concentrations that differ from those of the other laboratories.

3.4 Vegetation

The biomass densities (cf. Table A-X) show no differences between laboratories or locations; the ANOVA is shown in Table X. The mean biomass density is 0.22 ± 0.02 ($n=18$) kg m^{-2} . The concentrations of ^{137}Cs in the grass show highly sig-

nificant differences between locations, but no differences between the laboratories. The mean concentration of ^{137}Cs in grass at Ennerdale is 199 ± 30 ($n=12$) Bq kg^{-1} and 786 ± 70 ($n=12$) Bq kg^{-1} at Corney Fell. The ANOVA is shown in Table XI. An ANOVA for ^{134}Cs in the grass gives similar results as for ^{137}Cs . An ANOVA of the ratios between the levels of ^{134}Cs and ^{137}Cs in the grass is shown in Table XII. Here no differences are found between the laboratories or the locations; the mean activity ratio is 0.104 ± 0.002 ($n=24$) which compares well with the activity ratio of the 0-5 cm soil layers.

Table X. ANOVA of biomass densities (kg m^{-2}), cf. Table A-X.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 0.00260 | 2 | 0.00130 | 0.176 | 0.841 |
| Locations | 0.00823 | 1 | 0.00823 | 1.112 | 0.312 |
| Interactions | | | | | |
| Lab \times Loc | 0.00313 | 2 | 0.00157 | 0.211 | 0.812 |
| Residual | 0.08886 | 12 | 0.00741 | | |

Table XI. ANOVA of ^{137}Cs in grass (Bq kg^{-1}), cf. Table A-X.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 127655 | 3 | 42552 | 1.396 | 0.292 |
| Locations | 1412461 | 1 | 1412461 | 46.34 | 0.000 (***) |
| Interactions | | | | | |
| Lab \times Loc | 27936 | 3 | 9312 | 0.306 | 0.821 |
| Residual | 365751 | 12 | 30479 | | |

Table XII. ANOVA of the activity ratios of the radiocaesium isotopes ($^{134}\text{Cs}/^{137}\text{Cs}$) in grass samples, cf. Table A-X.

| Source of variation | Sum of Squares | df | Mean square | F-ratio | Sign. level |
|---------------------|----------------|----|-------------|---------|-------------|
| Main effects | | | | | |
| Laboratories | 0.001013 | 3 | 0.000338 | 2.181 | 0.143 |
| Locations | 0.000097 | 1 | 0.000097 | 0.625 | 0.452 |
| Interactions | | | | | |
| Lab \times Loc | 0.000080 | 3 | 0.000027 | 0.172 | 0.913 |
| Residual | 0.001858 | 12 | 0.000155 | | |

The observed ratio for the levels of ^{137}Cs between vegetation and the 0-5 cm soil layer is considerably larger at Corney Fell (67 Bq kg⁻¹ per kBq m⁻²) than at Ennerdale (19 Bq kg⁻¹ per kBq m⁻²). This may be explained by the different chemical soil properties at the two locations as determined by the Institute of Terrestrial Ecology. The organic content of the 0-5 cm soil layer soil from Corney Fell is about 85% which is similar to that from Ennerdale, but the cation exchange capacity is greater at Corney Fell (49 meq per 100 g) than that at Ennerdale (24 meq per 100 g).

3.5 Summary of ANOVA's and Site Variabilities

Table XIII summarises the results of the previous ANOVA's concerning differences between the laboratories. The table shows the levels of significance found for the four groups of data mentioned in the previous sections. For all the cases where significant differences are found, the differences can be related to the data from a single laboratory. This means that if the data from that laboratory are removed, the significance levels become less significant (corresponding to p

> 1%) when the ANOVA's are repeated. The table identifies these laboratories and shows the effects causing the significant differences by ranking the levels.

For the soil bulk density the results from laboratory D at Ennerdale are significantly lower than those of the other three laboratories among which the results are not significantly different. The results from laboratory C are lower than those from laboratory B which again are lower than those from laboratory A. This ranking is indicated in the table in the column labelled 'Cause of significance'.

