



Activity ratios of ^{137}Cs , ^{90}Sr and $^{239+240}\text{Pu}$ in environmental samples

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Abstract

Both global and Chernobyl fallout have resulted in environmental contamination with radionuclides such as ^{137}Cs , ^{90}Sr and $^{239+240}\text{Pu}$. In environmental samples, ^{137}Cs and $^{239+240}\text{Pu}$ can be divided into the contributions of either source, if also the isotopes ^{134}Cs and ^{238}Pu are measurable, based on the known isotopic ratios in global and Chernobyl fallout. No analogous method is available for ^{90}Sr . The activity ratios of Sr to Cs and Pu, respectively, are known for the actual fallout mainly from air filter measurements; but due to the high mobility of Sr in the environment, compared to Cs and Pu, these ratios generally do not hold for the inventory many years after deposition.

In this paper we suggest a method to identify the mean contributions of global and Chernobyl fallout to total Sr in soil, sediment and cryoconite samples from Alpine and pre-Alpine regions of Austria, based on a statistical evaluation of Sr/Cs/Pu radionuclide activity ratios. Results are given for Sr:Cs, Sr:Pu and Cs:Pu ratios. Comparison with fallout data shows a strong depletion of Sr against Cs and Pu.

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1. Introduction

The most prominent long-lived radionuclide in Chernobyl fallout is ^{137}Cs , because of its predominant contribution to long-term dose, due to both its long physical half-life (30 years) and long ecological half-life (several years) in most environmental media. However, there are other radionuclides worth investigating, even if they have a rather negligible dosimetric significance, at least in Central Europe (the situation is different in affected areas of Southern Belarus, Northern Ukraine and the Bryansk region of Russia). It is important to study radionuclides such as ^{90}Sr , Pu isotopes, ^{241}Am and others because: (1) we need to establish a status quo of environmental contamination (this is one of the tasks of “classical” radio-ecology); (2) they can be used as relatively easily measurable tracers for other (chemically similar) pollutants whose likely behaviour in the environment can thus be assessed; and (3) this gives insight into the functioning of environmental transport, dissipation and speciation mechanisms, which can help us to understand how ecosystems function altogether.

The ^{137}Cs , ^{90}Sr and Pu radionuclides, which are the subject-matter of this paper, are of particular interest because they were injected into the environment twice: first, following atmospheric atomic bomb tests and the crash of a ^{238}Pu -powered US-satellite in the late 1950s and early 1960s, and, later, in 1986 by the Chernobyl accident. We may therefore get some information about the temporal aspect of their long-term behaviour in the environment if we succeed in separating the respective contributions of these two input events.

The radionuclide ratios in the Chernobyl clouds were not spatially and temporally constant. Reasons include the release history (variable temperature, different zones of the reactor affected), the physical–chemical nature of released matter (gaseous, solid particles, condensed particles) and the atmospheric transport conditions. Even the ^{134}Cs : ^{137}Cs ratio is not exactly constant in fallout, not even over the relatively small area of Austria, although these nuclides have identical chemical and transport properties. Therefore, the ratios of ^{90}Sr and Pu isotopes to ^{137}Cs cannot be expected to be constant over a region such as Austria, which has been affected by several passing Chernobyl clouds, corresponding to different phases of the release and transport history. Also, the activity ratios in the surface deposition are not necessarily equal to the time-integrated ratio in the cloud, because the deposition rate from the cloud was not constant during its passage, but depended strongly on local precipitation history at that time. Therefore, the fallout composition found in soil and other media may reflect the composition of the cloud at one time, i.e. when it was raining in that area, rather than the mean over time. However, as reference values for fallout we have no choice but to use actual fallout measurements (not available in our case) or aerosol measurements.

The location closest to our investigation area where aerosols have been sampled during the passage of the Chernobyl cloud is the city of Salzburg, where an automatic sampling station is located (AT1216, 13.053°E, 47.791°N; <http://eurdep.jrc.it/>). The ^{90}Sr and $^{239+240}\text{Pu}$: ^{137}Cs ratios determined in the Chernobyl cloud at that location are given in Table 1 (Irlweck and Khademi, 1993). The very low Pu:Cs ratio in Chernobyl fallout seems to be typical for locations far from the source. Pu has been transported as particles, most of which were deposited in the near zone; therefore, the Pu:Cs ratio is much higher near Chernobyl (6 km south of the NPP, a ratio 0.0088 has been found, Bossew et al., 2004). To a lesser degree, the same applies to the Sr:Cs ratio (0.30 at the same site).

The situation is different for global fallout, which has been deposited rather evenly over many years, and which represents a “mixture” of many release events, the intense atmospheric atomic bomb tests from the late 1950s until 1963. Due to a relatively long residence time in the

Table 1
Ratios of activity concentrations in global and Chernobyl fallout

	Global	Chernobyl
$^{90}\text{Sr}:^{137}\text{Cs}$	0.641 ^a	0.017 ^b
$^{239/240}\text{Pu}:^{137}\text{Cs}$	0.0180 ± 0.0024^c	$6.6\text{E}-6^b$
$^{90}\text{Sr}:^{239/240}\text{Pu}^d$	36	2600

^a UNSCEAR (2000); calculated from the figures in annex C, table 10, p. 214 f, as the ratio of the sum of the decay corrected, quoted annual depositions of ^{90}Sr and ^{137}Cs in the Northern hemisphere. Decay corrected to 2000.

