SPATIAL DISTRIBUTION OF ^{137}CS IN FOREST SOILS OF SWITZERLAND

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Abstract. In the framework of the second Swiss forest soil inventory, ^{137}Cs-activity from 172 sites was measured systematically for the first time in the topmost soil layer (0–5 cm) and represented on a map. The spatial distribution of ^{137}Cs contamination was similar to the pattern observed in 1986 from dose equivalent measurements following the Chernobyl nuclear power plant accident. Forest soils from regions with high precipitation in 1986 showed a higher ^{137}Cs activity than regions with low precipitation. At sites with high caesium activities it was possible to discriminate between ^{137}Cs originating from global fallout of the fifties and sixties and ^{137}Cs from the Chernobyl accident. The results indicate that radiocaesium persists in the top soil layers and is recycled in forest ecosystems.

Keywords: Chernobyl, global fallout, vertical migration

1. Introduction

The nuclear power plant accident in Chernobyl at the end of April 1986 and the subsequent deposition of radioactive fallout over western Europe required the geographical delineation of exposure so that dose assessments of the population could be made by the corresponding national authorities. In Switzerland, maps were compiled for the distribution of radiocaesium (^{137}Cs) by using dose equivalent measurements (μR hr^{-1}, 1 m above ground) from permanent surveillance stations using a Geiger-Müller counter and by measuring the dose equivalent at single spots along defined routes and combining them with a few nuclide-specific soil activity measurements at selected, representative sites (Figure 1, Czarnecki *et al.*, 1986; Wernli, 1987). It became also obvious that the soil deposition was positively correlated with precipitation during the passage of the radioactive cloud (Wernli, 1987). Contamination of pastures, forage crops and agricultural products were measured predominantly, while forested areas were not included and the spatial distribution of soil contamination in natural or semi-natural ecosystems of Switzerland was not urgently needed.

Many studies have since shown that vertical migration of radiocaesium is slow in forest soils (e.g., Bergman, 1994; Rühm *et al.*, 1996) and up to 90% of the deposited radiocaesium is still found in the superficial soil layers (Desmet *et al.*, 1991), mainly the holorganic horizon (Belli *et al.*, 1994). When the possibility

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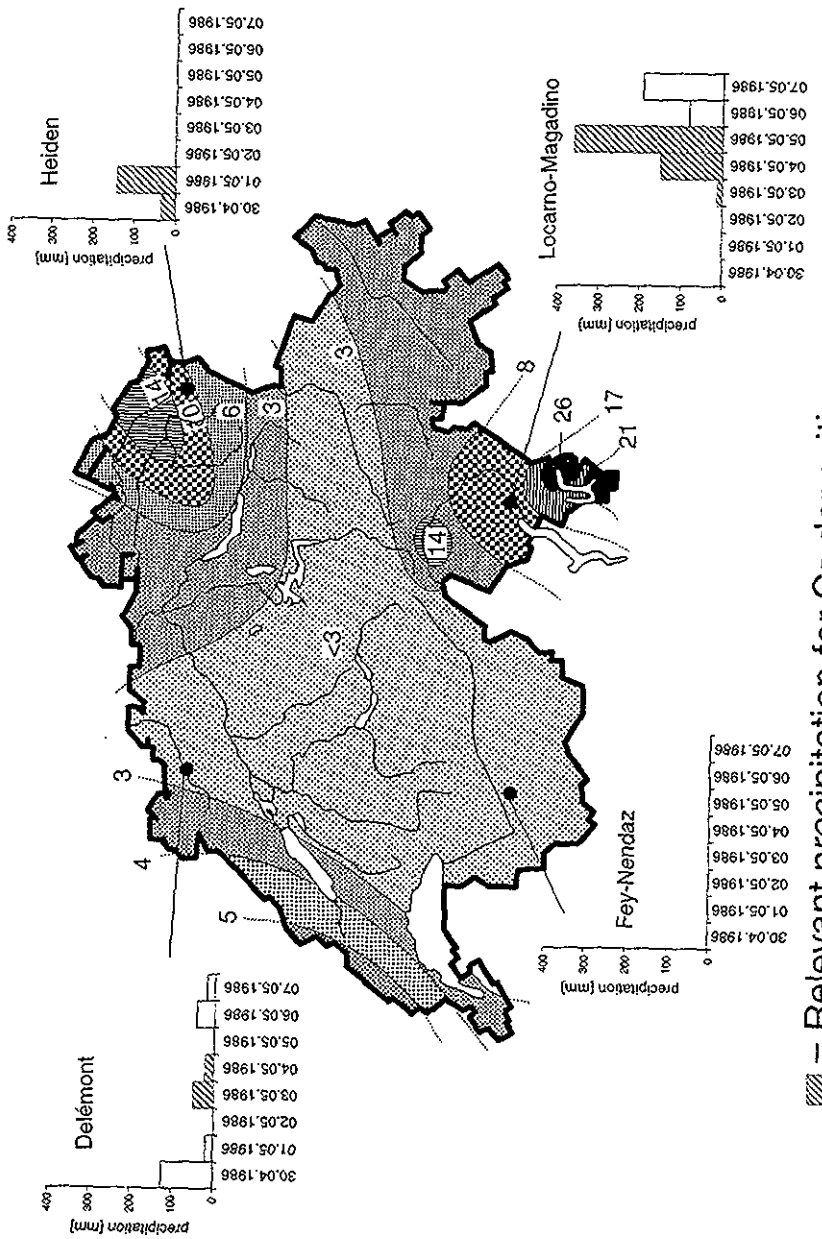