A significant two-factor interaction is found for the deposition of ^{137}Cs in soil for the 5-10 cm depth section which is caused by the results from laboratory A. The significant interaction is due to higher levels at Ennerdale than at Corney Fell reported by this laboratory in contrast to the trend observed by the other laboratories.

For the activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) laboratory D gives values at Ennerdale which are significantly lower than those of the other laboratories, whereas at Corney Fell laboratory A finds values which are higher than those of the other laboratories.

*Table XIII. Summary of the results from the ANOVA's highlighting differences between the laboratories. In case of significant differences (**, p < 1%) the laboratory whose data differ from those of the other laboratories is indicated as well as the cause of the significance (ranking of the levels).*

| Data | Sign. (Lab) | Cause of significance |
|--|-------------|-----------------------|
| Soil bulk density (kg m ⁻³) | | |
| Ennerdale | ** (D) | D < C, B, A |
| Corney Fell | * | |
| Deposition of ^{137}Cs in soil (kBq m ⁻²) | | |
| Total | ns | |
| 0-5 cm layer | * | |
| 5-10 cm layer | ** (A) | Interaction (loc.) |
| 10-20 cm layer | ns | |
| $^{134}\text{Cs}/^{137}\text{Cs}$ ratios in soil | | |
| Ennerdale | ** (D) | D > C, A, B |
| Corney Fell | ** (A) | A > E, B, C, D |
| Vegetation | | |
| Biomass (kg m ⁻²) | ns | |
| ^{137}Cs conc. (Bq kg ⁻¹) | ns | |
| $^{134}\text{Cs}/^{137}\text{Cs}$ ratios | ns | |

The results from the vegetation samples analysed in terms of biomass, ^{137}Cs concentration and of activity ratios yield no significant differences between the laboratories indicating the consistency of sampling and analytical procedures for vegetation samples in all laboratories.

The significant differences between the laboratories mentioned above have been determined in view of the within-laboratory variabilities of the results. In order to study this variability in greater detail, Table XIV summarises the results for the two locations showing means and both absolute and relative standard deviations. The statistics have been calculated from the data without those sub-sets that have been identified as significantly different from the rest.

It is noted from the table that the relative standard deviations at Ennerdale are generally some-

what higher than at Corney Fell. This difference has been studied by forming the ratios of the relative standard deviations at Ennerdale to those at Corney Fell and comparing these ratios to unity. When these ratios are tested with a Students t-test for each of the four groups of data, the means are higher than but not significantly different from unity. However, the entire set of data gives a mean ratio of 1.52, a standard deviation of 0.42 and a t-value of 4.23 (11 degrees of freedom) which is statistically significant at the 1% level. The data thus indicate that the field variability across all results is larger at the Ennerdale site than at the Corney Fell site, and that the variability in quantitative terms when expressed as a relative standard deviation is about 50% larger at Ennerdale than at Corney Fell.

Table XIV. Comparison of data variabilities between Ennerdale and Corney Fell showing means and absolute and relative standard deviations. The statistics do not include the sub-sets of data identified from the ANOVA's as significantly different from the rest.