^b For Salzburg; determined from time-integrated aerosol measurements (Irlweck and Khademi, 1993).

^c Bunzl and Kracke (1988).

^d Calculated from the rows above.

high atmosphere and the stratosphere, the residues have reached the earth's surface until several years later. The spatial variability of the fallout level is correlated to average precipitation intensity, and the spatial variability of the fallout compositions seems to be negligible over Central Europe, at least.

In this investigation we attempt to estimate activity ratios of $^{90}\text{Sr}:^{137}\text{Cs}$, $^{90}\text{Sr}:^{239+240}\text{Pu}$ and $^{239+240}\text{Pu}:^{137}\text{Cs}$ for the contributions of global and Chernobyl fallout in soil, lake sediment and cryoconite samples from the province of Salzburg, Austria. Motivated by the existence of particularly vulnerable ecosystems (Alpine agriculture) and regionally high, and spatially very variable, fallout levels, radio-ecological investigations have been performed for many years in this area. The values obtained here will be compared with the reference values as discussed in Table 1.

2. Materials and methods

2.1. Investigation regions

The samples were taken as part of various radio-ecological survey projects carried out between 1996 and 2004 in Alpine and pre-Alpine regions of the province of Salzburg. Cryoconite (sediments which can be found on glacier surfaces, known for their high capacity for accumulating radionuclides) and some of the soil samples come from the high Alpine Hohe Tauern region (part of the Central Alpine range), while some other soil samples stem from Alpine pastures in valleys and mountain slopes below. The investigated sediments were taken in the pre-Alpine Lake Wallersee.

The regions are known for, in some areas, high Chernobyl fallout (even exceeding 150 kBq/m^2 ^{137}Cs), due to heavy precipitation around 1 May 1986. The Northern slope of the Alps is also subject to heavy annual precipitation (up to more than 2000 mm/year), which resulted in high global fallout inventories.

2.2. Sampling and sample processing

Soil samples were taken as cores with soil coring devices or as cubes with spades. They were sliced into horizontal layers in order to investigate the vertical distributions of the radionuclides. The samples were dried (100–110 °C), large stones removed, and the remaining samples (soil, vegetation fibres, small stones) were homogenized as much as possible and subjected to radiometry.

Cryoconites were taken by grab sampling from the glacier surface. In the laboratory they were separated from snow and ice, and dried. Apparent dirt particles (larger fibres) were removed, but the samples were generally very fine-grained and sufficiently homogenous that no further treatment was required.

Sediment samples were collected with a gravity corer with sampling core lengths from 17.0 to 27.5 cm depending on the penetration resistance of the sediment. After samples had been collected, still in wet

conditions, every core was sliced into 5 mm layers. Samples were dried at 105 °C for gamma spectrometry, and aliquots of four layers were pooled for Pu and Sr analysis.

The samples discussed in this paper were taken between 1996 and 2004.

2.3. Radiometry

2.3.1. Gamma spectrometry – ^{137}Cs and ^{134}Cs

Some measurements were made with the HPGe detector from Canberra by the Institute of Applied Ecology and Institute of Risk Research at the University of Vienna, while others with the HPGe detectors from ORTEC by the Institute of Physics and Biophysics at the University of Salzburg. Coincidence (summation) correction was performed for ^{134}Cs (Salzburg: empirically; Vienna: MC calculation). This is important in order to separate ^{137}Cs into its fractions stemming from global fallout and Chernobyl, respectively, based on the relatively well-known ratio $^{134}\text{Cs}:^{137}\text{Cs}$ in Chernobyl fallout (see Section 2.4).

2.3.2. Plutonium and strontium analysis

Between 1.5 and 2.6 g of each sample were placed in a muffle furnace to be ashed at a temperature of 500 °C for at least 24 h. After cooling, 130 mBq ^{242}Pu and 10 mg stable Sr were added to the cryoconite ash. The sample was put in a Teflon beaker and evaporated to dryness with repeated addition of 65% HNO_3 , and 32% HCl mixed with H_3BO_3 . The residue was converted to nitrate by evaporating three times with 10 ml 65% HNO_3 . Finally, the sample was dissolved in 70 ml of 1 M HNO_3 and filtered. All plutonium species were oxidized to Pu(IV). Dissolved in 8 M HNO_3 , the sample was loaded onto a preconditioned anion exchange resin column (AG 1-X8 resin, BioRad, CA, USA). The effluent was collected and stored in plastic containers for subsequent analysis of ^{90}Sr . The column was rinsed with 50 ml 8 M HNO_3 and subsequently with 100 ml 10 M HCl . Plutonium was stripped with 100 ml 9 M HCl containing 0.1 M NH_4I . The strip solution was evaporated. Finally Pu was oxidized to Pu(IV) and collected on a propylene filter after co-precipitation with NdF_3 (the radiochemical procedure is described in more detail by LaRosa et al. (1992)). The $^{239+240}\text{Pu}$ activity was determined with a partially depleted PIPS detector (Canberra, active area 900 mm², thickness 100 µm) with a counting efficiency of approximately 40%. The uncertainty of all $^{239+240}\text{Pu}$ measurements was estimated to be below 15% (1 SD).

