Deposition of artificial radionuclides from atmospheric Nuclear Weapon Tests estimated by soil inventories in French areas low-impacted by Chernobyl

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A B S T R A C T

Soil inventories of anthropogenic radionuclides were investigated in altitudinal transects in 2 French regions, Savoie and Montagne Noire. Rain was negligible in these 2 areas the days after the Chernobyl accident. Thus anthropogenic radionuclides are coming hypothetically only from Global Fallout following Atmospheric Nuclear Weapon Tests. This is confirmed by the isotopic signatures ($^{238}Pu/^{239+240}Pu$; $^{137}Cs/^{239+240}Pu$; and $^{241}Am/^{239+240}Pu$) close to Global Fallout value. In Savoie, a peat core age-dated by $^{210}Pb_{ex}$ confirmed that the main part of deposition of anthropogenic radionuclides occurred during the late sixties and the early seventies. In agreement with previous studies, the anthropogenic radionuclide inventories are well correlated with the annual precipitations. However, this is the first time that a study investigates such a large panel of annual precipitation and therefore of anthropogenic radionuclide deposition. It seems that at high-altitude sites, deposition of artificial radionuclides was higher possibly due to orographic precipitations.

1. Introduction

Artificial radionuclides in soils of Western Europe are mainly due to global fallout from Nuclear Weapon Tests (NWTs) in the late fifties and sixties, to the American SNAP 9A satellite explosion in 1964, and to deposition from Chernobyl accident in May 1986. To discriminate and evaluate separately these three causes of soil contamination, different strategies were considered in the past:

- measurements of soils before and after 1986 (e.g. Aoyama et al., 2006),
- measurements of radionuclide or radionuclide ratio specific of one event (e.g. Mitchell et al., 1990),
- use of rain-deposition map considering that annual precipitations have driven the deposition of artificial radionuclides due to global fallout and that precipitations at the beginning of May 1986 have driven deposition of artificial radionuclides after the Chernobyl accident (e.g. Almgren et al., 2006).

In France, no reliable maps for artificial radionuclides in soils before 1986 were available and therefore Renaud et al. (2003) used annual precipitation map and map of precipitation of 1–5 May 1986 to estimate respectively global fallout deposition and Chernobyl deposition. To estimate global fallout, Renaud et al. used the relationship established in Ireland (Mitchell et al., 1990), which is based on $^{134}Cs/^{137}Cs$ ratio considering that there was no ^{134}Cs in western Europe coming from Nuclear Weapon Test (NWT). It appeared that some French areas were theoretically unaffected by wet deposition of artificial radionuclides from Chernobyl accident.

We investigate soil inventories of artificial radionuclides in two of these areas: Savoie and Montagne Noire. In addition, these 2 areas are mountain ranges and give us information on how the deposition of artificial radionuclides was in altitude sites. Finally, the contamination of the Montagne Noire was already investigated by a non-profit organisation (CRIIRAD, 2003) and it was shown that the contamination was quite high in the soil up to 1800 Bq kg^{-1} for ^{137}Cs.

The aims of this research study were 1) to validate rain-deposition estimation of global fallout and check if there were also

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[1] Dedicated to the memory of Françoise Vray.

artificial radionuclides from Chernobyl accident; 2) to investigate into details global fallout in mountain areas where deposition of atmospheric pollutants (POP, heavy metals) is known to be higher due to occult and orographic deposition (Graustein and Turekian, 1989; Roe, 2005; Daly and Wania, 2005; Le Roux et al., 2008).

2. Methods

2.1. Site description

Soils were sampled between 2004 and 2006 along transects of increasing altitude and subsequently increasing annual precipitation rate. Annual precipitation rates were given at each sampling site by data from Météo-France following AURELHY method (Bénichou and Le Breton, 1986, 1987).

Based on the map established by Renaud et al. (2003, 2004, 2005) and Roussel-Debet et al. (2007), areas where no precipitation occurred in the first week of May 1986 were chosen: Savoie and Montagne Noire (Fig. 1; Table 1). These mountains have different landscape and climate features. The Montagne Noire is a small mountain massif (Pic de Nore – $z_{max} = 1210$ m) in the Southern Part of Massif Central influenced both by Atlantic and Mediterranean air masses under a Mediterranean climate. Savoie (Tarentaise and Beaufortin) is a region part of the Western Alps and is characterised by a high mountain climate bordered by le Mont Blanc Massif ($z_{max} = 4810$ m) in the North and Vanoise Massif in the South.