| Data | Ennerdale | Corney Fell |
|--|-------------------------|-------------------------|
| Soil bulk density (kg m^{-3}) | | |
| 0-5 cm | 182 ± 40 (22%) | 160 ± 31 (19%) |
| 5-10 cm | 505 ± 117 (23%) | 225 ± 27 (12%) |
| 10-20 cm | 295 ± 225 (76%) | 254 ± 97 (38%) |
| ^{137}Cs deposition in soil (kBq m^{-2}) | | |
| Total | 14.2 ± 4.6 (33%) | 18.4 ± 4.7 (26%) |
| 0-5 cm | 10.5 ± 4.6 (44%) | 11.7 ± 2.2 (19%) |
| 5-10 cm | 3.0 ± 1.8 (59%) | 4.5 ± 1.6 (36%) |
| 10-20 cm | 0.5 ± 0.3 (71%) | 2.5 ± 2.0 (81%) |
| $^{134}\text{Cs}/^{137}\text{Cs}$ ratios in soil | | |
| 0-5 cm | 0.096 ± 0.017 (18%) | 0.102 ± 0.013 (13%) |
| 5-10 cm | 0.041 ± 0.011 (27%) | 0.064 ± 0.016 (25%) |
| 10-20 cm | 0.049 | 0.040 ± 0.019 (48%) |
| Vegetation | | |
| Biomass (kg m^{-2}) | 0.195 ± 0.078 (40%) | 0.238 ± 0.076 (32%) |
| ^{137}Cs concentration (Bq kg^{-1}) | 193 ± 115 (60%) | 724 ± 211 (29%) |
| $^{134}\text{Cs}/^{137}\text{Cs}$ ratios | 0.103 ± 0.009 (9%) | 0.107 ± 0.015 (14%) |

4 CONCLUSIONS

The present intercomparison exercise has covered sampling procedures used by five laboratories for the determination of radiocaesium deposition in vegetation and peaty soil. The soil sampling procedures included the collection of soil sections for the depths 0-5 cm, 5-10 cm and 10-20 cm in order to obtain information on the vertical distribution of radiocaesium in the soil in addition to the total deposition. Three laboratories used techniques of cutting horizontal slices of soil blocks to obtain the desired depth segments, while two laboratories used a coring technique. The techniques of sampling the vegetation did not differ much among the laboratories; the main difference was the cutting height which varied from 1 to 2.5 cm. The number of samples taken by each laboratory varied from one to five. The multiple sampling done by three of the laboratories gives information on the field variability of the parameters studied at each location.

For the determination of total deposition of ^{137}Cs the results give no indication of differences between the laboratories compared to the observed within-laboratory variability. The observed variabilities at the two locations correspond to standard deviations of 33% at Ennerdale and 26% at Corney Fell.

The total deposition of ^{134}Cs and ^{137}Cs observed at Ennerdale has been compared with levels obtained in 1988 by Baker and Cawse (1990). The two sets of data were found to be in good agreement with each other.

The results from the soil samples of depth sections do indicate significant differences between the laboratories. This covers the vertical distributions of ^{137}Cs deposition including the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios as well as the soil bulk densities. The laboratory (D) using the coring technique observed difficulties during sampling with efficient separation of the depth sections during sampling due to compression of the wet peaty soil. These difficulties are clearly reflected in the soil bulk densities obtained at the Ennerdale site and to some extent also at the Corney Fell site. Furthermore, the activity ratios at the Ennerdale site for this laboratory are also affected, probably due to the same problem. One laboratory (A) using the technique of horizontal slicing to obtain the soil depth profile of ^{137}Cs deposition produced results which are significantly different from those of the other laboratories. This is par-

ticularly reflected in the results of the 5-10 cm soil layer, but also in the results of the activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) from Corney Fell. It should be noted, however, that this laboratory collects soil samples on a routine basis for total deposition only, and usually not for depth profiles. The significant differences of the results of the two laboratories have been found on basis of the within-laboratory variabilities of the data. If we do not consider the results which have been identified as significantly different, the following variabilities in terms of standard deviations have been observed. For the soil bulk densities the variabilities across locations and depths range from 12% to 76%. For the deposition of ^{137}Cs in the 0-5 cm soil layer the variabilities range from 19% to 44%, for the 5-10 cm layer from 36% to 59%, and for the 10-20 cm layer from 71% to 81%. For the activity ratios the variability of the results range from 18% to 27% at Ennerdale and from 13% to 48% at Corney Fell.