Details for the analytical procedure for ^{90}Sr are given by Vajda et al. (1992): After oxalate precipitation – using the effluent of the Pu analysis – at pH 5–6, which removes the bulk of potassium and other alkali metals, the sample was dissolved in 3 M HNO_3 and loaded onto a chromatographic column filled with 2.5 g of a supported crown ether (Sr.Spec., EiChrom Industries, IL, USA). The column was rinsed with 110 ml 3 M HNO_3 and the Sr stripped with 20 ml distilled water. The chemical yield was determined by gravimetry after a strontium oxalate precipitation at pH 9–10. After ^{90}Y had reached equilibrium with ^{90}Sr (in practice after at least three weeks), the precipitate was dissolved in 2 ml 1.0 M HNO_3 , 4 ml scintillation cocktail (ReadyGel, Beckman, USA) was added and the sample was measured by liquid scintillation counting. For all samples the uncertainty of the measurement was about 15% (1 SD).

2.4. Isotopic ratios

^{137}Cs and $^{239+240}\text{Pu}$ are due to global and Chernobyl fallout. Taking advantage of the fact that ^{134}Cs is contained practically only in Chernobyl fallout (Cigna et al., 1971) and from the estimated $^{137}\text{Cs}:^{134}\text{Cs}$ isotopic ratio in Chernobyl fallout, $Q_{\text{Cs}} = 1.715$ for the Hohe Tauern region (reference date 1 May 1986; estimated from measurements of environmental media of the region with high ^{134}Cs concentrations, made not long after Chernobyl; to be published), we calculate the contribution of global- ^{137}Cs as follows:

$$^{137}\text{Cs}(\text{global}) = ^{137}\text{Cs}(\text{total}) - Q_{\text{Cs}} \times ^{134}\text{Cs}.$$

Similarly, the ratios of $^{238}\text{Pu}:$ $^{239+240}\text{Pu}$ are 0.461 ± 0.032 in Chernobyl and 0.035 in global fallout (Irlweck and Wicke, 1998; Perkins and Thomas, 1980). From measured activities of ^{238}Pu and $^{239+240}\text{Pu}$ the contributions of either source can be calculated. This method has also been used by other authors, e.g. Mietelski and Was (1995).

2.5. Assigning ^{90}Sr to global and Chernobyl fallout

The situation is different for ^{90}Sr , which stems both from global and Chernobyl fallout. Only one isotope is available, thus the mathematical separation in either source is not possible. Furthermore, the high solubility of Sr in water leads to its depletion from surface soil and cryoconite, to a much larger extent than for Cs and Pu, which are more strongly fixed, Cs in clay minerals (ubiquitous in soil and cryoconite), Pu also in organic matter depending on the chemical conditions (e.g., SCOPE 50, sec. 5.2.4.2; Smith and Amonette, 2006; sec. 3.2).

In order to assess the fractions of ^{90}Sr in global and Chernobyl fallout, respectively, we propose the following procedure.

Let us briefly denote the activities of ^{90}Sr (Chernobyl fallout), ^{90}Sr (global fallout), ^{137}Cs (Chernobyl), ^{137}Cs (global), $^{239+240}\text{Pu}$ (Chernobyl) and $^{239+240}\text{Pu}$ (global) as Sr(C), Sr(g), Cs(C), Cs(g), Pu(C) and Pu(g), and the total activities as Sr etc., without bracket, respectively.

Furthermore, let

$$q\text{SrCs(C)} := \text{Sr(C)}/\text{Cs(C)},$$

$$q\text{SrCs(g)} := \text{Sr(g)}/\text{Cs(g)},$$

and similarly for $q\text{SrPu(C)}$, $q\text{SrPu(g)}$, $q\text{PuCs(C)}$ and $q\text{PuCs(g)}$.

We then have

$$\text{Sr} = \text{Sr(C)} + \text{Sr(g)} = q\text{SrCs(C)} \times \text{Cs(C)} + q\text{SrCs(g)} \times \text{Cs(g)},$$

or

$$\text{Sr}/\text{Cs(C)} = q\text{SrCs(C)} + q\text{SrCs(g)} \times (\text{Cs(g)}/\text{Cs(C)}), \tag{1}$$

and, similarly,

$$\text{Sr}/\text{Pu(C)} = q\text{SrPu(C)} + q\text{SrPu(g)} \times (\text{Pu(g)}/\text{Pu(C)}). \tag{2}$$

Sr is a measured quantity, Cs(C) and Cs(g) can be calculated from measured ^{137}Cs and ^{134}Cs activities. Therefore, if the activity ratios $q\text{SrCs(C)}$ and $q\text{SrCs(g)}$ are approximately constant in one type of sample, Eq. (1) has the form $y = a + bx$, and we can estimate a and b , which are the mean activity ratios we seek, by linear regression analysis of Eq. (1). The same applies for $q\text{SrPu(C)}$, g in Eq. (2).

