

^{137}Cs AND ^{90}Sr CONCENTRATION IN LOMBARDIA SOILS: MEASUREMENTS AND MIGRATION PROFILES ANALYSIS

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Introduction

It is well known that environmental behaviour of different radionuclides in natural ecosystems depends on their chemical-physical properties, their speciation and, to some extent, their origin.

After the accident at the Chernobyl nuclear power plant, considerable fallout occurred in Northern Italy in early May 1986. Because of rainfall during the first days after the explosion, the most affected area in Lombardia was the so-called 'Triangolo Lariano', lying between the cities Como and Lecco. Moreover, in Lombardia Chernobyl fallout mainly occurred in areas heavily interested by the previous nuclear weapons testing fallout of the 1950s and early 1960s.

ARPA Lombardia (Regional Environmental Protection Agency of Lombardia) is in charge of monitoring certain environmental radioactive pollution; moreover, ARPA Lombardia promotes programs intended to improve knowledge of the behaviour of the major pollutants in the environment. This work is aimed at the study of artificial radiocontamination and the long-term behaviour of radionuclides in soils.

Undisturbed grassland soils were sampled in 8 different Lombardia sites: three sites located in a mountainous area in the north-west of Lombardia (namely Ballabio, Valbrona and Carenno), three sites located in a hill central Lombardia area (Cantù, Montevecchia and Besana Brianza) and two sites located in a flat southern Lombardia area (Magenta and Trucazzano). These three areas were selected because of their differences in average annual rainfall: higher in northern Lombardia and lower in south.

The samples were collected during 1996, ten years after the Chernobyl accident. Selected sites were reported to have been undisturbed since 1986, but actually we suppose that these sites have been undisturbed for more than 50 years.

In each site 5 cm thick sample layers were separately collected down to a depth of 40 cm; ^{137}Cs and ^{134}Cs were measured by high resolution gamma ray spectrometry on dried and homogenised samples and ^{90}Sr by total beta counting after chemical separation.

Chemical-physical features (organic fraction, pH, cation exchange capacity, calcium concentration and texture) were determined too.

Methods

In each of the 8 selected sampling sites soil was collected down to a depth of 40 cm by coring. Soil samples from 0 to 20 cm were divided in 4 layers, each 5 cm thick; soil samples from 20 to 40 cm were divided in 2 layers, each 10 cm thick.

Each sample was dried in an oven at 40°C to constant weight; the dry soil was crushed and sieved (2 mm). The resulting sample was weighed and transferred into a Marinelli beaker. ^{137}Cs and ^{134}Cs

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concentrations were measured using a high-resolution gamma ray spectrometer (HPGe detector - 30 % efficiency at 1173 keV).

A fraction of the sample was used for ^{90}Sr determination. 500 g of soil were heated in a muffle furnace (450°C, 12 hours) and transferred into a flask; 1 l of HNO_3 and 2 g of strontium nitrate were added, the mixture was reflux boiled for 6 hours and then filtered (Oughton, 1993). 500 g of oxalic acid and 12 g of sodium acetate were added to the filtrate. The pH was brought to 4 by adding ammonium hydroxide to obtain oxalate precipitation. Oxalates were filtered, dried and heated in a muffle furnace at 750°C. Residual salts were converted into nitrates and dissolved in 100 ml of 0,1 M HCl. 1 ml of the solution was saved for strontium yield determination by atomic absorption spectrometry, the remaining was extracted twice by a 20% HDEHP (bis-2-ethyhexyl phosphoric acid) solution in toluene. Yttrium nitrate (10 mg) was added to the aqueous phase which was stored for ^{90}Y ingrowth (2 weeks). Yttrium was then extracted by a 5% HDEHP solution in toluene. The organic phase was extracted twice by 3 M HNO_3 and yttrium oxalate was precipitated from the nitric solution by oxalic acid addition at pH 2,5.

The yttrium oxalate was deposited on a filter and counted using a plastic scintillation counter. Each sample was counted four times for 8 hours each time and the correspondence of the experimental sample decay time to the expected one ($T_{1/2} (^{90}\text{Y}) = 64$ hours) was checked; samples which gave experimental $T_{1/2}$ out of the correct time range ($64 \pm 10\%$ hours) were discarded and the complete analysis was repeated.