The results from the sampling of pasture show no indications of differences between the laboratories. The results cover biomass yields, concentrations of ^{137}Cs and activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$). The observed variabilities for the grass biomass range from 32% to 40%, concentrations vary by 29% to 60%, and activity ratios by 10% to 16%.

In addition to the significant differences ($p < 1\%$) found in the data material, several probably significant differences have been found ($p < 5\%$). In most cases these differences do not form a consistent pattern, and they are interpreted as due to natural field variability in connection with the sampling procedures applied.

A comparison across all results at the two locations indicate a 50% higher field variability at Ennerdale than at Corney Fell in terms of relative standard deviations.

In addition to the significant differences ($p < 1\%$) found in the material, several probably significant differences have been found ($p < 5\%$). In most cases these differences do not form a consistent pattern, and they are interpreted as due to natural field variability in connection with the sampling procedures applied. The natural field variability observed for the soil and vegetation samples emphasizes the importance of carrying out multiple sampling in order to obtain representative average values.

When comparing the results for the different soil sampling techniques, the coring technique is not to be recommended for the study of vertical profiles in peaty soil unless this technique is used with extreme care. Instead a technique using horizontal slicing should be applied. However, in case of obtaining data on total deposition the two techniques are expected to work equally well.

Acknowledgement

The present work was carried out in connection with a project on the Radioecology of Semi-natural Ecosystems (B17-044) supported by the Radiation Protection Programme of the Commission of the European Communities.

References

- Baker, S. J. and P. A. Cawse; A survey of radioactive caesium in soils of Cumbria and North Lancashire. AEA-EE-0047. AEA Environment and Energy, Harwell Laboratory, 1990.
- Federer, P., H. W. Schmitt, H. Sticher. Wie Repräsentativ sind Bodenanalysen? *Bulletin Bodenkundliche Gesellschaft der Schweiz*, 1989, 13, pp. 37-42.
- McGee, E.J., Colgan, P.A., Keatinge, M., Horrill, A.D., Kennedy, V.H., Johansson, K.J., Aarkrog, A. and Nielsen, S.P. Bias and measurement errors in radioactivity data from four European research laboratories. *The Analyst*, 1992. Accepted for publication.

Appendix

The following tables contain the results of the determinations by the participating laboratories of radiocaesium in soil and vegetation in addition to quantities derived from these results. The term *na* means that the samples were not available because no samples were or could be taken, and *nd* means not detected.

The mean values given in the tables are arithmetic means of the data.

Table A-I. Total deposition of radiocaesium isotopes (kBq m⁻²) at the two locations sampled and measured by the five laboratories.

| Total (kBq m ⁻²) | | Cs-137 | | Cs-134 | |
|------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | 13.42 | 19.02 | 1.04 | 2.06 |
| A | 2 | 23.37 | 14.40 | 1.52 | 1.47 |
| A | 3 | 10.55 | 14.54 | 0.86 | 1.67 |
| B | 1 | 13.25 | 20.60 | 0.99 | 2.10 |
| B | 2 | 16.97 | 20.24 | 1.30 | 1.78 |
| B | 3 | 16.99 | 23.27 | 1.23 | 1.85 |
| B | 4 | 22.00 | 14.73 | 1.55 | 1.53 |
| B | 5 | 15.65 | 14.72 | 1.11 | 1.35 |
| C | 1 | 11.58 | 12.88 | 1.02 | 0.98 |
| C | 2 | 11.14 | 24.71 | 0.72 | 2.10 |
| C | 3 | 11.48 | 27.07 | 0.91 | 1.97 |
| D | 1 | 11.31 | 19.54 | 1.26 | 1.35 |
| E | 1 | 7.20 | 13.00 | nd | 0.95 |
| Mean | | 14.22 | 18.36 | 1.13 | 1.63 |

Table A-II. Deposition in the 0-5 cm soil layer of radiocaesium isotopes (kBq m⁻²) at the two locations sampled and measured by the five laboratories.