However, we must be aware that there may be considerable uncertainties attached to y and x , due to small ^{134}Cs activity concentrations in many samples (the samples were taken and measured 10–18 years, or 5–9 half-lives of ^{134}Cs after Chernobyl), and also due to the small overall contribution of Pu(C) to total $^{239+240}\text{Pu}$. Therefore, due to uncertainty propagation, uncertainties of the components (global or Chernobyl) of ^{137}Cs and $^{239+240}\text{Pu}$ can be high. Also, since the fractions of Sr(C) in total ^{90}Sr and Pu(C) in total Pu are known to be small, in particular the estimates of $q\text{SrCs(C)}$ and $q\text{PuCs(C)}$ must be expected to be rather uncertain. The relative uncertainties will turn out to be of similar size for most samples, and samples with relative uncertainty that is “too” high, $1\sigma > \approx 120\%$ (chosen deliberately), will be discarded from some analyses (indicated when applicable).

Mathematically, parameter estimation in regression involving predictors (x) that are uncertain is not trivial and can have serious consequences on the estimate of the regression coefficient (e.g., Wikipedia, 2006; Riggs et al., 1978; Frost, 2000; Carroll et al., 1999; Carroll and Ruppert, 2002). However, apart from a short comment in the conclusions, we shall not discuss this in more depth in this paper.

If the q values have been determined, the ratio Sr(C):Sr(g) can be estimated,

$$q\text{SrPu(g)}/q\text{SrPu(C)} = (\text{Sr(g)}/\text{Sr(C)}) \times (\text{Pu(C)}/\text{Pu(g)}),$$

or

$$Q\text{Sr} := \text{Sr(C)} : \text{Sr(g)} = (q\text{SrPu(C)}/q\text{SrPu(g)}) \times (\text{Pu(C)}/\text{Pu(g)}) \quad (3)$$

(similarly for $q\text{SrCs}$). The fractions of Sr(C) and Sr(g) in total Sr are then

$$\text{Sr(g)}/\text{Sr} = 1/(1 + Q\text{Sr}),$$

$$\text{Sr(C)}/\text{Sr} = Q\text{Sr}/(1 + Q\text{Sr}).$$

3. Results

3.1. Strontium—caesium

In Fig. 1 the ratios $^{90}\text{Sr}:^{137}\text{Cs}$ (Chernobyl) are plotted against the Cs(global):Cs(Chernobyl) ratios according to Eq. (1) for all sample categories (no values were available for the sediments). The regression results are summarized in Table 2. It does not seem reasonable to perform a regression on all samples, given the very different behaviour of soil and cryoconite samples (Fig. 1). Fig. 2 shows the ratios for soil samples only, but separated for different soil layers. The rationale is that one can expect that the different mobilities of Cs and Sr in soil will lead to different $q\text{SrCs}$ ratios in different soil layers. The values are taken from various project reports, some of which do not quote uncertainties, unfortunately. Fig. 3 shows the ratio for cryoconite samples, separated into sampling sites at high (2800–2900 m) and low (2500–2600 m) altitudes on the glacier, together with regression lines. (The linear regression looks distorted in the log–log plot.) From the, on the average, higher Cs(g)/Cs(C) ratio of the “low” cryoconites than that of the “high” cryoconites, we conclude that the contribution of Chernobyl to the former is lower.

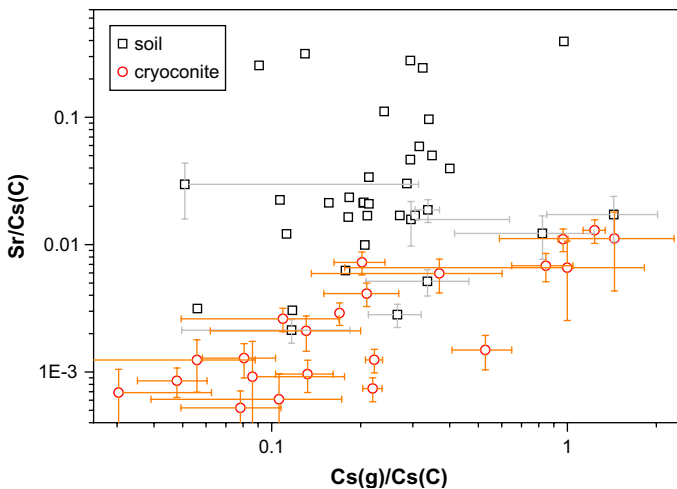


Fig. 1. Scatterplot of $^{90}\text{Sr}:^{137}\text{Cs}$ (Chernobyl) vs. Cs(global):Cs(Chernobyl) ratios; all samples.

Table 2
Ratios $^{90}\text{Sr}:^{137}\text{Cs}$ in global and Chernobyl fallout, different media

	Global	Chernobyl	<i>n</i>	qSrCs(C)/qSrCs(g)
All categories	—	—	58	
Soil	(0.068 ± 0.063)	0.044 ± 0.025°	36	0.65 ± 0.70
	§(0.066 ± 0.065)	§0.045 ± 0.026°	35	0.68 ± 0.78
0–3 cm	§((4.7 ± 6.3)E–3)	§0.0058 ± 0.0032°	17	1.2 ± 1.8
0–6 cm	§0.096 ± 0.051°	§(0)	25	
Cryoconite	(8.00 ± 0.96**)E–3	((8.1 ± 5.4)E–4)	22	0.10 ± 0.07
High altitude	((1.2 ± 1.0)E–3)	(8.9 ± 1.9**)E–4	12	0.74 ± 0.64
Low altitude	(6.1 ± 1.3**)E–3	(3.1 ± 1.1*)E–3	10	0.51 ± 0.21

** $p \leq 0.01$, * $p \leq 0.05$, ° $p \leq 0.1$, () $p > 0.1$. § Data with high uncertainty removed.

3.2. Strontium–plutonium

In Fig. 4 the ratios $^{90}\text{Sr}:^{239+240}\text{Pu}$ (Chernobyl) are plotted against the Pu(global):Pu(Chernobyl) ratios according to Eq. (2), for all three sample categories. The regression results are summarized in Table 3. Here the soil layers have not been separated, because the data are not sufficient for this purpose.