The yttrium yield was finally determined by gravimeter method; the filter was burned at 900°C after counting and weighed.

Chemical-physical features of each sample were determined according to official Italian procedures (G.U. 79/1992). Both pH in water and in KCl solution were measured. Organic fraction was determined by Walkley and Black method. Calcium concentration was determined by atomic absorption spectrometry on nitric leachate. To measure exchange capacity, barium chloride triethanolamine method was used. Texture was determined by sedimentation method and classified on the basis of USDA textural diagram.

Results

Table 1 resumes ^{137}Cs and ^{90}Sr total deposition values, measured summing radionuclides content of different soil sections (from 0 to 40 cm depth). Sites are listed from northern to southern.

Table 1

Sampling site	^{137}Cs Bq/m ²	^{90}Sr Bq/m ²	$^{137}\text{Cs}/^{90}\text{Sr}$
Ballabio (M)	25680	3260	8
Valbrona (M)	39580	3000	13
Carenno (M)	19010	2140	9
Cantù (H)	11690	2290	5
Montevecchia (H)	12050	680	18
Besana Brianza (H)	8330	440	19
Magenta (F)	15050	370	41
Trucuzzano (F)	19460	510	38

M: mountain; H: hill; F: flat

^{137}Cs deposition values range from about 8000 Bq/m² (Besana Brianza) to 40000 Bq/m² (Valbrona); ^{90}Sr values range from 370 Bq/m² (Magenta) to about 3300 Bq/m² (Ballabio). ^{137}Cs content is

always higher than ^{90}Sr one, and their ratio ranges from 5 (Cantù) to 41 (Magenta). Higher radionuclide content is generally measured in northern Lombardia sites.

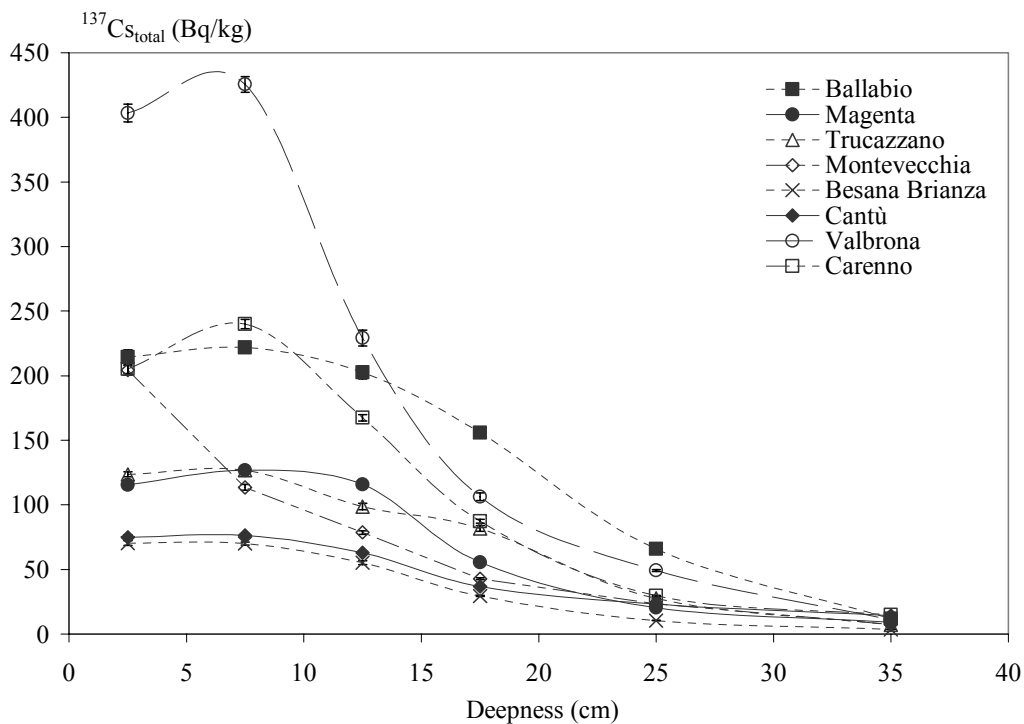
Caesium results

Results of ^{137}Cs content in different samples are reported in table 2. ^{137}Cs depth profiles at different sites are presented in figure 1.

