Spatial variability of fallout Caesium-137 in Austrian alpine regions

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Abstract

As part of the Austrian soil contamination survey, total process standard deviations of soil deposition measurement originating from spatial variability of fallout deposition and measuring uncertainties have been determined. The total coefficient of variation (CV) of ^{137}Cs fallout in a 1 ha area, measured by soil core sampling, amounts to 21.5%. The measuring process contributes with 12.2% leaving a 17.7% CV purely due to spatial variability of the fallout. Representativity of the measured contamination is being assessed by calculating confidence intervals (95%), which vary between $\pm$ 25% and 42% of the measured values for different sampling methods used for the survey on which the Austrian caesium map is based. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Chernobyl fallout; Spatial variability; Total process coefficient of variation

1. Introduction

In order to assess the statistical significance of soil deposition data measured by soil sampling or in situ gamma spectrometry, information about process standard deviations in general and spatial variability of the fallout in particular is necessary. Distinguishing between different contributions to the total measuring process standard deviation requires a series of experiments especially designed to uncover these sources.

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In a field experiment, the total process standard deviation, measured as a coefficient of variation and hereafter called TPCV (Total Process CV), has been determined by multiple fallout measurements on an undisturbed meadow in Salzburg with previously determined relatively high Chernobyl-borne contamination. The contributions to the TPCV originating from the measuring process itself were determined by separate experiments or estimated from practice.

The data derived from this project are being used in the Austrian soil contamination survey ("Caesium Map", UBA, 1996; Bossew, Ditto, Falkner, Henrich, Kienzl & Rappelsberger, 1998) as input to the calculation of confidence levels of the deposition figures.

2. Methods

2.1. Sampling site

A meadow in the city of Salzburg which is known to have been undisturbed since the Chernobyl accident in 1986 has been selected for the investigation. Only grass and hay are harvested on the sampling site and it is more or less horizontal, the slope not being larger than 1%. It is a typical and representative meadow used for grass and hay production for feeding cows with the difference that in the urban area it is not used for cows grazing grass on it while this practice is very common in the rural areas. The flat terrain and the missing influence of large animals minimize the chance of redistribution effects for contaminated soil material by running water or grazing cows. For these reasons the meadow seems to be suitable for studying the primary distribution pattern of the radioactive contamination. The average ^{137}Cs soil contamination is relatively high at 43 kBq/m^2 (ref. 1. May 1986) thus allowing short measuring times. The site has also been selected because it is easily accessible from the physics department of the university of Salzburg. A 100×100 m^2 section of the entire 5 ha area has been sampled extensively according to a 3-fold grid as follows (Fig. 1):

grid 3: the 100×100 m^2 area divided in 25 20×20 m^2 squares, 9 of them (i.e. every second in each row and column) containing 3×3 sampling points, i.e. a total of 81 sampling points.

grid 2: starting area 10×10 m^2, subdivision according to the same pattern.

grid 1: starting area 1×1 m^2, subdivision according to the same pattern.

Grid 2 is located in one corner of grid 3, and grid 1 in a corner of grid 2, in turn. As some of the points coincide a total of 235 soil samples were taken. The soil cores were collected as cylindrical samples with 8.1 cm diameter and 14 cm length. They could be removed from the sampler as compact cores, therefore allowing their division into different layers in the laboratory. Smearing by drawing down contaminated material into deeper layers did not have any significant effect on the overall activity and was not considered for the further calculations.