Figure 1. Calculated mean deposition of ^{137}Cs [kBq m^{-2}] in Switzerland after the Chernobyl accident based on dose equivalent measurements from Geiger-Müller instruments ($\mu R\ hr^{-1}$, 1 m above ground) and representative soil measurements made in 1986 (redrawn with permission of the Swiss Federal Nuclear Safety Inspectorate).

arose to obtain soil samples which were systematically collected from 172 sites during 1993, in the framework of the second forest soil inventory, we took the opportunity to measure their caesium activity to:

(1) Create a current map of the top soil contamination by caesium in forested soils of Switzerland;
(2) Compare the current caesium activity in forest top soil layers (0–5 cm) with the map of ground contamination compiled in 1986;
(3) Relate the measured activities with precipitation in 1986 for 4 regions; and
(4) Differentiate between caesium from global fallout and Chernobyl fallout for 12 sites.

2. Materials and Methods

Soil samples were collected systematically from 172 sites. Sites were selected by placing an 8 × 8 km grid over a geographic map of Switzerland and choosing the intersections located in a forested area. All samples were collected in 1993. At each site, one single sample of approximately 1 kg of soil was taken from each horizon, as well as from predefined depths, and thoroughly mixed to obtain representative samples. Samples were dried at 60 °C and sieved (2 mm mesh). An aliquot from each of the 172 top soil samples (generally A_h-horizon, 0–5 cm) was placed in a 50 mL polyethylene bottle for determining radiocaesium activity. The total activity of ^{134}Cs and ^{137}Cs was measured by γ-spectrometry on a solid state germanium detector linked to a multichannel-analyser. The counting time was adjusted so that the uncertainty for the counting statistics was less than 10% for the majority of the samples. Activity of samples was decay corrected to July 1, 1993. Differences between the soil densities of samples, which ranged from 0.4 to 1.2 g cm^{-3} (dry density), and the calibration standard solution (1 g cm^{-3}), were not corrected since the gamma rays of radiocaesium have high energies and thus sample self absorption is minimal (Hubbel, 1983). All results are given in Bq per kg soil dry weight. The homogeneity of one sample was verified by measuring 10 subsamples of the same horizon and site and determining the standard deviation.