Table 2

Sampling site	^{137}Cs (Bq/kg) - Depth profile					
	0-5 cm	5-10 cm	10-15 cm	15-20 cm	20-30 cm	30-40 cm
Ballabio	215 ± 6	222 ± 3	203 ± 5	156 ± 4	66 ± 2	11.8 ± 0.2
Valbrona	403 ± 7	425 ± 6	229 ± 6	106 ± 3	49.2 ± 0.7	10.9 ± 0.2
Carenno	205 ± 4	240 ± 4	167 ± 3	87 ± 2	29.7 ± 0.4	14.7 ± 0.4
Cantù	75 ± 2	76 ± 2	63 ± 1	37 ± 1	23.2 ± 0.3	14.2 ± 0.2
Montevecchia	204 ± 4	114 ± 2	79 ± 2	43 ± 1	23.6 ± 0.6	7.5 ± 0.2
Besana Brianza	71 ± 2	70 ± 1	55 ± 1	30 ± 1	10.5 ± 0.1	3.55 ± 0.09
Magenta	116 ± 3	127 ± 2	116 ± 2	55 ± 1	20.6 ± 0.3	9.2 ± 0.3
Trucazzano	124 ± 2	127 ± 2	99 ± 3	82 ± 2	27.6 ± 0.4	7.0 ± 0.1

FIGURE 1 : Total ^{137}Cs depth profile



In most cases ^{137}Cs concentration shows a maximum between 5 and 10 cm. Grass radical apparatus is usually contained in the first 5 cm of soil; therefore caesium uptake by grass roots should reduce the amount of radionuclide in the first layer.

Fifty percent of radiocaesium is contained in the first 10 or 15 cm layer, eighty percent is contained in the first 20 cm; caesium content of the deepest section (30-40 cm) is generally lower than 5%. Therefore we can reasonably assume that total radiocaesium fallout is actually contained in the first 40 cm of soil.

Caesium depth profiles follow an exponential law as 10 cm layers are considered. Resulting $S^{1/2}$ (half value deepness) ranges from 5 to 9 cm.

During the Chernobyl accident great amounts of ^{134}Cs were thrown into the atmosphere; $^{137}\text{Cs}/^{134}\text{Cs}$ ratio R , evaluated on the basis of Chernobyl reactor radionuclide content, was equal to 2. Ratio R was expected to change during the following years as a consequence of different half decay times of the two radionuclides ($T_{1/2} (^{134}\text{Cs}) = 2.065 \text{ y}$, $T_{1/2} (^{137}\text{Cs}) = 30.17 \text{ y}$).

Since ^{134}Cs was absent in environmental samples collected before the Chernobyl accident (ENEA 1985), it is possible to evaluate expected ^{137}Cs content of soils due to the Chernobyl accident through the following equations:

$$^{137}\text{Cs}_{\text{Chernobyl}} = ^{134}\text{Cs} * R_t \quad \text{and} \quad ^{137}\text{Cs} = ^{137}\text{Cs}_{\text{Chernobyl}} + ^{137}\text{Cs}_{\text{Nucl. Weap.}}$$

where ^{134}Cs and ^{137}Cs are the measured radionuclide concentrations and R_t is the value of the expected ratio $^{137}\text{Cs}/^{134}\text{Cs}$ at the sampling date.

Calculated ^{137}Cs concentrations due to nuclear weapons tests are reported in table 3. ^{137}Cs is almost constant in each layer up to 20 or 30 cm depth (Fig. 2); slightly higher concentrations are observed in the 10-20 cm layer. Lower values are always measured in the deepest layer (30-40 cm). Average concentration values in 0-20 cm layer ranges from about 20 to 40 Bq/kg.