The soil cores were dried, homogenised and measured with HPGe detectors. 24 out of the 235 soil cores were divided in 7 layers (each 2 cm thick) in order to get information about the depth distribution.
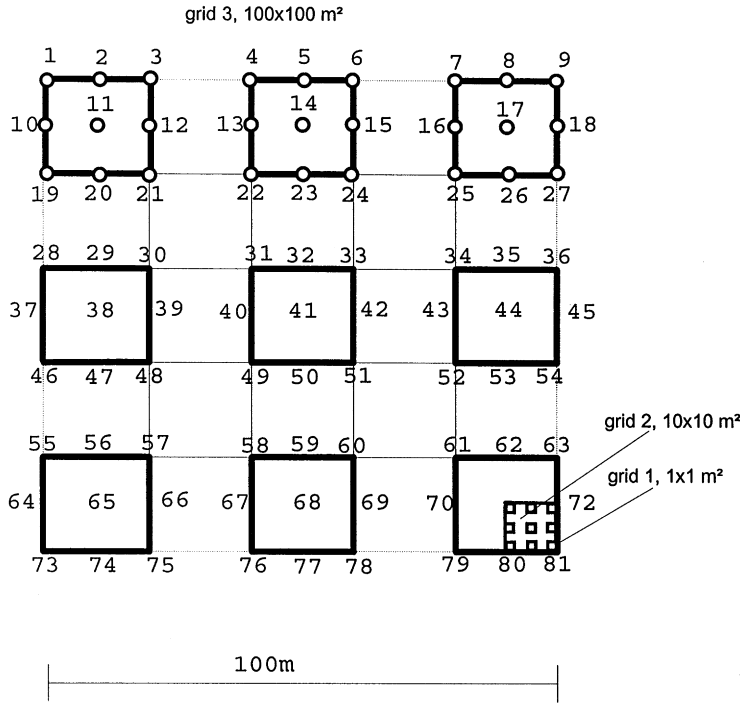
Fig. 1. Scheme of the grid arrangement designed for soil sample collection for investigating the spatial distribution of the Chernobyl fallout. Location: Urban area of Salzburg city.

2.2. Sampling and measuring errors

The sampling and measuring process involves various sources of errors, some of them stochastic, some systematic, like the error of determining the cross-section of the soil core, cut-off of the profile, i.e. neglecting contamination of deeper layers (systematic error), preparing aliquots from the total sample for gamma-spectrometry, positioning of the sample on the detector, detector calibration (systematic), photon attenuation in the sample (systematic) and counting statistics (stochastic).

Sample "management" errors like weighing errors and loss of soil during handling were estimated from practice. Several more sophisticated laboratory experiments were carried out in order to get information about still "trivial" error sources like non-central positioning of the sample on the detector; errors due to aliquotation and sample inhomogeneity were determined by measuring a series of aliquots out of one large original soil sample and by producing "artificially" inhomogenous samples, respectively, in order to see how such "sample mismanagement" would influence the overall error.

Possible errors which arise from the process of detector efficiency calibration were determined from literature data (uncertainties of photon peak yields), calibration source manufacturer's data (uncertainty of calibration source activity), the counting

error of the calibration measurement and the deviation of the calculated (fitted) efficiency function, which is used in routine measurements from measured efficiency values. These errors are partly stochastic and partly systematic as their origin is concerned; the efficiency values being used as constants for sample measuring, however, contribute as systematic errors.

A contribution to the overall uncertainty which is quite difficult to determine is due to the error which is almost necessarily made when correcting for photon attenuation in the sample. We believe, however, that the correction methods applied (Monte Carlo-based calculations and interpolations of empirically determined factors, respectively) guarantee this type of (systematic) error to be low, at least for medium and high energies like that of the ^{137}Cs photopeak (662 keV).

A serious source of uncertainty is the calculation of the amount of contamination below the cut-off depth of the soil sample. It is assessed by extrapolating known soil profiles as follows:

(1) A theoretical distribution, the diffusion–convection model, was fitted to the empirical ^{137}Cs profiles (Bossew, 1997). Parameters are the migration time from fallout (Chernobyl) until sampling (a known number, 7.41 years in this case), the apparent diffusion constant, the apparent convection velocity and the total radionuclide input, respectively (unknown).

(2) Fitting of each profile yields values for the three latter parameters. From these, we calculated the ratio

$$q = \left(\int_0^\infty C(x)\, dx \right) \bigg/ \left(\int_0^{14\ cm} C(x)\, dx \right),$$

where $C(x)$ is the theoretical distribution.

(3) The values $100(q - 1)$, calculated for each profile, indicate which part (in percent) of the total fallout is missed by the cut-off at 14 cm. The statistical evaluation gives $(8.3 \pm 6.6)\%$ (1 SD). The AM = 8.3% can be interpreted as a systematic error produced by the cut-off, the SD = 6.6% as a contribution to the stochastic uncertainty, due to the spatial inhomogeneity of the soil parameters (diffusion and convection constant) and hence the inhomogeneity of the profile shape.