| Soil, 0-5 cm (kBq m ⁻²) | | Cs-137 | | Cs-134 | |
|-------------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | 6.58 | 13.52 | 0.73 | 1.52 |
| A | 2 | 15.95 | 9.54 | 1.37 | 1.13 |
| B | 1 | 9.87 | 15.52 | 0.88 | 1.74 |
| B | 2 | 14.20 | 14.26 | 1.30 | 1.50 |
| B | 3 | 15.66 | 12.39 | 1.23 | 1.31 |
| B | 4 | 17.02 | 9.34 | 1.38 | 1.06 |
| B | 5 | 14.21 | 10.01 | 1.05 | 1.09 |
| C | 1 | 5.50 | 9.40 | 0.70 | 0.78 |
| C | 2 | 4.65 | 12.77 | 0.53 | 1.41 |
| C | 3 | 7.19 | 11.71 | 0.79 | 1.34 |
| D | 1 | 8.89 | 12.51 | 1.07 | 1.03 |
| E | 1 | 6.08 | 9.06 | nd | 0.76 |
| Mean | | 10.50 | 11.67 | 1.00 | 1.22 |

Table A-III. Concentrations in the 0-5 cm soil layer of radiocaesium isotopes (kBq kg⁻¹) at two locations sampled and measured by the five laboratories.

| Soil, 0-5 cm (kBq kg ⁻¹) | | Cs-137 | | Cs-134 | |
|--------------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | 0.907 | 1.595 | 0.101 | 0.180 |
| A | 2 | 1.589 | 1.300 | 0.137 | 0.154 |
| B | 1 | 0.994 | 1.872 | 0.089 | 0.210 |
| B | 2 | 1.125 | 1.242 | 0.103 | 0.130 |
| B | 3 | 1.511 | 1.294 | 0.118 | 0.136 |
| B | 4 | 1.817 | 1.278 | 0.147 | 0.145 |
| B | 5 | 1.338 | 1.291 | 0.099 | 0.141 |
| C | 1 | 0.710 | 1.516 | 0.087 | 0.124 |
| C | 2 | 0.781 | 1.819 | 0.083 | 0.201 |
| C | 3 | 1.014 | 1.752 | 0.110 | 0.199 |
| D | 1 | 1.200 | 1.676 | 0.144 | 0.138 |
| E | 1 | 0.680 | 1.130 | nd | 0.095 |
| Mean | | 1.139 | 1.480 | 0.111 | 0.154 |

Table A-IV. Deposition in the 5-10 cm soil layer of radiocaesium isotopes ($kBq m^{-2}$) at the two locations sampled and measured by the five laboratories.

| Soil, 5-10 cm ($kBq m^{-2}$) | | Cs-137 | | Cs-134 | |
|--------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | 5.317 | 3.808 | 0.395 | 0.290 |
| A | 2 | 6.473 | 2.548 | 0.231 | 0.279 |
| B | 1 | 3.380 | 4.242 | 0.107 | 0.327 |
| B | 2 | 2.457 | 3.625 | nd | 0.193 |
| B | 3 | 1.325 | 5.100 | nd | 0.380 |
| B | 4 | 4.460 | 3.562 | 0.146 | 0.325 |
| B | 5 | 1.125 | 3.442 | 0.055 | 0.185 |
| C | 1 | 5.879 | 3.468 | 0.317 | 0.200 |
| C | 2 | 1.845 | 7.403 | 0.192 | 0.501 |
| C | 3 | 4.131 | 7.030 | 0.117 | 0.400 |
| D | 1 | 1.719 | 4.825 | 0.155 | 0.161 |
| E | 1 | 0.807 | 2.336 | nd | 0.169 |
| Mean | | 3.626 | 4.282 | 0.190 | 0.284 |

Table A-VI. Deposition in the 10-20 cm soil layer of radiocaesium isotopes ($kBq m^{-2}$) at the two locations sampled and measured by the five laboratories.