Table 3: Nuclear test ^{137}Cs concentration in soil layers

Sampling site	$^{137}\text{Cs}_{\text{nuclear weapons}}$ (Bq/kg) - Depth profile						Avg. Conc. 0-20 cm (Bq/kg)	% of ^{137}Cs due to nucl. weap.
	0-5 cm	5-10 cm	10-15 cm	15-20 cm	20-30 cm	30-40 cm		
Ballabio	34 ± 14	40 ± 13	47 ± 12	48 ± 9	32 ± 5	2.0 ± 1.4	43	26
Valbrona	27 ± 23	43 ± 23	44 ± 14	16 ± 8	20 ± 3	6.0 ± 0.9	33	16
Carenno	45 ± 12	39 ± 13	51 ± 10	35 ± 6	12 ± 2	0.7 ± 1.4	42	24
Cantù	10 ± 7	15 ± 6	20 ± 5	19 ± 2	17 ± 1	11.3 ± 0.2‡	17	49
Montevecchia	16 ± 13	22 ± 7	26 ± 6	11 ± 4	11 ± 3	0.4 ± 0.2‡	18	22
Besana B.za	30 ± 5	19 ± 5	17 ± 5	19 ± 2	7.8 ± 0.1‡	1.3 ± 0.1‡	21	44
Magenta	25 ± 9	26 ± 8	28 ± 7	32 ± 3	13.4 ± 1.2	6.2 ± 0.2‡	28	36
Trucuzzano	7 ± 8	20 ± 8	19 ± 6	25 ± 5	12 ± 2	1.3 ± 1.2	18	21

‡: value calculated considering ^{134}Cs minimum detectable activity

The last column in table 3 shows % of current ^{137}Cs contamination due to nuclear weapons over total ^{137}Cs . In 5 sites most of the ^{137}Cs contamination is due to the Chernobyl accident; in 3 sites (Cantù, Besana Brianza and Magenta) a relevant fraction of the current ^{137}Cs contamination (from 36% to 49%) is still due to nuclear weapon test fallout.

Depth profile of total ^{137}Cs and calculated Chernobyl ^{137}Cs are quite similar since ^{137}Cs from weapons tests gives only a minor and constant contribution (fig. 1 and 3).

FIGURE 2 : Nuclear weapons ^{137}Cs depth profile

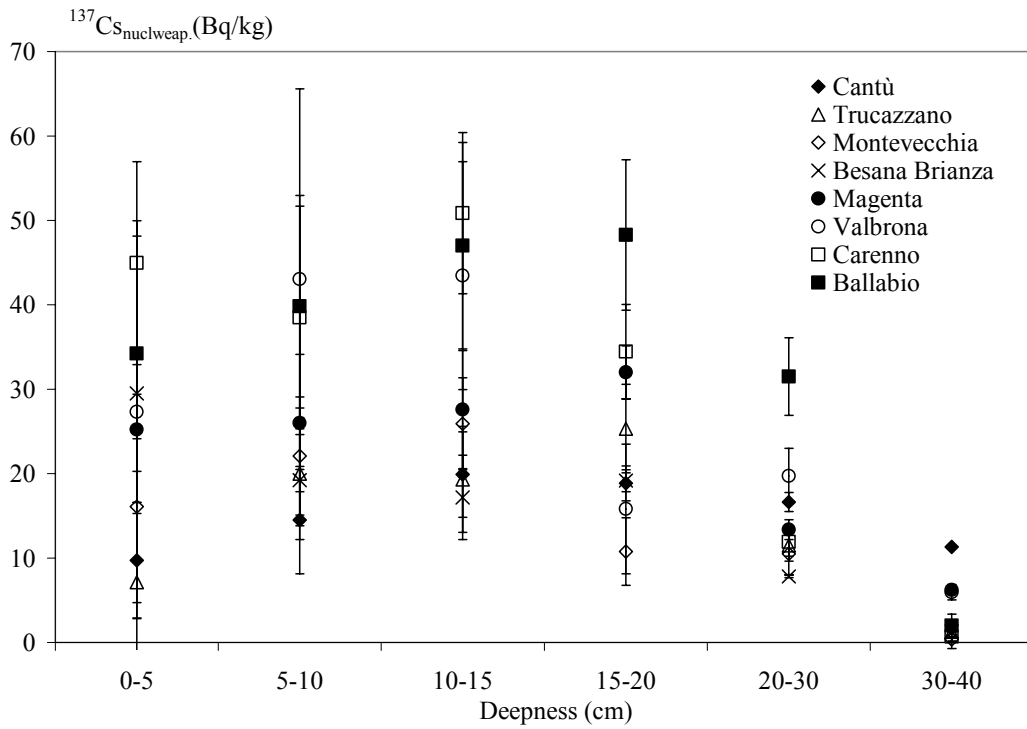
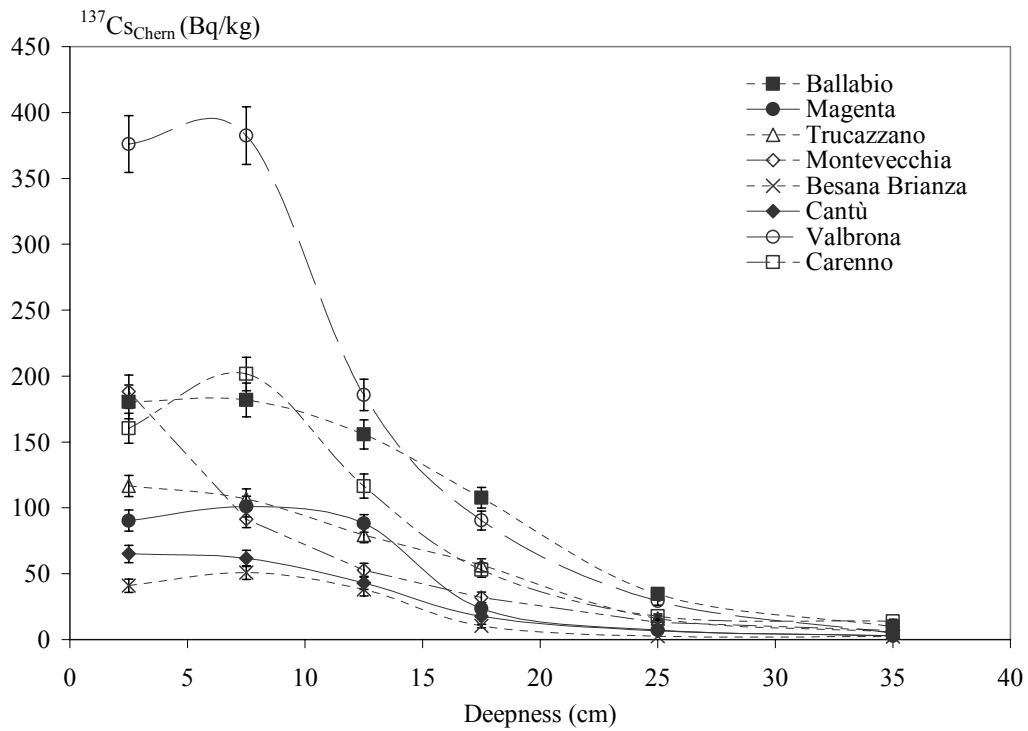