2.2. Soil sampling and preparation

Sampling sites were located in undisturbed areas, i.e. non-ploughed grassland or woodland, that were assumed flat enough (slope < 5%) to prevent soil migration and heavy run-off erosion. In each site, undisturbed soil samples were collected using an 8 cm diameter stainless steel corer. Three cores (where possible) were collected together at each sampling site in order to minimize heterogeneity. The depth of each core was at least 30 cm. Some cores were sub-sampled (5 or 10 cm) in order to investigate the vertical distribution of the radionuclides. Sub-sections for the same depth interval of the three cores were then bulked and each composite interval was prepared separately. Soil profiles exhibiting very low anomalous inventory were discarded from data set.

Soil samples were homogenised, dried at 80 °C and sieved to remove coarse particles (greater than 2 mm) prior to spectrometry analyses. Soil samples were discarded when the amount of coarse material exceeds 30% of total material.

In addition, a peat core was taken in a minerotrophic peatland at the little Saint-Bernard Pass with a plexiglas tube of 1 m long and 9.4 cm diameter. The core was frozen, transported to the Institute of Environmental Geochemistry, Heidelberg, Germany and was prepared according to the protocol of Givelet et al. (2004). The first 55 cm powder samples were analysed by Gamma spectrometry like soil samples.

2.3. Gamma spectrometry

Direct gamma spectrometry analyses were performed on closed volumes of 60 ml of soil using γ-spectrometers with low background level HPGe-detectors with a 0.5 mm thickness beryllium window at the "Laboratoire de Mesure de la Radioactivité dans l'Environnement" (IRSN Orsay, France) (Bouisset and Calmet, 1997). Samples were measured for 24–48 h. Detectors were located underground, under a 3 m slab of concrete, in a room shielded with 10 cm low activity lead and 5 mm electrolytic copper. Efficiency calibrations were obtained using different densities of pitchblende sources prepared in the same geometry as the samples. The calibration energy range was 46 keV to 2.7 MeV. ^{137}Cs activity (half-life: 30.2 years) was determined based on its peak at 661.7 keV. For peat samples, ^{241}Am, ^{214}Pb and ^{210}Pb were measured respectively based on their peaks at 59.4; 351.9 and 46.5 keV. ^{214}Pb was used to estimate supported ^{210}Pb (Appleby, 2001). Self-adsorption corrections were done according to sample type, density and gamma energy using in-house standards.

2.4. $^{239+240}$Pu and ^{241}Am determination

Pu isotopes and ^{241}Am were measured at the "Laboratoire de Mesure de la Radioactivité dans l'Environnement" (IRSN Orsay, France), by alpha-counting