Based on the ^{134}Cs/^{137}Cs ratio, which was 0.6 for the Chernobyl fallout in Switzerland in 1986 (Czarnecki *et al.*, 1986), and the half-lives of ^{134}Cs and ^{137}Cs, which are 2.1 and 30.2 yr, respectively, the ^{134}Cs/^{137}Cs ratio for July 1, 1993 was calculated. This ratio, and the fact that all the ^{134}Cs detected in the samples of 1993 originated from Chernobyl fallout, allowed us to calculate the portion of ^{137}Cs originating from the Chernobyl fallout. The difference between the calculated and measured ^{137}Cs-activity in 1993 corresponded to the ^{137}Cs-activity due to the global fallout.

Within the framework of the Swiss National Forest Inventory, topsoil samples had been collected at the intersections of a 1 × 1 km grid prior to the Chernobyl

accident and stored at the Swiss Federal Institute for Forest, Snow and Landscape Research (WSL). At each sampling point, one sample from the mineral soil layer (0–20 cm) was taken after removing the litter layer. By choosing intersections which had the same coordinates in the pre- and post-chernobyl sampling campaigns the opportunity arose to compare ^{137}Cs activity of soil samples from the same sites before and after the deposition of the Chernobyl fallout. Sampling points of the two campaigns were within a radius of 9 to 17 m at the corresponding intersections which had been marked during the first campaign.

Pre- and post-Chernobyl comparisons were done for four regions of Switzerland with different radioactive soil contamination based on the radiation map of 1986 (Figure 1). In each region three sites were selected. All samples were decay corrected to July 1, 1993.

Precipitation data for the period of the Chernobyl fallout in 1986 were used from the measuring stations of the Swiss Meteorological Institute, which are most representative for the four regions.

3. Results and Discussion

Figure 2 shows a map of the radioactive contamination of forest soil in Switzerland in 1993 resulting from the measurements of the topmost layer (0–5 cm). Based on our measurements we can define three arbitrary, not geostatistically interpolated ranges of soil contamination with ^{137}Cs and allocate Swiss regions to them:

(1) 0–100 Bq kg^{-1}: forest soil in the Swiss midland, the Rhone valley and the Canton of Grisons;
(2) 100–500 Bq kg^{-1}: forest soil at the north rim of the Jura mountains and Southwest of the Lake of Constance; and
(3) >500 Bq kg^{-1}: forest soil in the southern part of Switzerland with some values exceeding 1000 Bq kg^{-1}.

Although the activities are given in Bq kg^{-1} and are not representing the total caesium inventory of the corresponding site, the distribution pattern of ^{137}Cs in forest soils (Figure 2) agrees well with the map of radioactive deposition in Bq m^{-2} (Figure 1), which was created seven years earlier in the weeks following the Chernobyl nuclear power plant accident. In both figures, regions with a higher caesium deposition coincide. The use of the topsoil samples for this comparison is justified because it is generally accepted that the vertical migration of radiocaesium along the forest soil profile is slow (Calmon *et al.*, 1996; Tikhomirov and Shcheglov, 1994) and that the activity is confined mainly to the superficial organic layers (Ritchie *et al.*, 1970, Rühm *et al.*, 1996, Velasco *et al.*, 1993). This view is supported by the fact that even global fallout caesium can still be detected in our topsoil samples (cf. Table I) and also was verified at six of our sites by measuring