FIGURE 3 : Chernobyl ^{137}Cs depth profile



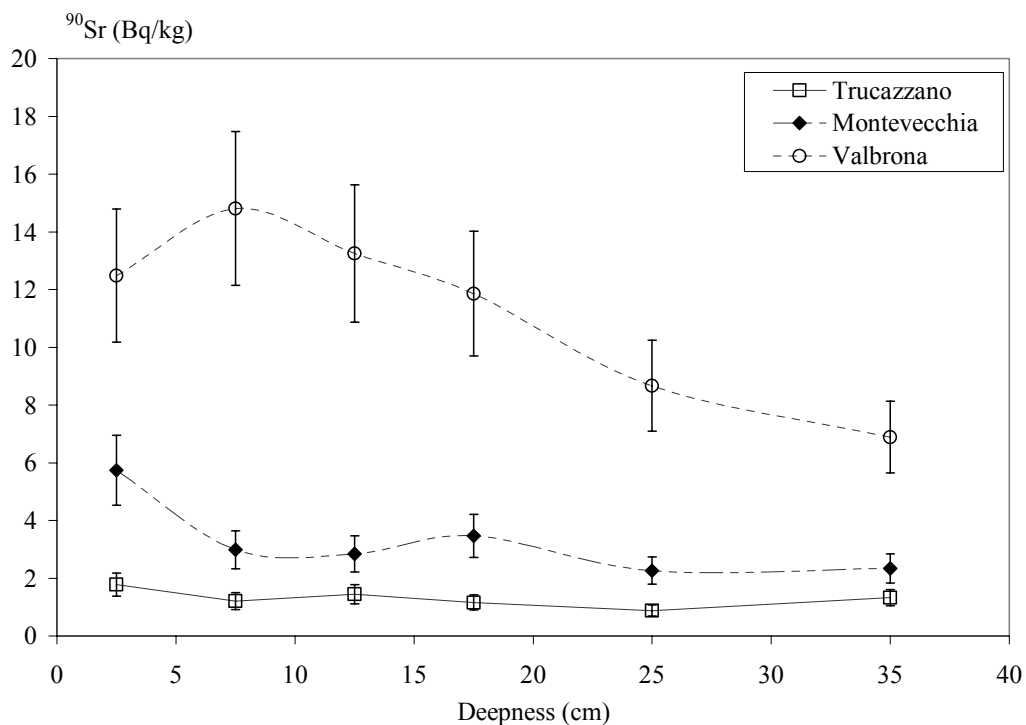
Strontium results

^{90}Sr concentration in different sample layers are reported in table 4. Examples of measured depth profiles are shown in figure 4.