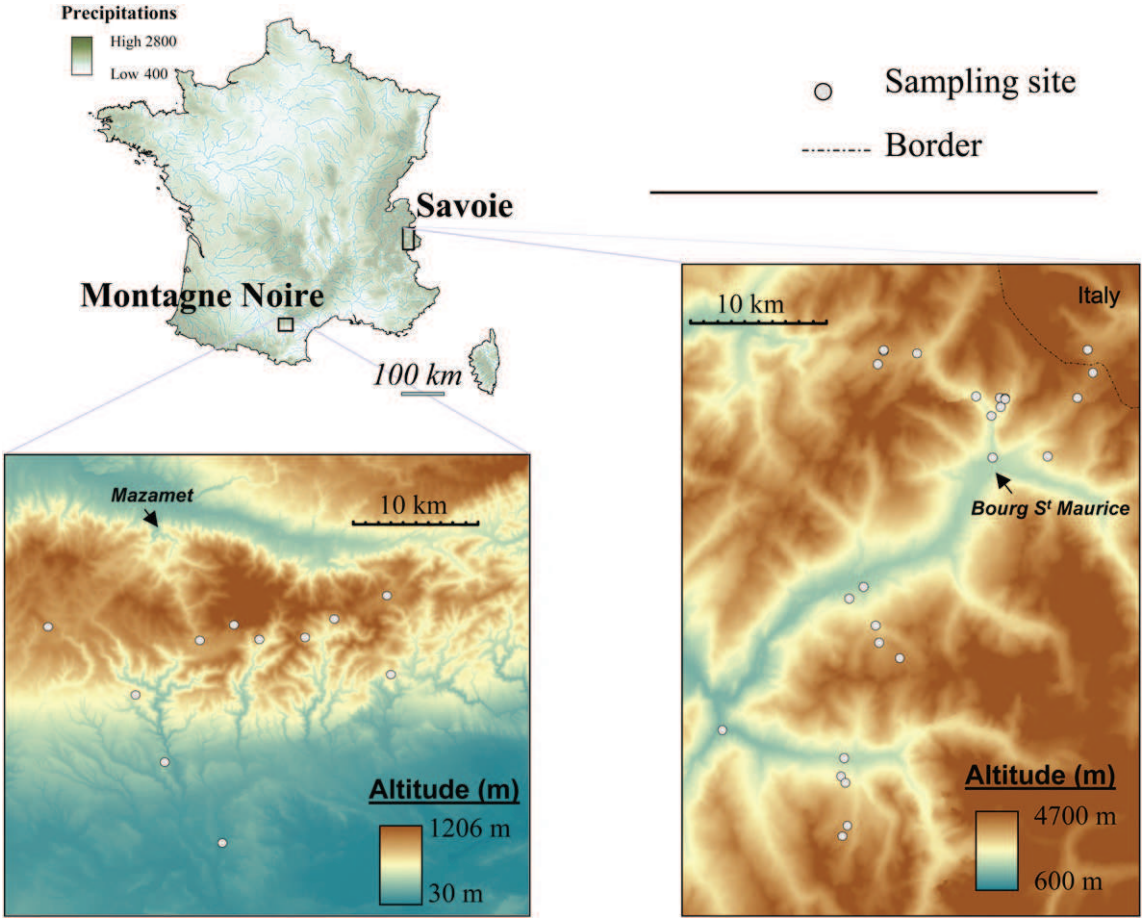

Fig. 1. Location of the sampling zones in France with respect to mean annual precipitations inferred from AURELHY Météo-France data and altitude.

after radiochemical treatment, using a protocol of Goutelard et al. (1998). Briefly, samples are first ashed at 480 °C in order to eliminate organic matter and then spiked with ^{242}Pu and ^{243}Am, to control the recovery efficiency of the treatment. A double-step leaching using concentrated HNO_3 and H_2O_2 makes the radionuclides soluble. Pu and Am are then separated from stable elements and natural alpha-emitting radioisotopes by co-precipitation exchange chromatography and extraction chromatography (Goutelard et al., 1998). The 2 fractions (respectively Pu and Am) are then electro-deposited and alpha-counted for seven days.

2.5. Radionuclide inventories

Radionuclide inventories were calculated as follows:

$$I = \sum_i A_i \times \delta_i \times z_i \quad \text{(SI units)} \tag{1}$$

where I is the total soil inventory of the measured radionuclide, A_i the activity for the radionuclide in each layer I of the <2 mm dry soil fraction, δ_i the soil density for each layer I and z_i the depth interval for each layer I,

3. Results and discussion

3.1. Artificial radionuclide distribution

There is a large scale of variation in ^{137}Cs, $^{239+240}$Pu and ^{241}Am inventories, for Savoie respectively 1500–8700 Bq m^{-2}, 50–350 Bq m^{-2} and 20–150 Bq m^{-2} and for Montagne Noire respectively 2300–6900 Bq m^{-2}, 76–190 Bq m^{-2} and 30–73 Bq m^{-2}.

We show that more than 60% of these radionuclides are preserved in the first 20 cm of the soils, ascertaining no major loss by vertical migration [ESI 1] and have the same behaviours in the soil [Fig. 2]. Because we collected soil up to at least 30 cm depth, we think that we get a complete recovery of the radionuclides deposited onto the soils. In addition, the similar distribution of the three elements (Cs, Am and Pu) [for Pu and Cs relationship see Fig. 2, for Am and Pu relationship see ESI 2] indicates principally a physical, mechanical migration (i.e. bioturbation, infiltration by soil cracks…) and not a chemical migration since these elements have different chemical behaviours.

3.2. Origin of the artificial radionuclides

It is now well-established that ^{137}Cs from Chernobyl accident was deposited on soils mainly during raining events at the beginning of May 1986 (Almgren et al., 2006; Mitchell et al., 1990; Renaud et al., 2003). In Montagne Noire and Savoie, no precipitation occurred between 1 and 5th May 1986 and our hypothesis was that the artificial radionuclides were mainly coming from Nuclear Weapon Tests (NWTs) global fallout. This is confirmed by the mean ratio $^{239-240}$Pu/^{137}Cs of 0.034 ± 0.006 (Savoie samples) [Fig. 3] compared to the ratios measured by Mitchell et al. (1990), Hodge et al. (1996) and Bunzl and Kracke (1988) between 0.03 and 0.033. This is also confirmed by the constant values of $^{239-240}$Pu/^{137}Cs with increasing ^{137}Cs values [Fig. 3]. Indeed any additional input of ^{137}Cs coming from Chernobyl will decrease the $^{239-240}$Pu/^{137}Cs ratio in soils since there was negligible plutonium deposition in Western Europe after Chernobyl. Concerning ^{137}Cs, the ratio to plutonium is thus constant and typical of global fallout taking into account radioactive decay until 2007.