The lower qSrPu ratios in cryoconite than in soil and sediment indicate a faster depletion of Sr in cryoconite than in soil and sediments. We also observe that the ratio qSrPu(C)/qSrPu(g), namely the factor with which one has to multiply the Pu(C):Pu(g) ratio in order to obtain the Sr(C):Sr(g) ratio in Eq. (3), is quite different for different media. Even if the statistics are not very good in some cases, we see that this ratio is lower in soil and sediment than for cryoconite.

3.3. Plutonium–caesium

We now regress Pu(g) against Cs(g) and Pu(C) against Cs(C) in order to find the respective activity ratios. In a first step we perform a regression of type $y = a + bx$, and check if the

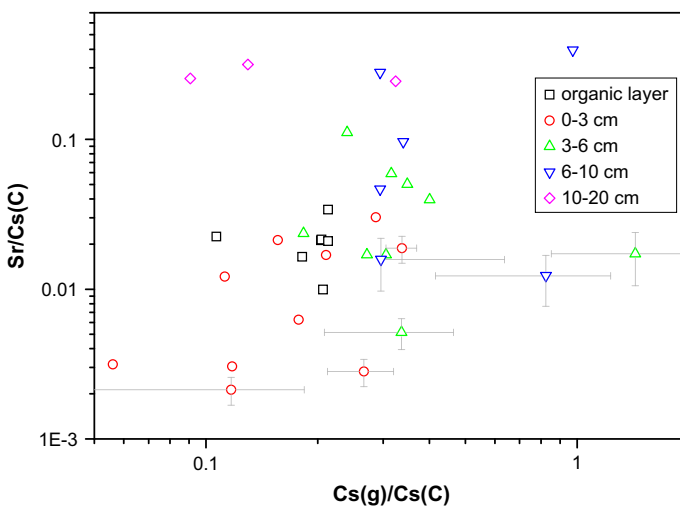


Fig. 2. Scatterplot of $^{90}\text{Sr}:^{137}\text{Cs}$ (Chernobyl) vs. $\text{Cs}(\text{global}):\text{Cs}(\text{Chernobyl})$ ratios; only soil samples. Different soil layers are separated. Only for some values are uncertainties available.

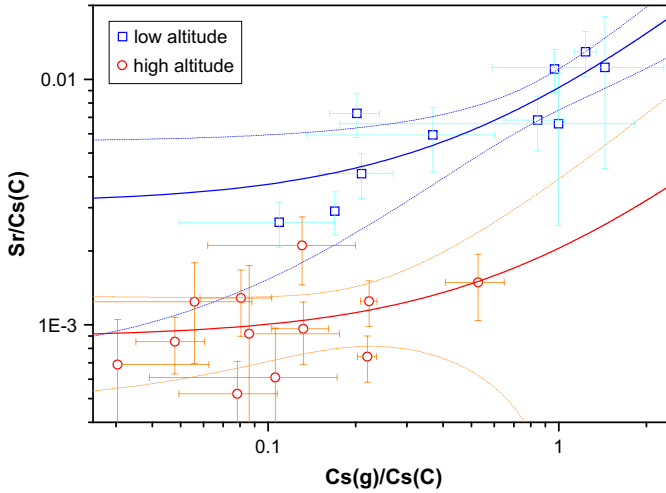


Fig. 3. Scatterplot of $^{90}\text{Sr}:^{137}\text{Cs}$ (Chernobyl) vs. $\text{Cs}(\text{global}):\text{Cs}(\text{Chernobyl})$ ratios; only cryoconite samples; samples from higher (altitude 2800–2900 m) and lower (2500–2600 m) sites on the glacier are plotted separately. Solid lines: linear regression, dotted: 95% confidence limits.

intercept (a) is not significantly different from zero [criterion: $p(\text{H}_0:a = 0) < 0.05$]. If this is the case, then in a second step we force the regression through zero, by setting $y = bx$; the slope b is then the ratio we seek. If the intercept is significantly different from zero [$p(\text{H}_0:a = 0) \geq 0.05$], the second step cannot be strictly applied, and no ratio can be properly determined. We still do it in the analysis, but label the results with # in Table 4, and interpret the results with due suspicion. We can observe, however, that even if the results are methodologically questionable, they agree quite well with the statistically sounder ones (those with intercept = 0 with $p < 0.05$).