Table 4

Sampling site	^{90}Sr (Bq/kg) - Depth profile						Avg. Conc. 0-40 cm (Bq/kg)
	0-5 cm	5-10 cm	10-15 cm	15-20 cm	20-30 cm	30-40 cm	
Ballabio	15.0 ± 3.2	7.7 ± 1.7	17.4 ± 3.8	16.5 ± 3.5	14.6 ± 3.0	9.0 ± 1.9	13
Valbrona	12.5 ± 2.3	14.8 ± 2.7	13.2 ± 2.4	11.9 ± 2.2	8.7 ± 1.6	6.9 ± 1.2	10
Carenno	14.7 ± 2.8	15.2 ± 2.7	12.1 ± 2.2	13.8 ± 2.5	10.2 ± 1.9	5.6 ± 1.0	11
Cantù	8.4 ± 1.7	6.1 ± 1.2	3.5 ± 0.9	4.9 ± 1.0	5.9 ± 1.1	7.1 ± 1.3	6.2
Montevecchia	5.7 ± 1.2	3.0 ± 0.7	2.8 ± 0.6	3.5 ± 0.7	2.3 ± 0.5	2.3 ± 0.5	2.9
Besana B.za	2.1 ± 0.5	1.6 ± 0.4	1.6 ± 0.4	1.8 ± 0.4	1.1 ± 0.3	0.9 ± 0.2	1.3
Magenta	0.8 ± 0.2	1.5 ± 0.3	1.6 ± 0.3	1.5 ± 0.3	0.9 ± 0.2	0.2 ± 0.2	1.1
Trucazzano	1.8 ± 0.4	1.2 ± 0.3	1.5 ± 0.3	1.2 ± 0.3	0.9 ± 0.2	1.3 ± 0.3	1.2

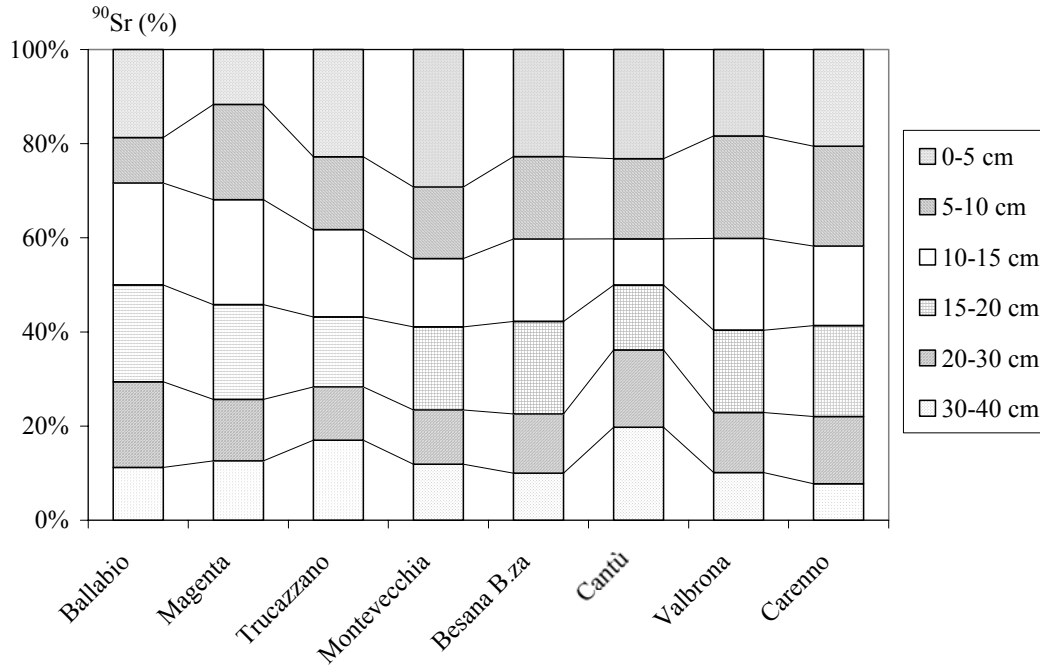
FIGURE 4: Examples of ^{90}Sr depth profiles