Apart from these considerations, it must be stressed that the systematic cut-off error increases with time, because due to the downward migration of the ^{137}Cs, the fraction below the sampling depth increases. (A paper dealing with the detailed discussion of the spatial inhomogeneity of the depth distribution of ^{137}Cs is in preparation.)

2.3. Statistical evaluation

The analysis of homogeneity of the contamination of the Salzburg test site was carried out by analysis of variance (ANOVA). This method investigates if a random sample can be considered as grouped basically by comparing the arithmetic means (AM) of subgroups. In our case, the sub-squares of each grid were defined as the subgroups (see Fig. 1). For a more detailed explanation see the Results in Section 3.1. A condition for the ANOVA analysis to be carried out correctly is the normal
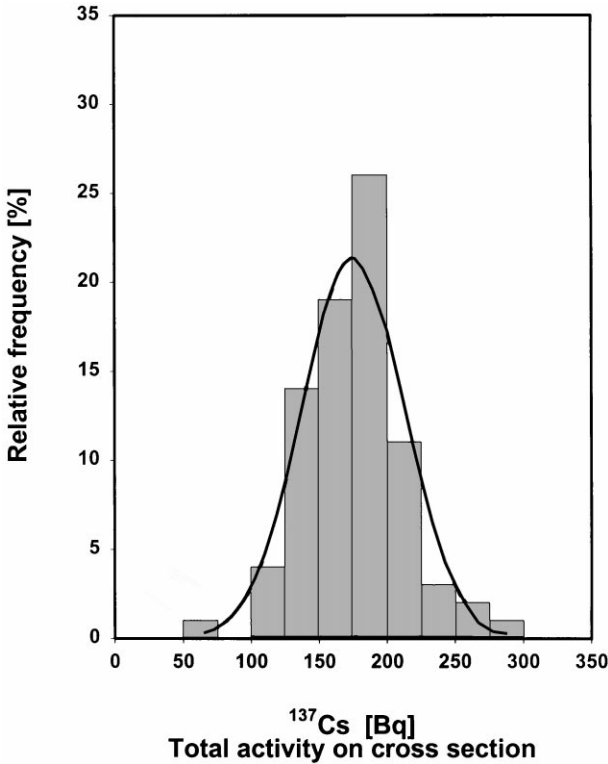
Fig. 2. Frequency distribution of the specific activity per cross section on grid 3 and calculated normal distribution with AM = 175.3 Bq and SD = 37.8 Bq. Cross-section of core sampler: 51.5 cm², yielding a specific activity per unit area of 34. kBq/m² at t_0 = Oct. 1993. Considering radioactive decay and activity below sampling depth of 14 cm, the activity per unit area = 43 kBq/m² at the time of surface deposition, May 1986.

distribution of the input data, in our case the measured deposition values on the three different grids. Normal distribution was confirmed **for all grids** by χ^2-testing. An example of the frequency distribution is shown in Fig. 2 using the data of the surface contamination on grid 3, the largest grid with 10 000 m². Grids 1 and 2 have similar shape. The ANOVA data were then used for calculating the confidence interval for a measured value (= the AM of contamination of N random samples taken from an area).

In practice, sampling can often not be carried out on a 100×100 m² standard area because of natural or technical reasons, but it can be done on a smaller area. In spite of this, the contamination data used in the Austrian caesium map are always considered as estimations of the mean contamination of the standard area centred around the indicated geographical location (Bossew, 1997). In other words, the mean contamination determined from a sub-area is interpreted as an estimation of the AM of the larger area. Statistically, this implies a further contribution to the overall

uncertainty of the calculated AM value, in addition to the uncertainty resulting from the spatial variability on the actual sampling area.

In order to assess this additional uncertainty, the ANOVA calculation was performed for differently sized subgroups and the corresponding confidence levels (CLs) plotted against the size of the area for getting information about how much this type of restriction related to sampling would affect the overall uncertainty.