Plutonium isotopes [Fig. 4] have a typical isotopic ratio representative of global nuclear fallout (^{238}Pu/$^{239+240}$Pu = 0.0284 ± 0.005 for Savoie samples) which is very closed to the value of 0.03 according to Hardy et al. (1973) and soil values of 0.034 ± 0.013 from Italy measured by Jia et al. (1999). ^{241}Am/$^{239+240}$Pu ratio displays also relative uniform value (0.41 ± 0.06 for Savoie samples) with 2 outliers from nearby sites displaying one higher value and one lower. These results are also in good agreement with soil results from South-West France (IRSN, unpublished results) [Fig. 4] and Switzerland (Eikenberg et al., 2004; Froidevaux et al., 2004).

3.3. Relationship between soil inventories of artificial radionuclides and annual precipitations

AURELHY ("Analyse Utilisant le RELief pour l'Hydrométéorologie") is a method of precipitation estimation based on numerous meteorological stations from Meteo France and measurements over thirty years. AURELHY method is also based on the topographic features of terrain and therefore precipitations are well correlated with altitudes for a same mountain range (Bénichou and Le Breton, 1986, 1987). The resulting map is a grid data layer at 1-km resolution with a monthly average for each point. We use it to estimate annual precipitations.

The relationships between mean annual precipitation (MAP) and soil inventories of radionuclides from Nuclear Weapon Tests in undisturbed locations are well-established (e.g. Pourcelot et al., 2007; Sanchez-Cabeza et al., 2007, for a summary see table 2 in Sutherland, 1996). Our rain-deposition relationships for plutonium [Fig. 5a] and for ^{137}Cs [Fig. 5b] in Savoie and Montagne Noire agree well with previous relationships established in neighbouring countries in Ireland by Mitchell et al. (1990) and Germany by Bunzl and Kracke (1988). At both sites, plutonium and ^{137}Cs total inventory are correlated with annual precipitations. For ^{137}Cs, where the data set is more complete than for plutonium, the general relationship between ^{137}Cs total inventory (I_{cesium}) in Bq m^{-2} and MAP in mm is:

$$I_{cesium} = 3.04 \times MAP - 110 \quad \left(n = 51; r^2 = 0.415, P_{0.05} < 0.01\right) \tag{2}$$

In order to see if there was a difference between the 2 areas, I/MAP_{Savoie} and $I/MAP_{M\text{-}Noire}$ ratios were calculated. Median latencies in ratio for $I/MAP_{M\text{-}Noire}$ and, I/MAP_{Savoie} are respectively 2.91 and 2.59 Bq m^{-2} mm^{-1}; the distributions in the two groups of I/MAP ratios are slightly different (Mann–Whitney test $U = 382$, $n_{Mt\_Noire} = 21$, $n_{Savoie} = 27$, $P = 0.041 < 0.05$, two-tailed). Indeed excluding samples from one site in Savoie like Cormet de Roselend for example will make the relationships for the 2 areas statistically identical (Mann–Whitney test $U = 150$, $n_{Mt\_Noire} = 21$, $n_{Savoie} = 21$, $P = 0.08 > 0.05$, two-tailed).

The slight differences between the two locations can be perhaps explained by the difference in altitude range and topographic features of the 2 areas. For example, high-altitude soils either in Montagne Noire (around 1400 mm y^{-1} precipitations corresponding to the altitude of around 1000 m) or in Savoie (2 sites: 1850 mm y^{-1}, 8696 Bq m^{-2}; 1808 mm y^{-1}, 7039 Bq m^{-2}) have larger inventories than predicted by "classical annual precipitation–deposition relationship" [Fig. 5b, ESI 3]. This can be due to snow accumulation (Pourcelot et al., 2003) or atmospheric processes such as orographic deposition (Le Roux et al., 2008), which are not well taken into account in the AURELHY method to spatialize annual precipitations. These different hypotheses should be further investigated, for example in Pyrenees mountain range.

3.4. Chronology of radionuclide deposition and comparison with other records

The age dating of the peat core was done using the ^{210}Pb Constant Rate of Supply (CRS) model (Appleby, 2001). The peak of

Table 1
Sample geographic coordinates, radionuclide inventories and ratios.