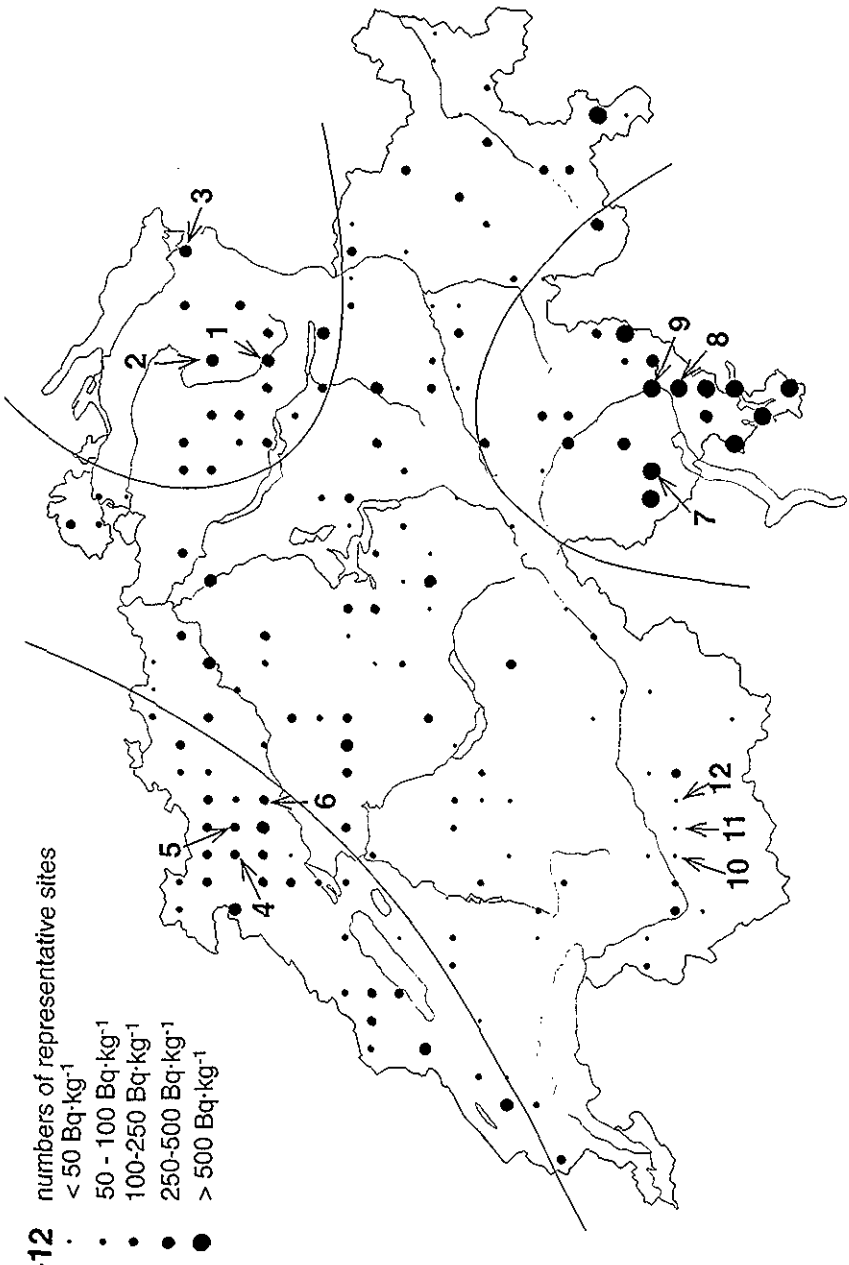

Figure 2. Activity of ^{137}Cs [Bq kg^{-1}] in the topsoil layer (0–5 cm) of forest soils collected in the framework of the second forest soil inventory 1993 in Switzerland (*radionuclide decay was considered to the reference date July 1, 1993*).

the caesium activity over the whole soil profile (Table II). ^{134}Cs-activities in the soil layers deeper than 5 cm are not included in the tables because the activities were too small and the counting errors too high to make a conclusive statement. The good agreement between calculated ($^{137}Cs_{total}/^{134}Cs$ ratio is 18.9 at July 1, 1993) and measured ^{134}Cs-activities in the 1993 samples (Table II) further confirms that radiocaesium (^{137}Cs and ^{134}Cs) of the Chernobyl fallout has not migrated into deeper soil layers until now. Contrary to agricultural soils with a high clay content, the low clay content in forest soils would suggest a high migration rate of caesium. However, the high fraction of organic matter, as well as temporary immobilisation and recycling processes of radiocaesium induced by soil microorganisms in the topmost soil layer cause the opposite (Bunzl *et al.*, 1995).