Higher values were found in northern sites. ^{90}Sr content of different soil sections, expressed as % of total content, is shown in figure 5. Strontium distribution in the first 40 cm of soil is almost constant. We can therefore suppose that a relevant amount of ^{90}Sr is actually contained in soil layers deeper than 40 cm, as a consequence of strontium high mobility in soil (Riise, 1990).

Current ^{90}Sr soil contamination is due both to Chernobyl and to nuclear weapon fallout, but there is no way to distinguish their different contributions.

FIGURE 5: ^{90}Sr % distribution in soil layers



Soil analysis

Each soil layer was analysed with regard to pH, cationic exchange capacity and concentration of organic matter. Percent of clay, silt and sand values were used to classify texture on the basis USDA textural diagram.

Average values measured in each site are reported in table 5.

Table 5: Chemical and textural features of soils

Sampling site	pH in H ₂ O	Cationic Exchange Capacity Cmol/kg	Organic Matter Conc. g/kg	% Clay	% Silt	% Sand	Soil Texture Class
Ballabio	6,43	18,38	33,4	6,3	50,7	43,0	Silt Loam
Valbrona	6,58	22,60	24,9	7,2	42,4	50,4	Loam
Carenno	5,99	22,17	28,5	7,8	48,6	43,6	Loam
Cantù	5,80	15,71	20,1	6,0	43,5	50,5	Sandy Loam
Montevecchia	5,05	22,92	18,6	7,5	53,8	38,7	Silt Loam
Besana Brianza	7,41	18,48	27,6	6,7	49,6	43,7	Silt Loam
Magenta	6,39	17,23	33,4	7,0	59,4	33,6	Silt Loam
Trucazzano	7,28	16,78	25,3	6,7	52,3	41,0	Silt Loam

All examined soils exhibit similar characteristics: pH is generally neutral, with the exception of Cantù and Montevecchia acid soils; cationic exchange capacity is from medium to high; organic matter content is always high. The last column of table 5 lists the resulting texture classes.

Discussion and conclusion

Measured radiocontamination levels display marked variation from site to site, even though close to each other; generally northern sites are more contaminated than southern ones.

^{137}Cs is contained in the first 40 cm of soil; a 40 cm deep sampling is therefore sufficient to measure total caesium radiocontamination.

Depth profile of ^{137}Cs shows a maximum between 5 and 10 cm; its concentration is usually lower in 0-5 cm layer than in 0-10 cm one. Therefore special care must be paid when comparing results of different studies where ^{137}Cs content of first layers of soil is measured.

^{137}Cs due to nuclear weapon testing fallout, which occurred about 45 years ago, is still contained in the first 40 cm deep soil, with maximum values in the 10-20 cm layer. Most Chernobyl ^{137}Cs is placed in the first 20 cm of soil. Caesium downward migration speed is extremely low; Chernobyl caesium is thus expected to remain in upper soil layers for many decades.

^{90}Sr depth distribution in soils is almost constant up to 40 cm; thus a 40 cm deep sampling will not likely be able to collect total deposited strontium. Depth profile analysis shows that strontium mobility is higher than caesium one. It can not be distinguished if present strontium contamination is due to nuclear weapon testing or to Chernobyl fallout contribution.

No general conclusions can be drawn about $^{137}\text{Cs}/^{90}\text{Sr}$ ratio both because of its dependence on sampling site and on soil layer. When considering $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in the total 0-40 layer, higher values are found in flat sites (40) than in hilly (20) and mountainous ones (10).

Chemical and textural features of examined soils are quite similar. Some correlations can be found between ^{137}Cs concentration and soil characteristics when single layer results are considered. Obtained data however are not sufficient to draw any conclusions. No correlations were found with ^{90}Sr concentrations.

Further work is in progress to analyse samples collected in other areas of the region; plutonium radionuclides will be determined too.

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