Mountainous area	Location	Site number	Sample type	Latitude	Longitude	Altitude (m)	Mean annual precipitation rate (mm y^{-1})	^{137}Cs inventory (Bq m^{-2})	$\Delta^{137}Cs$ inventory (Bq m^{-2})	^{241}Am inventory (Bq m^{-2})
Montagne Noire	Pic de Nore	PICNO2004-1	woodland	43.4185	2.4663	1112	1500	6348	292	73
	Castan	CASTA2004-2	woodland	43.4063	2.493	635	1480	3582	106	
	Pradelles-Cabardès	PRACA2004-2	woodland	43.4059	2.4324	814	1350	4411	259	
	Les Martys	MARTY2004-4	woodland	43.4172	2.2851	793	1390	5867	238	
	Mas-Cabardès	MASCA2004-5	woodland	43.3665	2.3685	357	920	2343	145	28
	Limousis	LIMOU2004-6	woodland	43.3187	2.3987	240	738	2555	108	30
	Lespinassière	SPINA2004-7	woodland	43.4077	2.5373	610	1390	3848	261	45
	Col de Salette	COLSA2004-8	woodland	43.422	2.5668	920	1420	6865	398	
	Col de Salette	COLSA2004-9	grassland	43.422	2.5668	920	1420	4646	170	
	Cassagnoles	CASSA2004-11	woodland	43.3829	2.621	485	1200	3775	203	
	Cassagnoles	CASSA2004-12	woodland	43.3829	2.621	690	1200	3448	125	
	Cassagnoles	CASSA2004-13	grassland	43.3829	2.621	690	1220	2951	119	
	Cassagnoles	CASSA2004-14	grassland	43.3829	2.621	526	1080	3144	207	
	Cassagnoles	CASSA2004-15	grassland	43.3829	2.621	382	1200	3309	235	
	Cassagnoles	CASSA2005-1	woodland	43.3829	2.621	524	1200	2361	163	
	Pic de Nore	PICNO2005-2	grassland	43.4185	2.4663	1180	1500	5889	507	
	Le Fournas, Ferrals-Les-Montagnes	FOURN2005-4	grassland	43.4375	2.6175	763	1380	3839	306	
	Le Fournas, Ferrals-Les-Montagnes	FOURN2005-7	woodland	43.4375	2.6175	761	1380	4812	386	
	Pradelles-Cabardès	PRACA2005-8	grassland	43.4059	2.4324	718	1350	5676	308	
	Les Martys	MARTY2005-11	grassland	43.4172	2.2851	794	1390	3752	267	
	Mas-Cabardès	MASCA2005-A12	grassland	43.3665	2.373	299	920	2352	233	
Savoie	Courchevel	COURC2006-2	grassland	45.39387	6.6327	2060	1368	2291	272	37
	Courchevel	COURC2006-3	grassland	45.40036	6.63788	1913	1284	3190	384	45
	Saint Bon	STBON2006-4	grassland	45.431715	6.634648	1199	945	1619	269	21
	Saint Bon	STBON2006-5	grassland	45.427347	6.638135	1401	980	3386	398	34
	Saint Bon	STBON2006-6	grassland	45.427347	6.638135	1401	980	2970	358	58
	Versoye	VERSO2006-7	grassland	45.664478	6.796135	1698	1550	4014	535	43
	Versoye	VERSO2006-8	grassland	45.663722	6.800828	1557	1537	3434	479	44
	Versoye	VERSO2006-9	grassland	45.663612	6.80061	1547	1535	4877	597	54
	Versoye	VERSO2006-10	grassland	45.658563	6.79606	1392	1459	3411	408	42
	Bonneval	BONNL2006-11	grassland	45.653383	6.787328	1080	1340	3228	342	48
	La Plagne	PLAGN2006-12	grassland	45.50395	6.69279	2145	1368	3256	316	46
	La Plagne	PLAGN2006-14	grassland	45.50395	6.69279	2145	1368	3300	417	41
	La Plagne	PLAGC2006-15	grassland	45.514468	6.675112	1817	1299	2967	368	43
	La Plagne	PLAGC2006-17	grassland	45.52547	6.67322	1521	1122	2808	350	38
	Saint Bon	STBON2006-18	grassland	45.44318	6.638212	820	908	3141	379	43
	Villarlurin	VILLR2006-19	grassland	45.46491	6.530807	559	906	3066	332	42
	Macôt la Plagne	BONNG2006-20	grassland	45.543362	6.650897	1030	912	3042	363	45
	Macôt la Plagne	BONNG2006-21	grassland	45.550247	6.664133	725	777	2247	331	34
	Col du Petit St Bernard	ROS2006-22	grassland	45.67702	6.8808	2175	1808	7039	670	71
	Col du Petit St Bernard	ROS2006-24	grassland	45.6616	6.86561	2006	1648	5205	519	77
	Cormelet de Roselend	ROS2006-25	grassland	45.69171	6.8771	1973	1850	8686	930	144
	Cormelet de Roselend	ROS2006-26	grassland	45.689886	6.68793	1962	1922	3910	404	62
	Cormelet de Roselend	ROS2006-27	grassland	45.69829	6.69401	1996	1905	4627	527	70
	Cormelet de Roselend	ROS2006-29	grassland	45.69554	6.72371	1721	1896	3768	417	54
	Cormelet de Roselend	ROS2006-30	grassland	45.66628	6.77428	1200	1644	4385	478	63
	Cormelet de Roselend	ROS2006-31	grassland	45.62704	6.78608	886	997	2580	338	62
	La Rosière	ROS2006-32	grassland	45.62587	6.83579	1564	1037	3641	446	44
	Col du Petit St Bernard	PSB-II	peatland	45.680287	6.883954	2050	1922	2910	200	82[a]

[a] Measured by gamma spectrometry.