As shown earlier (Wernli, 1987), the ground deposition of Chernobyl fallout was well correlated with rainfall in wide areas of Switzerland. The increased caesium deposition due to precipitation in 1986 (Figure 1) was still apparent from soil caesium activity collected in 1993 (Figure 2). In the Rhone valley both the caesium activity in soil and the ground deposition were low with 12–36 Bq kg^{-1} and <3 kBq m^{-2}, respectively (Table I, sites No. 10–12, Figure 2). No precipitation was registered for this region at the meteorological station of Fey-Nendaz during the whole period when the radioactive cloud passed (Figure 1). The corresponding values for the region of southern Switzerland are opposite: 1461–1896 Bq kg^{-1} and 8 kBq m^{-2}, respectively combined with heavy rainfall registered at Locarno during the whole period. The sites in the north eastern and north western part of Switzerland show intermediate values.

In Table I, measured and calculated ^{137}Cs-activities originating from global fallout are compared with each other based on the known $^{134}Cs/^{137}Cs$ ratio in the 1993 soil samples and the soil samples collected before the Chernobyl accident. The calculated values are generally higher than the measured ones except at the sampling sites representing the Rhone Valley (No. 10, 11 and 12) where the formation of the $^{134}Cs/^{137}Cs$ ratio was not possible (^{134}Cs below detection limit in the samples of 1993). The level of the differences between calculated and measured ^{137}Cs activities depends on the regions investigated. Differences were most significant in the southern part of Switzerland (sampling sites No. 7, 8 and 9), lowest in the northwestern part of Switzerland (sampling sites No. 4, 5 and 6) and intermediate in the north-eastern region of Switzerland (sampling sites No. 1, 2 and 3).

Although the counting errors for the ^{134}Cs measurements are high in some cases (cf. Table I) it was demonstrated before that the measured ^{134}Cs-activities correspond with the expected values and the Chernobyl caesium still is located in the top soil layer. The reason for the discrepancies between measured and calculated activities of the global fallout caesium are difficult to explain and remain subject of assumptions:

TABLE I

Caesium activity in the top soil (0–5 cm) of 12 forested sites collected before and after the Chernobyl accident in 1986 and in 1993, respectively. Comparison between the measured and calculated ^{137}Cs-activity of the global fallout on the basis of the soil measurements in 1993 and the known ^{134}Cs/^{137}Cs ratio which was 0.6 for the Chernobyl fallout in Switzerland in 1986 (activities were corrected to July 1, 1993)

Site No. (see. Figure 2)	^{137}Cs-activity 1993 [Bq kg^{-1}]	^{134}Cs-activity 1993 [Bq kg^{-1}]		Global fallout calculated from 1993 samples [Bq kg^{-1}]	Global fallout measured in 1986 samples [Bq kg^{-1}]	Counting error 2σ [%] ^{137}Cs, 1993	Counting error 2σ [%] ^{134}Cs, 1993	^{137}Cs/^{134}Cs 1993
		measured	calculated[a]					
1	466	20	25	148	53	3	12	23
2	414	21	22	93	44	3	10	20
3	398	21	21	70	11	3	11	19
4	102	4	5	42	26	4	52	27
5	144	8	8	15	16	4	23	18
6	138	6	7	49	58	4	35	24
7	1646	91	87	217	188	2	3	18
8	1896	97	100	378	17	2	4	20
9	1461	76	77	265	2	2	5	19
10	12	0	1	–	13	15	–	–
11	30	0	2	–	5	9	–	–
12	36	0	2	–	4	7	–	–

[a] Calculated with the ratio of $^{137}Cs_{total}$/^{134}Cs which was 18.9 at July 1, 1993.

TABLE II

Vertical distribution of ^{137}Cs (bomb plus Chernobyl fallout) in 6 forest soil profiles in Switzerland

Depth of soil layer	Site[a] 1	2	3	7	8	9
– (cm) –	([Bq kg^{-1}] dw[b])					
0–5	464[c]	433[c]	323[c]	1390[c]	1563[c]	1042[c]
5–8					183[c]	
5–10	188[c]	136[c]	160[c]	159		60
10–12				477		
10–15		54[c]			22	
10–20	8		63[c]			0
20–40	12	0	12	4	4	
40–60				0	5	0
60–80	0	0			0	
120–140			7		0	
130			0			

[a] Cf. Figure 2 for site location.
[b] Dry weight.
[c] A_h horizon.