3. Results and discussion

3.1. Analysis of the soil sample grids

Table 1 summarises the statistical parameters related to the three grids. $SD_{between.groups}$ and $SD_{within.groups}$ refer to the 9 sub-squares (= groups) of each grid. The principal result is that the subgroups of grid 3 (subgroups: 20 m × 20 m, grid 3: 100 m × 100 m) cannot be anticipated to be representative for the whole area. This important decision is based on the result of the *F*-test, which tests the null hypothesis of equal group means by comparing the standard deviation within the different groups (scatter of single values around the mean value of a certain group) with the standard deviation between the different groups (scatter of mean values of different groups around the total mean value). In other words, samples taken from grid 2 the size of which ($100\,m^2$) is of the same order of magnitude as the groups of grid 3 ($400\,m^2$), are *not* representative for the deposition on a $10\,000\,m^2$ area (grid 3). On the other hand, an additional *t*-test, which compares the AMs of grids 2 and 3 shows no significant difference ($p = 0.71 \gg 0.05$) from which we conclude that the location of grid 2 within grid 3 has been chosen by chance such as to yield representative deposition values for grid 3.

The Bartlett-test, finally, indicates homogeneity for the CVs of the subgroups of grid 3, which fits the result that $SD_{grid\,2} = 32.0 \approx SD_{within;\,grid\,3} = 33.0$. However, from $SD_{grid\,2} < SD_{grid\,3}$ with weak significance (additional *F*-test: $p = 0.072$) it follows

Table 1
Statistical parameters of the soil samples collected from the test site and results of the statistical tests applied

Parameter	Grid 1 ($1\,m^2$)	Grid 2 ($100\,m^2$)	Grid 3 ($10,000\,m^2$)
AM (Bq/51.5 cm²)[a]	171.3	173.3	175.3
SD (Bq/51.5 cm²)[a]	38.7	32.0	37.8
CV%	22.6	18.5	21.5
SD (between groups)	44.6	29.7	66.7
SD (within groups)	38.0	32.3	33.0
p (*F*-test)	0.22	0.56	0.005
p (Bartlett-test)	0.19	0.067	0.31

[a]The surface contamination refers to the size of the cylinder shape of the sample with a cross section of 51.5 cm²

that by selecting grid 2 we have found a sub-group with lower SD. If the CVs would not be homogenous, the selection of one sub-group would not necessarily yield a CV which is representative for an area of this size.

As grid 2 is concerned in the comparison with grid 1, AMs of subgroups of grid 2 are very homogenous (large $p(F)$). On the other hand, the almost significant Bartlett-value indicates a slight inhomogeneity of the CVs of the sub-groups of grid 2. This fits the observation that $SD_{grid\ 1} = 38.7$ is quite different from $SD_{within.groups;grid\ 2} = 32.3$. Therefore, arbitrary selection of a grid-1-sized area (1 m²) may not produce a CV which is representative of an area of the size of grid 2 (100 m²). Finally, both SDs between and within groups of grid 1 are relatively high indicating possible very small-scale (10 cm range) inhomogeneity.

It must be emphasised that, so far, all CV values include not only spatial variance but also uncertainties related to sampling and measuring, i.e. are TPCV values.

3.2. Contribution of process uncertainties

Table 2 gives the values for the different error contributions under certain conditions which we consider reasonable. For further calculations, however, values of the total stochastic error of 12.2%, determined for less favourable conditions and probably overly conservative, and of 5%, probably too optimistic, respectively, were used. The conservative value has been used as standard input value for the Austrian ceasium map. The figures hold for the sample management and measuring process as applied by the Austrian Ecology Institute.

All figures represent 1-Sigma relative standard deviations (CV%) and refer to ^{137}Cs measurements.

Table 2
Process-related sources of uncertainty for gamma spectrometric measurements of soil samples

Source of uncertainty	Uncertainty (%)
Stochastic	
Sampler deformation (affects cross-section)	5
Weighing	< 0.2
Aliquotation	3
Remaining sample inhomogeneity	4
Sample positioning on detector	2.7
Counting statistics (for ^{137}Cs), typically	3
Cut-off of profile at 14 cm, stochastic contribution	6.6
Total stochastic	10.5
Systematic	
Cut-off of profile at max. 14 cm, systematic contribution	$- \leqslant -8.3$
Loss of soil	$- \leqslant 0.2$
Detector efficiency calibration (for ^{137}Cs)	$+ 0.6–5$
Sample density self-absorption correction (for ^{137}Cs)	$< \pm 1?$

From a TPCV = 21.5% (grid 3) and a 12.2% process contribution follows the net CV due to spatial variability only, equaling 17.7%.