| Soil, 10-20 cm ($kBq m^{-2}$) | | Cs-137 | | Cs-134 | |
|---------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | na | 2.708 | na | 0.191 |
| A | 2 | 0.463 | 2.171 | 0.147 | 0.187 |
| B | 1 | na | 0.633 | na | 0.023 |
| B | 2 | 0.308 | 2.163 | nd | 0.073 |
| B | 3 | na | 5.644 | na | 0.147 |
| B | 4 | 0.512 | 1.274 | 0.025 | 0.089 |
| B | 5 | 0.310 | 0.959 | nd | 0.039 |
| C | 1 | 0.201 | 0.010 | nd | 0.000 |
| C | 2 | 1.174 | 4.237 | nd | 0.186 |
| C | 3 | 0.161 | 6.659 | nd | 0.214 |
| D | 1 | 0.700 | 2.201 | 0.039 | 0.154 |
| E | 1 | 0.225 | 1.376 | nd | nd |
| Mean | | 0.451 | 2.503 | 0.070 | 0.118 |

Table A-V. Concentrations in the 5-10 cm soil layer of radiocaesium isotopes ($kBq kg^{-1}$) at two locations sampled and measured by the five laboratories.

| Soil, 5-10 cm ($kBq kg^{-1}$) | | Cs-137 | | Cs-134 | |
|---------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | 0.229 | 0.341 | 0.013 | 0.026 |
| A | 2 | 0.198 | 0.303 | 0.007 | 0.033 |
| B | 1 | 0.130 | 0.404 | 0.004 | 0.031 |
| B | 2 | 0.117 | 0.290 | nd | 0.015 |
| B | 3 | 0.050 | 0.425 | nd | 0.032 |
| B | 4 | 0.223 | 0.285 | 0.007 | 0.026 |
| B | 5 | 0.033 | 0.270 | 0.002 | 0.014 |
| C | 1 | 0.226 | 0.311 | 0.012 | 0.018 |
| C | 2 | 0.267 | 0.750 | 0.011 | 0.059 |
| C | 3 | 0.229 | 0.611 | 0.007 | 0.038 |
| D | 1 | 0.421 | 0.239 | 0.038 | 0.008 |
| E | 1 | 0.033 | 0.208 | nd | 0.015 |
| Mean | | 0.180 | 0.370 | 0.011 | 0.026 |

Table A-VII. Concentrations in the 10-20 cm soil layer of radiocaesium isotopes ($kBq kg^{-1}$) at the two locations sampled and measured by the five laboratories.

| Soil, 10-20 cm ($kBq kg^{-1}$) | | Cs-137 | | Cs-134 | |
|----------------------------------|------------|-----------|-------------|-----------|-------------|
| Lab | Sample no. | Ennerdale | Corney Fell | Ennerdale | Corney Fell |
| A | 1 | na | 0.063 | na | 0.004 |
| A | 2 | 0.011 | 0.072 | 0.003 | 0.006 |
| B | 1 | na | 0.030 | na | 0.001 |
| B | 2 | 0.016 | 0.086 | nd | 0.003 |
| B | 3 | na | 0.166 | na | 0.004 |
| B | 4 | 0.021 | 0.057 | 0.001 | 0.004 |
| B | 5 | 0.004 | 0.050 | nd | 0.002 |
| C | 1 | 0.021 | 0.001 | nd | 0.000 |
| C | 2 | 0.043 | 0.196 | nd | 0.009 |
| C | 3 | 0.016 | 0.220 | nd | 0.007 |
| D | 1 | 0.069 | 0.065 | 0.004 | 0.005 |
| E | 1 | 0.008 | 0.057 | nd | nd |
| Mean | | 0.023 | 0.089 | 0.003 | 0.004 |

Table A-VIII. Densities of soil samples (kg m^{-3}) from the two locations inferred from the laboratory results of radionuclide soil inventories per unit weight and area.