Δ^{241}Am inventory (Bq m^{-2})	^{238}Pu inventory (Bq m^{-2})	Δ^{238}Pu inventory (Bq m^{-2})	$^{239+240}$Pu inventory (Bq m^{-2})	$\Delta^{239+240}$Pu inventory (Bq m^{-2})	$^{239+240}$Pu/^{137}Cs	$\Delta^{239+240}$Pu/^{137}Cs	^{238}Pu/$^{239+240}$Pu	Δ^{238}Pu/$^{239+240}$Pu	^{241}Am/$^{239+240}$Pu	Δ^{241}Am/$^{239+240}$Pu
7	5.5	0.5	190	8	0.030	0.003	0.029	0.004	0.39	0.05
3	2.0	0.3	76	4	0.032	0.004	0.027	0.005	0.37	0.06
3	2.2	0.4	82	4	0.032	0.003	0.027	0.006	0.37	0.06
5	3.0	0.4	111	4	0.029	0.003	0.027	0.004	0.41	0.06
4	2.3	0.4	89	4	0.039	0.006	0.026	0.005	0.41	0.07
5	3.1	0.5	111	5	0.035	0.006	0.028	0.006	0.40	0.06
3	1.5	0.3	49	2	0.030	0.006	0.031	0.007	0.42	0.07
4	3.3	0.5	121	5	0.036	0.006	0.028	0.006	0.28	0.05
6	2.8	0.5	100	4	0.034	0.006	0.028	0.006	0.58	0.08
6	3.2	0.5	112	5	0.028	0.005	0.028	0.006	0.38	0.07
8	3.0	0.8	107	6	0.031	0.006	0.028	0.009	0.41	0.10
7	4.4	0.7	132	6	0.027	0.004	0.033	0.006	0.41	0.07
5	3.4	0.7	106	6	0.031	0.005	0.032	0.008	0.40	0.07
5	3.7	0.6	113	5	0.035	0.005	0.033	0.007	0.42	0.06
4	2.8	0.3	107	4	0.033	0.004	0.026	0.004	0.43	0.06
5	2.5	0.4	100	4	0.030	0.005	0.025	0.005	0.41	0.06
5	2.9	0.4	103	4	0.035	0.006	0.029	0.005	0.42	0.07
6	2.6	0.4	94	4	0.034	0.006	0.028	0.006	0.41	0.08
6	3.2	0.4	106	4	0.034	0.005	0.030	0.005	0.40	0.07
5	2.9	0.4	107	4	0.035	0.005	0.027	0.005	0.40	0.06
5	2.8	0.5	104	4	0.034	0.006	0.027	0.006	0.43	0.07
4	2.6	0.4	81	4	0.036	0.007	0.032	0.007	0.41	0.07
7	5.2	0.4	183	7	0.026	0.003	0.029	0.003	0.39	0.05
7	5.1	0.5	194	7	0.037	0.005	0.026	0.003	0.40	0.05
12	8.6	0.6	350	11	0.040	0.006	0.025	0.003	0.41	0.05
7	4.2	0.5	146	5	0.037	0.005	0.029	0.005	0.43	0.06
7	4.7	0.5	164	6	0.035	0.005	0.029	0.004	0.43	0.06
6	3.3	0.5	124	5	0.033	0.005	0.026	0.005	0.43	0.06
6	4.6	0.7	151	6	0.035	0.005	0.030	0.006	0.42	0.06
7	4.9	1.0	154	8	0.060	0.011	0.032	0.008	0.40	0.06
5	3.2	0.4	111	4	0.031	0.005	0.029	0.005	0.39	0.06
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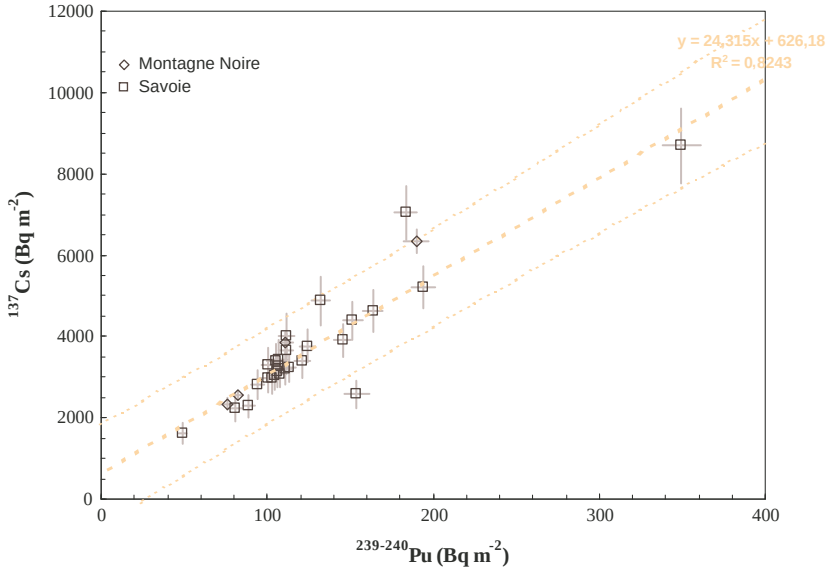