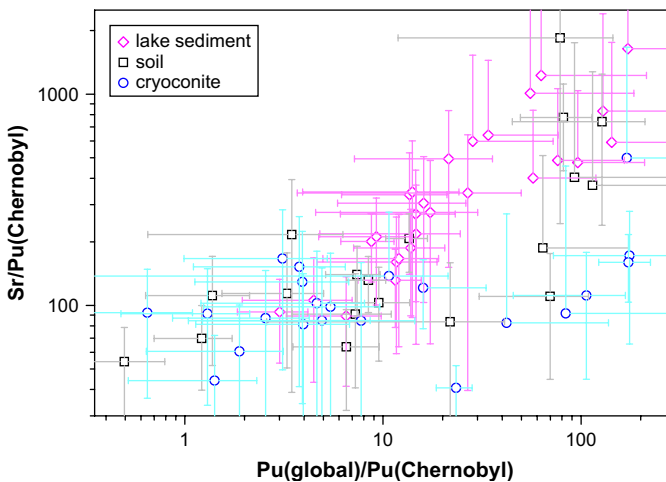


Fig. 4. Scatterplot of $^{90}\text{Sr}:^{239+240}\text{Pu}$ (Chernobyl) vs. $\text{Pu}(\text{global}):\text{Pu}(\text{Chernobyl})$ ratios.

Table 3
Ratios $^{90}\text{Sr}:^{239+240}\text{Pu}$ in global and Chernobyl fallout, different media

	Global	Chernobyl	<i>n</i>	qSrPu(C)/qSrPu(g)
All categories	3.65 ± 0.74**	158 ± 47**	67	43 ± 16
Sediment	6.2 ± 1.0**	191 ± 62**	27	31 ± 11
	§4.2 ± 1.0**	§185 ± 31**	21	46 ± 13
Soil	5.7 ± 2.0**	(92 ± 111)	19	(16 ± 20)
	§8.3 ± 3.0*	§ (65 ± 163)	12	(8 ± 20)
Soil + sediment	6.06 ± 0.99**	148 ± 58**	46	24 ± 10
	§6.77 ± 1.45**	125 ± 61*	33	18 ± 10
Cryoconite	0.91 ± 0.27**	87 ± 19**	22	96 ± 35
	§0.35 ± 0.14*	§96 ± 9**	19	274 ± 113
High altitude	(4.2 ± 3.4)	84 ± 16**	14	(20 ± 17)
Low altitude	(1.3 ± 0.7)	(31 ± 79)	8	(24 ± 62)
	§(0.53 ± 0.23)	§68 ± 28°	5	(128 ± 77)

** $p \leq 0.01$, * $p \leq 0.05$, ° $p \leq 0.1$, () $p > 0.1$. § Data with high uncertainty removed.

4. Discussion

The circumstances of the data – overall low ^{90}Sr and $^{239+240,238}\text{Pu}$ activity concentrations, on the average a low contribution of Chernobyl to these nuclides, ^{134}Cs measured quite long after fallout, not many samples due to their difficult acquisition in high alpine environment – inevitably gave rise to a high uncertainty. Nevertheless, we were able to produce reasonable regression statistics, and therefore reasonably reliable estimates of radionuclide activity ratios. The results are summarized and further discussed in the following: recall that they are specific to the investigated region, since radionuclide composition of the Chernobyl cloud was (to different degrees for different radionuclides) subject to spatio-temporal variability, as a result of the complicated release and transport history.

4.1. Strontium–caesium

In global fallout, we find an average $^{90}\text{Sr}:^{137}\text{Cs}$ ratio of about 0.07 in soil samples, and about 0.001–0.006 in cryoconite samples.

For Chernobyl fallout, we find a ratio of 0–0.045 for soil, and typically 0.001 in cryoconite. The estimates of the Sr/Cs ratio in soil are statistically not satisfying and require further

Table 4
Ratios $^{239+240}\text{Pu}:^{137}\text{Cs}$ in global and Chernobyl fallout, different media

	Global	Chernobyl
All categories	–	(9.8 ± 1.3**)E–6 (<i>n</i> = 32)#
		§(9.7 ± 1.0**)E–6 (<i>n</i> = 24)#
Soil	§0.0107 ± 0.0005** (<i>n</i> = 11)	(1.9 ± 0.2**)E–5 (<i>n</i> = 7)
		§(1.9 ± 0.1**)E–5 (<i>n</i> = 2)
Cryoconite	–	§(9.7 ± 1.1**)E–6 (<i>n</i> = 22)#
High altitude	–	(9.53 ± 0.64**)E–6 (<i>n</i> = 14)
Low altitude	0.0090 ± 0.0011** (<i>n</i> = 13)	§((4.7 ± 1.8)E–5) (<i>n</i> = 9)
Soil + cryo-low	§0.0090 ± 0.0008** (<i>n</i> = 23)	–

** $p \leq 0.01$, * $p \leq 0.05$, ° $p \leq 0.1$, () $p > 0.1$. § Data with high uncertainty removed. # Intercept sign. >0. No value: no correlation.

investigation. The reason is, probably, linked to the different mobilities of Cs and Sr in soil, which lead to different ratios in different soil depths.

Obviously Sr undergoes strong depletion in the media, compared with original fallout (Table 1), as far as the values can be considered representative of fallout in the investigation area (see discussion in Section). Depletion factors can be defined by

$$D := q_{\text{SrCs}}(\text{original fallout})/q_{\text{SrCs}}(\text{samples}).$$

Roughly estimated depletion factors are summarized in Table 6.