| Lab | Ennerdale | | | Corney Fell | | |
|------|-----------|---------|---------|-------------|---------|---------|
| | 0-5 cm | 5-10 cm | 10-20cm | 0-5 cm | 5-10 cm | 10-20cm |
| A | 145 | 605 | na | 170 | 223 | 430 |
| A | 201 | 655 | 426 | 147 | 168 | 304 |
| B | 199 | 520 | na | 166 | 210 | 211 |
| B | 252 | 420 | 193 | 230 | 250 | 252 |
| B | 207 | 530 | na | 191 | 240 | 340 |
| B | 187 | 400 | 244 | 146 | 250 | 224 |
| B | 212 | 682 | 738 | 155 | 255 | 192 |
| C | 155 | 521 | 95 | 124 | 223 | 71 |
| C | 124 | 363 | 274 | 140 | 197 | 216 |
| C | 142 | 361 | 100 | 134 | 230 | 302 |
| D | 148 | 82 | 101 | 149 | 404 | 338 |
| E | na | na | na | na | na | na |
| Mean | 179 | 467 | 271 | 159 | 241 | 262 |

Table A-IX. Activity ratios of radiocaesium isotopes (Cs-134/ Cs-137) in soil samples from the two locations.

| Lab | Ennerdale | | | Corney Fell | | |
|------|-----------|---------|---------|-------------|---------|---------|
| | 0-5 cm | 5-10 cm | 10-20cm | 0-5 cm | 5-10 cm | 10-20cm |
| A | 0.1108 | 0.0570 | na | 0.1127 | 0.0762 | 0.0705 |
| A | 0.0862 | 0.0356 | 0.3163 | 0.1181 | 0.1095 | 0.0861 |
| B | 0.0891 | 0.0315 | na | 0.1123 | 0.0771 | 0.0363 |
| B | 0.0913 | nd | na | 0.1048 | 0.0532 | 0.0337 |
| B | 0.0782 | nd | na | 0.1055 | 0.0745 | 0.0260 |
| B | 0.0808 | 0.0327 | 0.0488 | 0.1133 | 0.0912 | 0.0699 |
| B | 0.0742 | 0.0485 | nd | 0.1093 | 0.0537 | 0.0407 |
| C | 0.1270 | 0.0539 | nd | 0.0828 | 0.0577 | 0.0100 |
| C | 0.1089 | 0.0396 | nd | 0.1107 | 0.0676 | 0.0439 |
| C | 0.1102 | 0.0283 | nd | 0.1144 | 0.0569 | 0.0321 |
| D | 0.1198 | 0.0903 | 0.0558 | 0.0824 | 0.0334 | 0.0700 |
| E | nd | nd | nd | 0.0841 | 0.0723 | nd |
| Mean | 0.0979 | 0.0464 | 0.1403 | 0.1042 | 0.0686 | 0.0472 |

Table A-X. Results of biomass densities (kg m^{-2}) and radiocaesium concentrations (Bq kg^{-1}) in grass at the two locations.

| Lab | Samp no | Ennerdale | | | Corney Fell | | |
|------|------------|-----------------------------|-------------------------------|-------------------------------|-----------------------------|-------------------------------|-------------------------------|
| | | Yield kg m^{-2} | Cs-137 Bq kg^{-1} | Cs-134 Bq kg^{-1} | Yield kg m^{-2} | Cs-137 Bq kg^{-1} | Cs-134 Bq kg^{-1} |
| A | 1 | na | 202 | 17 | na | 1144 | 121 |
| A | 2 | na | 264 | 22 | na | 1052 | 132 |
| B | 1 | 0.209 | 131 | 14 | 0.075 | 453 | 50 |
| B | 2 | 0.327 | 233 | 28 | 0.263 | 714 | 68 |
| B | 3 | 0.179 | 87 | 8 | 0.247 | 575 | 63 |
| B | 4 | 0.067 | 98 | 9 | 0.295 | 990 | 113 |
| B | 5 | 0.172 | 36 | 5 | 0.360 | 847 | 89 |
| C | 1 | 0.295 | 261 | 27 | 0.228 | 515 | 50 |
| C | 2 | 0.198 | 271 | 29 | 0.223 | 1001 | 102 |
| C | 3 | 0.164 | 209 | 23 | 0.226 | 505 | 48 |
| D | 1 | 0.143 | 167 | 19 | 0.222 | 690 | 79 |
| E | 1 | na | 432 | 37 | na | 950 | 83 |
| Mean | | 0.195 | 193 | 20 | 0.238 | 724 | 75 |