3.3. Confidence limits

From the above findings confidence limits (CL) for measured deposition values can be calculated. The CL are here defined as the limits of the range around the measured value within which the "true value" lies with a pre-set probability (95% in our case), considering only the stochastic uncertainties, i.e. if there were no systematic uncertainties which are in general unknown (otherwise one would try to correct them). For the discussion of the spatial variability of the ^{137}Cs deposition in the present investigation this circumstance does not matter, because the same systematic uncertainty is common to all samples.

However, this defined CL clearly do not represent the "full" uncertainty of the measured value, since they do not account for the systematic uncertainty which may contribute significantly under unfortunate conditions. In other words, the true deposition value lies (with 95% probability) in the interval $[(x + \Delta x) - \delta x, (x + \Delta x) + \delta x]$, where x denotes the measured value, Δx the systematic and δx the stochastic contribution of the "full" uncertainty, respectively, ($\delta x = CL$ as defined above) rather than in $[x - \delta x, x + \delta x]$.

The contribution to the systematic uncertainty due to profile cut-off, generally unknown, is a function of sampling (cut-off) depth, radionuclide migration time and soil properties (diffusion and convection constants). With the evaluation of more profile data of different soils available it may be possible to produce rough estimates of the cut-off uncertainty as a function of sampling depth and migration time, parameters which are readily available as opposed to diffusion and convection constants. However, such estimates cannot be made so far, therefore the CL calculation had to be restricted so as to account for spatial variability and process-related stochastic uncertainties only.

The normalised CL (in %) for n random samples out of an area that is equal to the size of grid 3 (10 000 m^2)

$$CL_{\text{grid 3}} = x(P)\sqrt{\frac{CV_{\text{grid 3, special}}^2}{n} + CV_{\text{process}}^2}, \tag{1}$$

where $x(p)$ is the Gauss distribution value for significance level p. If n samples are randomly taken out of an area of the size of grid 2 (= 100 m^2), but interpreted as an estimation of the deposition AM of a grid 3 size area, the respective CL is clearly larger:

$$CL_{\text{grid 2;est.for grid 3}} = x(P)\sqrt{CV_{\text{grid 3,spat}}^2 - CV_{\text{within,grid 3,spat.}}^2\left(1 - \frac{1}{n}\right) + CV_{\text{process}}^2}, \tag{2}$$

$$CV_{\text{spatial}}^2 = TPCV^2 - CV_{\text{process}}^2. \tag{3}$$
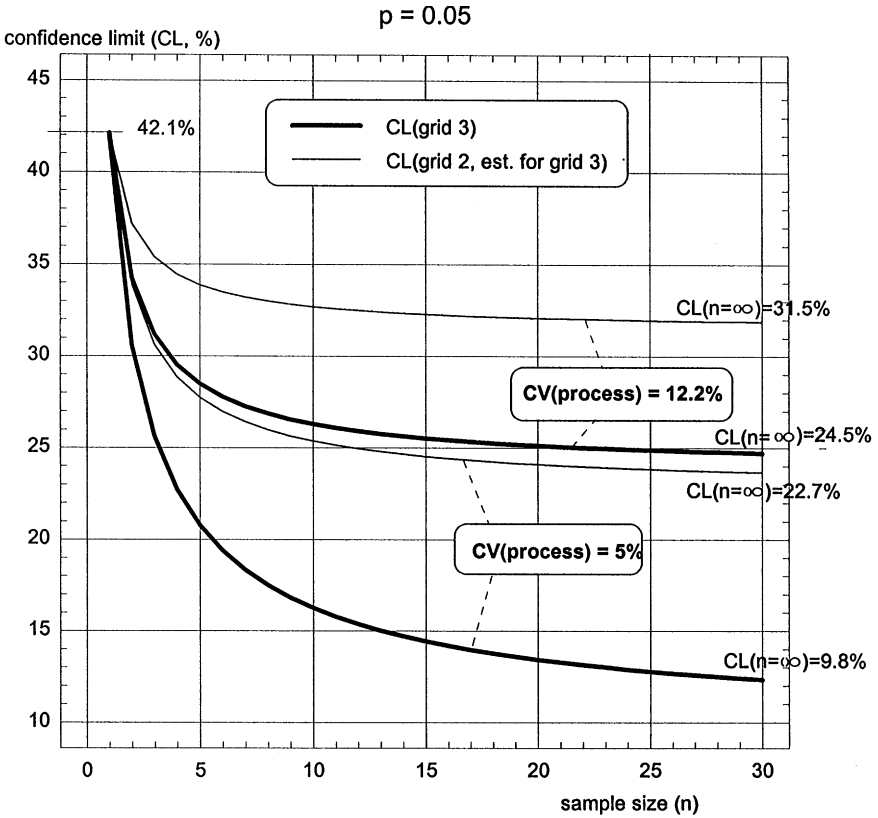
Fig. 3. Confidence limits for different sampling methods.