Fig. 2. Relationship between ^{137}Cs and $^{239+240}$Pu total soil inventories in Savoie (squares) and Montagne Noire (diamonds).

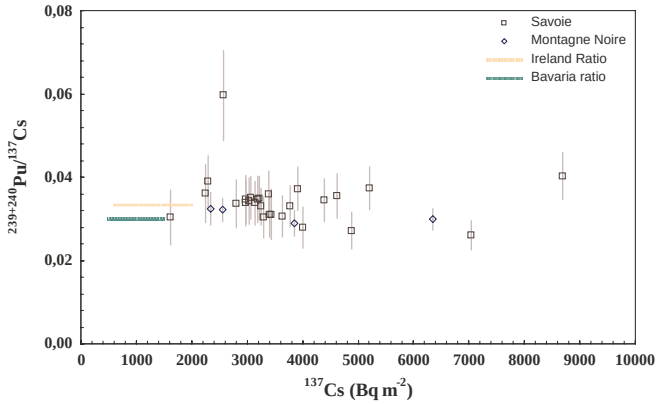

Fig. 3. $^{239+240}$Pu/^{137}Cs ratio vs. ^{137}Cs in total soil inventories in Montagne Noire (diamonds) and Savoie (squares).

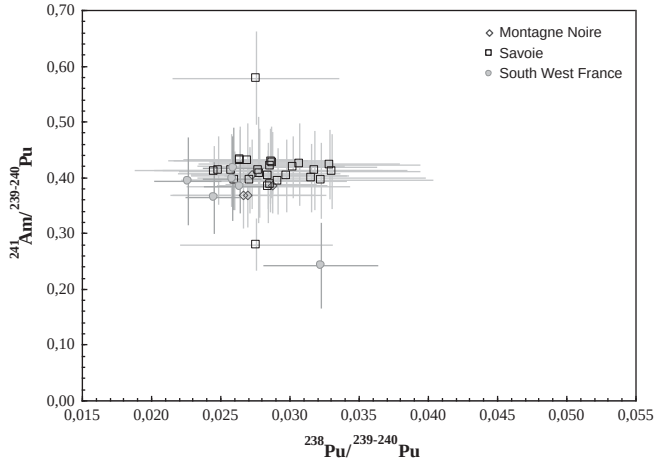

Fig. 4. ^{241}Am/$^{239+240}$Pu vs. ^{238}Pu/$^{239+240}$Pu in total soil inventories in Montagne Noire (diamonds) and Savoie (squares). Are also shown 6 samples from South-West France (full circles – unpublished data IRSN).