Bibliographic Data Sheet Risø-R-620(EN)

Title and author(s)

An Intercomparison of Sampling Techniques Among
Five European Laboratories for Measurements of
Radiocaesium in Upland Pasture and Soil

S.P. Nielsen, A. Aarkrog, P.A. Colgan, E. McGee, H.J.
Synnott, K.J. Johansson, A.D. Horrill, V.H. Kennedy
and N. Barbayiannis

| | |
|---------------|-----------|
| ISBN | ISSN |
| 87-550-1801-7 | 0106-2840 |

| | |
|-----------------|---------------|
| Dept. or group | Date |
| Ecology Section | February 1992 |

| | |
|------------------------|------------------------|
| Group's own reg. No(s) | Project contract No(s) |
|------------------------|------------------------|

| | | | |
|-------|--------|---------------|------------|
| Pages | Tables | Illustrations | References |
| 25 | 24 | 9 | 3 |

Abstract (Max. 2000 characters)

An intercomparison of sampling procedures used by five European laboratories for the determination of radiocaesium in vegetation and peaty soil was carried out at two locations in Cumbria. The soil sampling procedures included the collection of depth profiles in order to obtain information on the vertical distribution of radiocaesium in addition to the total deposition. The number of samples taken by each laboratory varied from one to five. The multiple sampling has given information on the homogeneity of the parameters studied at each location. The parameters comprise soil bulk densities, total deposition of ^{137}Cs , deposition of ^{137}Cs in three soil layers, biomass densities, concentrations of ^{137}Cs in pasture, and activity ratios ($^{134}\text{Cs}/^{137}\text{Cs}$) in soil and vegetation.

The determination of total deposition of ^{137}Cs gave no indication of differences between the laboratories. The total depositions of ^{134}Cs and ^{137}Cs observed at one site were compared with levels obtained from another study and the two sets of data were found to be in good agreement. The results from the soil profiles do indicate significant differences between laboratories. This covers the vertical distributions of ^{137}Cs deposition including the $^{134}\text{Cs}/^{137}\text{Cs}$ -activity ratios as well as the soil bulk densities. One laboratory using a coring technique observed difficulties during sampling due to compression of the soil. The coring technique should thus be avoided or applied with extreme care for the sampling of depth profiles in peaty soil. The results from the sampling of pasture show no indication of differences between the laboratories.

For the parameters studied the observed variabilities across soil depths and locations range from 10% to 81% in terms of relative standard deviations. A comparison across all results at the two locations indicate a 50% higher field variability at one of the sites relative to the other.

Descriptors INIS/EDB

CESIUM 134; CESIUM 137; COMPARATIVE
EVALUATIONS; DATA COMPILATION;
DEPOSITION; INTERLABORATORY COMPARISONS;
PASTURES; PLANTS; SAMPLING; SOILS

Available on request from Risø Library, Risø National Laboratory,
(Risø Bibliotek, Forskningscenter Risø), P.O. Box 49,
DK-4000 Roskilde, Denmark.
Telephone +45 42 37 12 12, ext. 2268/2269
Telex 43 116. Telefax +45 46 75 56 27.

Available on request from:
Risø Library
Risø National Laboratory,
P.O. Box 49, DK-4000 Roskilde, Denmark
Phone + 45 42 37 12 12, ext. 2268/2269
Telex 43116, Telefax + 45 46 75 56 27

ISBN 87-550-1801-7
ISSN 0106-2840