In our case, $CV_{grid\ 3,spat.} = 17.7\%$, $CV_{within,grid\ 3,spat.} = 14.5\%$, $CV_{process} = 12.2\%$; $p = 5\%$ and two-sided, $x(p) = 1.96$. An assumption made here is that $CV_{process}$ is independent of n, which is probably not entirely correct. For example the sample aliquot error probably even increases with n, because the more soil cores, the more total mass out of which the aliquot for measuring must be drawn thus increasing the resulting error. If the CVs are being estimated from an actual sample, $x(p)$ must be replaced by the value of the t-distribution $t(p, n - 1)$.

Fig. 3 shows the CLs as a function of the sample size n for two different sampling methods: (Samples from an area of the size of grid 3 (10 000 m^2); samples from an area of the size of grid 2 as an estimation for the AM of the grid 3 size area) and two different assumptions for the process-related uncertainty $CV_{process}$. Eq. (1) demonstrates, that with increasing contribution of the process related CV ($CV_{process}$) to the TPCV, the difference between the CLs resulting from the two sampling methods decreases. Consequently, as the assumption $CV_{process} = 5\%$ is probably the more realistic one, sampling is much more effective (in terms of minimising the CL) if done over the whole 1 ha area rather than just over a 100 m^2 one.
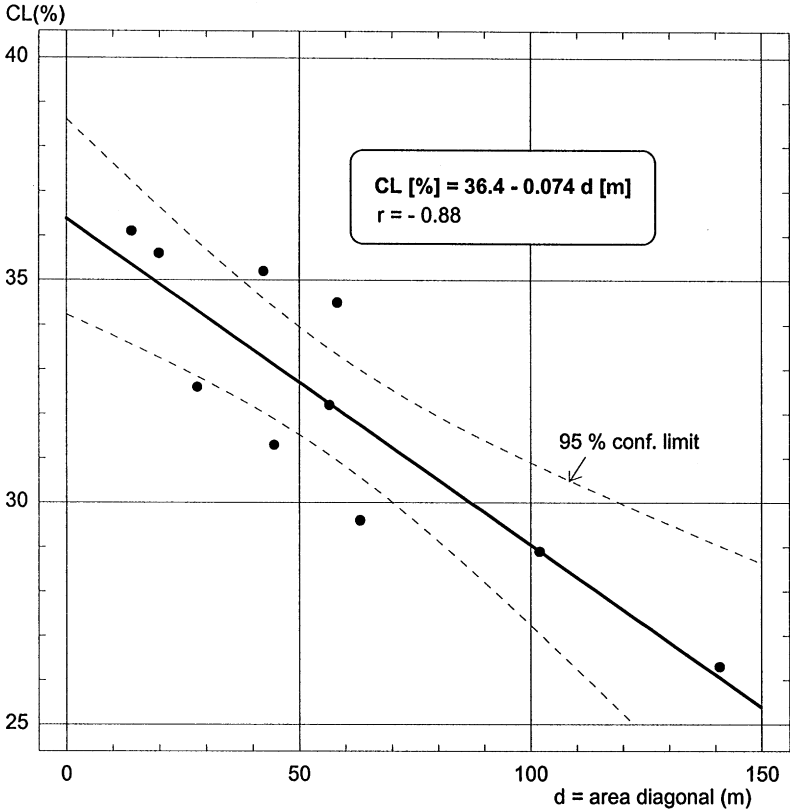
Fig. 4. Regression of confidence limits (CL) as a function of area size used for sampling. The data points were experimentally derived for $n = 10$ samples (standard sampling method).

The same procedure can be carried out using differently sized subgrids rather than only grid 2: Squares of different sizes from grid 3 were defined and the procedure described above was repeated using the data from these squares, in order to see the influence of the size of the area on the CL. The results are shown in Fig. 4 where CLs are plotted against the diagonal d of the areas defined and a linear regression analysis has been performed. The regression formula for CL(d) obtained in this way was used to calculate the CLs associated with the contamination data of the caesium map (UBA, 1996, Bossew et al., 1998), which had in fact been derived from sampling on very different sized areas.