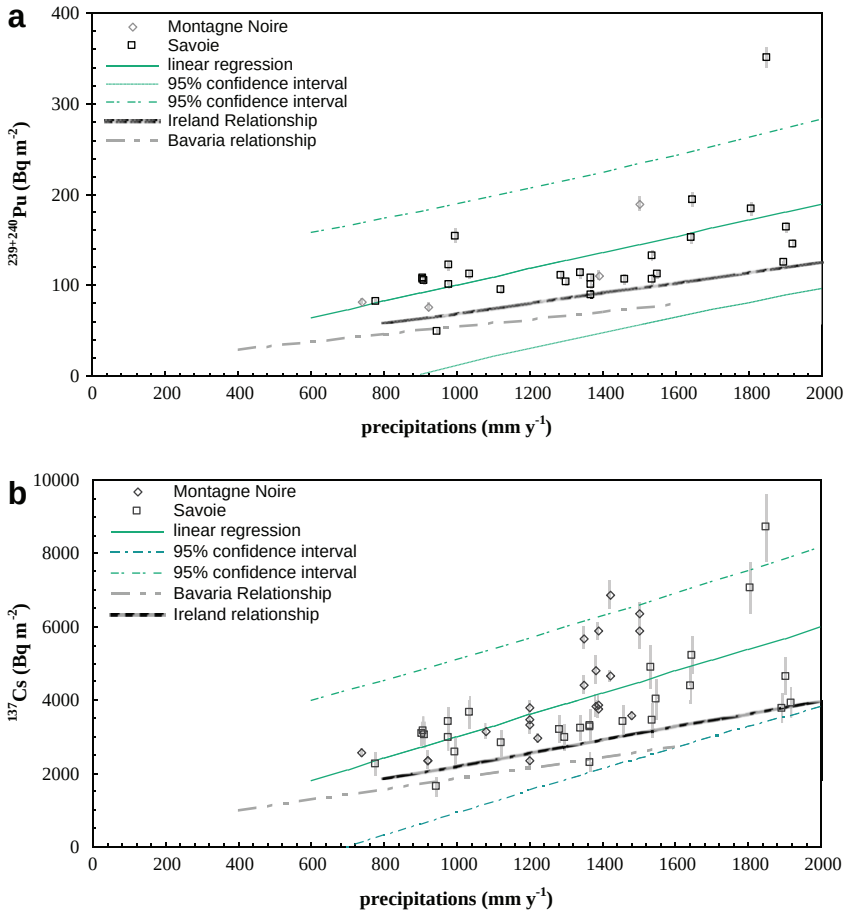

Fig. 5. a. relationship between ^{137}Cs total soil inventories and annual precipitations in Savoie (squares), in Montagne Noire (diamonds). b. relationship between $^{239+240}$Pu total soil inventories and annual precipitations in Savoie (squares), in Montagne Noire (diamonds).

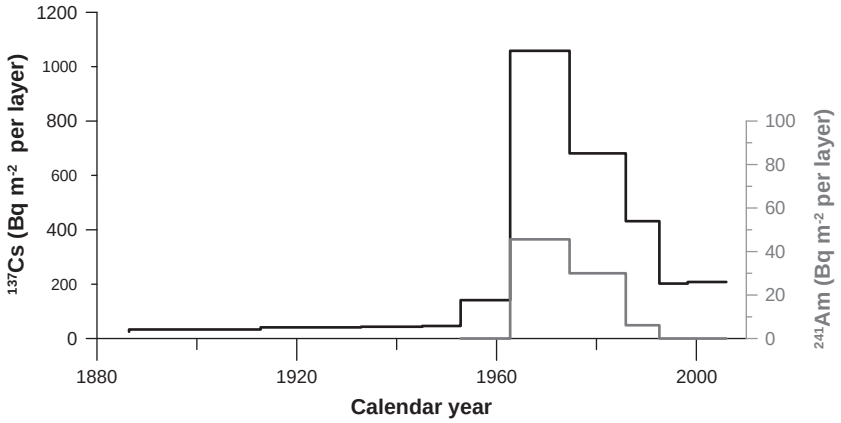

Fig. 6. ^{137}Cs and ^{241}Am distribution (Bq m^{-2} per layer) vs. calendar age estimated by ^{210}Pb CRS model.

^{137}Cs and ^{241}Am is dated between 1963 and 1975 [Fig. 6], which is in good agreement with records in western Europe (Monna et al., 2009 – Fig. 1). ^{137}Cs is subject of mobilization in the peat core because of similar behaviour than K^+ and plant retention (Turetsky et al., 2004; Rausch et al., 2005), however ^{241}Am, daughter of ^{241}Pu is relatively immobile with only three peat layers where this radionuclide is measurable. Mobility of ^{137}Cs in the peat core can perhaps explain the low ^{137}Cs inventory (2910 Bq m^{-2}) in comparison with nearby soils (5200–7000 Bq m^{-2}). ^{241}Am soil inventory, as measured by gamma spectrometry, is 82 Bq m^{-2}. This is in good agreement with nearby soil inventories for ^{241}Am (71–77 Bq m^{-2}).

4. Conclusions

Our results on deposition of artificial radionuclides in two mountainous areas not impacted by Chernobyl-derived radionuclides, show that deposition over a large spectrum of precipitation

rate is mainly explained by mean annual precipitation in good agreement with previous European studies. In high mountainous sites, radionuclide soil inventory in soils can be higher than predicted due to snow accumulation and orographic precipitations. Isotopic ratio of ^{137}Cs, ^{241}Am and plutonium isotopes confirm their Global Fallout origin. ^{241}Am and ^{137}Cs peaks found in peat layer age-dated from the end of the sixties to beginning of the seventies confirm that radionuclides were deposited following the Nuclear Weapon tests.

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Appendix. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jenvrad.2009.10.010.

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