It is difficult to say why cryoconites taken from different altitudes on the same glacier behave so differently. It may have to do with different climatic conditions, which result in different freeze–thaw cycles of the cryoconite holes, and with different influence of microbial life in the cryoconites (Psenner and Sattler, 1998; Sattler et al., 2002), which seem to influence the chemical milieu and thus, possibly, the mobility of the elements. Further research on these phenomena is under way in a joint project between the Universities of Innsbruck and Salzburg, Austria.

4.2. Strontium–plutonium

In global fallout, we found an average $^{90}\text{Sr}:^{239+240}\text{Pu}$ ratio of about 6 in soil and sediment samples, and about 0.3–4 in cryoconites. Given a value of 35 from the literature for the fallout (calculated from Table 1, $\text{Sr}/\text{Pu} = (\text{Sr}/\text{Cs}) \times (\text{Cs}/\text{Pu})$), this result points to a very strong depletion of Sr; this is no surprise given its high mobility in the environment compared to Cs and Pu.

For Chernobyl fallout, we determined a ratio of about 65–200 for soil and lake sediments, and 30–80 for cryoconites. The respective literature value for fallout, calculated as above, is 2600. Again we find a strong depletion of Sr (Table 6).

For some reason, depletion of Sr against Pu is stronger in Chernobyl than in global fallout, although one would expect the opposite (as found for Sr vs. Cs), given a longer time available for depletion mechanisms (e.g. leaching) for the latter. This unexpected result must be left open to speculation at this point. Assuming a lower Sr(C):Pu(C) ratio in Table 1 (i.e. a higher contribution of Pu in Chernobyl fallout at the sampling locations than measured in air filters in Salzburg) could resolve the paradox. Another explanation may be found in the different physical speciation of Pu in global and Chernobyl fallout, the former contained in smaller particles, which may have an influence on weathering and leachability.

4.3. Plutonium–caesium

For global fallout, we found a $^{239+240}\text{Pu}:^{137}\text{Cs}$ ratio of about 0.01, which is in good accordance with literature (0.018). Obviously, Cs and Pu are both strongly fixed in the investigated media (soil and cryoconite) so that no significant, or at least no strong, depletion of Pu element is observable. The tendency of Pu to form complexes with organic matter (humic and fulvic acids) may, however, make a part of it more mobile for colloidal transport.

For Chernobyl fallout, the ratio is about $1-2 \times 10^{-5}$, apparently somewhat higher in soil than in cryoconite, but the data are not sufficient for a strong conclusion. In any case, the ratio seems to be somewhat higher than the one observed in air filters of Salzburg (Table 1), less than 100 km away, 6.6×10^{-6} . In any case, a Pu(C)/Cs(C) ratio that is higher in the investigated areas

than in integrated air in Salzburg is no reason to doubt the results, in view of the considerations given in Section 1.

4.4. Strontium global—Strontium Chernobyl

We use the results from tables to estimate the mean fraction of ^{90}Sr stemming from global fallout. For each sample, we multiply the fraction $\text{Cs(C)}/\text{Cs(g)}$ or $\text{Pu(C)}/\text{Pu(g)}$ with the $\text{qSrCs(C)}/\text{qSrCs(g)}$ or $\text{qSrPu(C)}/\text{qSrPu(g)}$ ratio, taken from Tables 2 and 3, whichever is applicable, according to Eq. (3). From this QSr value we calculate the fraction $\text{Sr(g)}/\text{Sr(total)}$.

In spite of the occasionally high uncertainties of the ratios $\text{qSrCs(C)}/\text{qSrCs(g)}$ etc., the results of Table 5 appear quite consistent. Here, the SDs are the external ones, calculated from the means of the individual data, but not taking into account their (internal) uncertainties, which can be quite high due to uncertainty propagation. The mean $\text{Sr(g)}/\text{Sr}$ ratios for the cryoconites calculated out of Sr/Cs and Sr/Pu are not too different; however, for individual samples the differences between the two fluctuate between (-28) and $(+61)\%$ -points, which does not make the method appear very reliable for partitioning the ^{90}Sr in individual samples.

For all samples we observe a surprisingly low $\text{Sr(g)}/\text{Sr}$ ratio, given that, if all Sr would be preserved in the samples, and considering that $\text{qSrCs(C)}/\text{qSrCs(g)} = 0.027$ (from Table 1) in “pure” (undepleted) fallout, the fraction $\text{Sr(g)}/\text{Sr}$ would be 89% in soil “Nassfeldalm” and 85% in “Postalm”. Since much more time has passed from Sr(g) deposition in the early 1960s to sampling than between Sr(C) deposition around 1 May 1986 and sampling, the high mobility of Sr has shifted the ratio towards Chernobyl- ^{90}Sr , which has had not so much time to be depleted, so to say.

However, the statistical quality of these estimates is still not satisfying, and we hope that by evaluating more samples we may reduce uncertainties.