(i) Bomb fallout caesium has migrated into deeper soil layers during the last 20 to 30 yr which is supported by the numbers given in Figure 2 and the fact that Chernobyl caesium is still located in the A_h horizon as shown before.

(ii) Heavy rainfall during the deposition of the Chernobyl fallout may have caused runoff (Bergman, 1994) at the sampling sites No. 8 and 9 which are situated at a steep slope. As a consequence the calculated ^{137}Cs-activity is too high.

(iii) ^{137}Cs-activity in soil was reduced because of an increasing plant availability and uptake over the years (Thiry *et al.*, 1999).

Other reasons may be different sampling techniques used which is less probable as they would cause a systematic error, or the well-known heterogeneity of the horizontal contamination which could not be checked in this case as only one sample was collected per site and horizon. Within samples, ^{137}Cs was homogenous distributed which was verified by the measurement of 10 subsamples of a sample from site No. 9 in 50 mL polyethylene bottles (30.8±1.7 Bq/subsample, counting error <5%).

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References

Belli, M., Sansone, U. and Menegon, S.: 1994, *Sci. Total Environ.* **157**, 257.

Bergman, R.: 1994, 'The Distribution of Radioactive Caesium in Boreal Forest Ecosystems', in H. Dahlgaard (ed.), *Nordic Radioecology, The Transfer of Radionuclides Through Nordic Ecosystems to Man, Studies in Environmental Science* **62**, pp. 335–379.

Bunzl, K., Kracke, W., Schimmack, W. and Auerswald, K.: 1995, *J. Environ. Radioactivity* **28**(1), 17.

Calmon, P., Jouve, A. and Maubert, H.: 1996, *Journal of Radioecology* **4**(1), 27.

Czarnecki, J., Cartier F., Honegger P. and Zurkinden, A.: 1986, 'Bodenverstrahlung in der Schweiz aufgrund des Reaktorunfalls in Chernobyl', in L. André *et al.* (eds.), *Radioaktivitätsmessungen in der Schweiz nach Tschernobyl und ihre Wissenschaftliche Interpretation.* Tagungsbericht Band 1, 20–22.10.86, Universität Bern, p. 581.

Desmet, G., Nassimbeni P. and Belli M.: 1990, *Transfer of Radionuclides in Natural and Semi-Natural Environments*. Elsevier Science Publishers Ltd, London a.o., p. 693.

Hubbel, J. H.: 1983, *Int. J. Appl. Radiat. Isot.* **33**, 1269.

Ritchie, J. C., Clebsch, E. E. C. and Rudolph, W. K.: 1970, *Health Phys.* **18**, 479.

Rühm, W., Steiner, M., Wirth, E., Dvornik, A., Zhuchenko, T., Kliastshtorin, A., Rafferty, B., Shaw, G. and Kuchma, N.: 1996, 'Dynamic of Radionuclides Behaviour in Forest Soils', in A. Karaoglou *et al.* (eds.), *The Radiological Consequences of the Chernobyl Accident*, **EUR 16544 EN**, pp. 225–228.

Thiry, Y. Riesen, T. and Lewyckj, N.: 1999, 'Woody Biomass Production and Radiocaesium Accumulation Rate in Pine (*Pinus silvestris* L.) from a Contaminated Forest in the Vetka Area', in I. Linkov (ed.), *Contaminated Forests: Recent Developments in Risk Identification and Future Perspectives*, NATO ASI Series 2-, Kluwer Academic Publishers, Dordrecht (in press).

Tikhomirov, F. A. and Shcheglov, A. I.: 1994, *Sci. Total Environ.* **157**, 45.

Velasco, R. H., Belli, M., Sansone, U. and Menegon, S.: 1993, *Health Phys.* **64**(1), 37.

Wernli, C.: 1987, *Radiat. Prot. Dosim.* **19**(4), 235.