4. Conclusions

The spatial variability of the ^{137}Cs deposition on a 1 ha area found in this investigation is 17.7%, with a total process coefficient of variation (TPCV) of 21.5%.
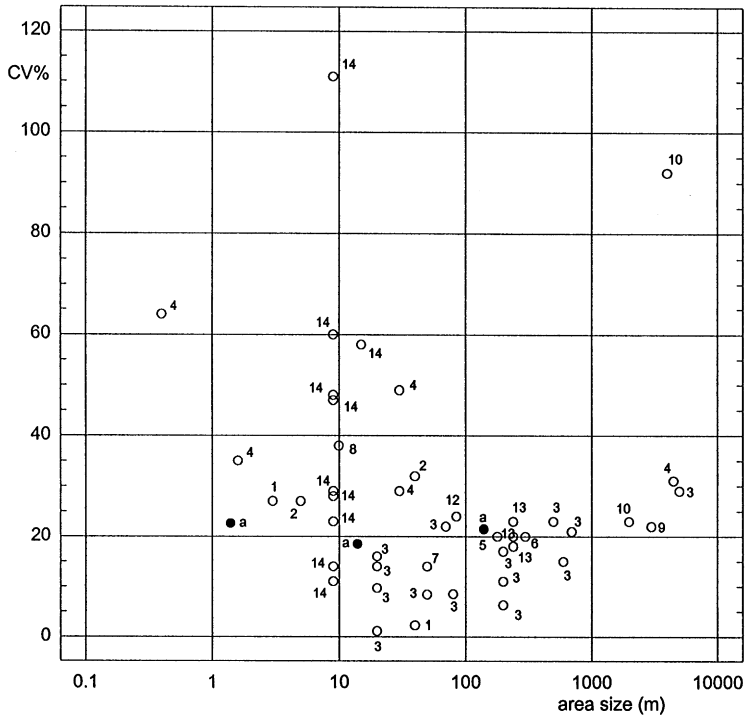
Fig. 5. Coefficient of variation (CV) of the ^{137}Cs-depositon as a function of the area size used for sampling. (1) Austrian Ecology Institute, in Lettner, Bossew and Hubmer (1994) (2) Austrian Federal Environmental Agency, radionuclides in a forest ecosystem, unpublished (3) Austrian Federal Environmental Agency, investigations of an agrarian ecosystem, unpublished (4) Austrian Ecology Institute, investigation of an area near Gmunden, Upper Austria, Lettner et al. (1994) (5) Bachhuber, Bunzl and Schimmack (1987) (global fallout) (6) Gustafsson and Skalborh et al. (1987) (7) Henrich, Friedrich, Weiß and Zapletal (1988) (8) Nyhan, White, Schofield and Trujillo (1983) (global fallout) (9) Padovani, Contento, Giovano and Malisan (1990) (10) Simopoulos (1989) (12) Mcgee, Keatinge, Synnott and Colgan (1995) (13) Sutherland and de Jong (1990) (14) Haugen (1992).

This value is in good agreement with the data reported in the literature, as can be seen in Fig. 5 showing literature data for TPCV's of ^{137}Cs deposition versus area size.

If the mean deposition density of a 1 ha area has to be determined, the samples should in fact be taken from points distributed over an area as close as possible to 1 ha, as 100 m^2 sub-areas, for example, do not yield estimates which are representative for the 1 ha area, or in other words, the result from any smaller area, seen as an estimate for the deposition density of the 1 ha area, has a larger uncertainty attached to it as if the same number of samples would have been taken from the 1 ha area. If 10 samples are collected on a 10 m × 10 m area in order to determine the nuclide inventory of an area of 100 m × 100 m, and assuming a process uncertainty of 12.2%, the 95% confidence limit would be 32.7%, as compared to 26.3%, if the samples were taken on the 100 m × 100 m area itself.

The CV as a measure of the variability actually overestimates it, because it ignores the correlation of the values from adjacent points. Therefore, derived parameters like confidence interval based on the CV concept are probably too conservative and should rather be considered as upper bounds.

Acknowledgements

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