The linear regressions performed here involve predictors (x) with measurement uncertainty, which is quite substantial in some cases. It is known from theory that the regression coefficient (the slope) is then $b^* = b/[1 + (\sigma_{\delta x}/\sigma_x)^2]$, where b is the true (unknown) regression coefficient, σ_x^2 the variance of the x data, and $\sigma_{\delta x}^2$ the variance of the uncertainty of x (supposed to be the same for all x , which is another idealization), i.e. the “true” slope b is systematically underestimated. However, since the data spread is quite large in the cases which have been discussed here, in spite of often large x uncertainties ($\sigma_{\delta x}$), the systematical error in estimating b (due to the denominator in b^* , called dilution or attenuation factor) should not be larger than, roughly estimated, 20–30%. It may prove necessary to investigate this effect further (it has been broadly discussed in epidemiology and dosimetry, e.g. Heid, 2002).

Table 5
Estimated fraction of ^{90}Sr which is attributable to global fallout

Sample	% $^{90}\text{Sr}(\text{global})/^{90}\text{Sr}(\text{total})$	
	From Sr/Cs ratios (%)	From Sr/Pu ratios (%)
Soil, Nassfeldalm	(27 ± 10)	
Soil, Postalm	(21 ± 8)	
Cryoconite, high altitude	(15 ± 10)	(15 ± 9)
Cryoconite, low altitude	(54 ± 23)	(39 ± 17)
Lake sediments		(36 ± 22)

AM ± SD(external) of the samples.

Table 6

Estimated depletion of Sr against Cs and Pu and Pu against Cs in various environmental media, against original fallout

	Medium	$D(\text{global})$	$D(\text{Chernobyl})$
Sr/Cs	Soil	~ 10	~ 0.4–29
	Cryoconite	~ 80	~ 17
Sr/Pu	Soil, sediment	~ 6	~ 14–40
	Cryoconite	~ 10–100	~ 30–80
Pu/Cs	All	~ 2	~ 0.7

D : qSrCs(original fallout; Table 1)/qSrCs(actual samples), etc.

Due to their strong fixation in soil, sediment and cryoconites, almost the entire deposition of Cs, and most of Pu are still contained in these media (i.e. the activity apart from physical decay), while only a small fraction of Sr is left. It must be anticipated that it has been carried into ground and surface water bodies, and later via surface watercourses into seas and oceans as its final sinks. It would be interesting to check if sediments of the Black Sea (fed by the Danube and the Dnepr which drain regions with relatively high Chernobyl fallout), or the water itself, show an excess of ^{90}Sr with respect to original fallout. In fact, Egorov et al. (1999) report a $^{90}\text{Sr}/^{137}\text{Cs}$ ratio of 1.0 in the discharge of the Danube into the Black Sea between May 1986 and 1994 (weighted mean; calculated from the figures given in the article). This is much higher than the respective ratios in both global and Chernobyl fallout in environmental samples, as determined in our paper, and as known from original, both global and Chernobyl fallout.

Table 6 shows that depletion of Sr against Cs not only differs between media, but also for different times that are available between fallout and sampling. We try to derive a very rough residence half-life (or ecological half-life) of Sr relative to Cs by assuming an exponential depletion model (physically plausible),

$$1/D(t) := \text{depletion ratio} = \text{prop. exp}(-\beta t),$$

where t is depletion time, β depletion constant and $T_{1/2} = \ln(2)/\beta$ the residence half-life. Comparing $D(t)$ at two times (i.e. for global and Chernobyl fallout), we find that

$$\beta = -\ln(D(t_2)/D(t_1))/(t_1 - t_2). \quad (4)$$

Evaluating this with a simulation method (see Appendix), we find the values for (AM $\pm 1\sigma$ SD) listed in Table 7. No increase of depletion of Sr against Pu with time can be found, but numerically instead a decrease (negative β , labelled with \$ in the table), the reason for which is unknown (see also the discussion above).

Table 7

Estimated ecological depletion (or residence) half-lives, $T_{1/2}$ (years)

	Sr vs. Cs	Sr vs. Pu	Pu vs. Cs
All samples	5.0 ± 3.0 a	(23 ± 59 a)	8.6 ± 2.6 a
Soil	4.6 ± 1.6 a	\$	
Cryoconite	5.2 ± 1.5 a	\$	

\$: numerically negative; () not significant.

5. Conclusions and outlook

In this paper, we demonstrated a way to estimate the mean contribution of global and Chernobyl fallout to ^{90}Sr found in soil, lake sediment and cryoconite samples, based on regression with ^{137}Cs and $^{239+240}\text{Pu}$. The activity concentrations of the latter radionuclides can be separated into global and Chernobyl parts. We also gave results of activity ratios $^{90}\text{Sr} : ^{239+240}\text{Pu} : ^{137}\text{Cs}$ for global and Chernobyl fallout, which show a strong depletion of Sr against Cs and Pu, and an approximate preservation of the original ratio in fallout, for Pu:Cs, with possible slight depletion of Pu.

As research on Alpine and glacial radio-ecology goes on at the University of Salzburg, more data will be available in the future, and statistical scores of the results can be expected to improve. However, time runs out for sufficiently accurate radiometry of ^{134}Cs , since already 10 half-lives have passed since its deposition (after the Chernobyl accident in April 1986), and therefore separation of ^{137}Cs in its fractions stemming from global and Chernobyl fallout becomes increasingly difficult, if not impossible altogether, by this method. An alternative method may be to use ^{207}Bi as a proxy for global- ^{137}Cs (Bossew et al., 2006): it seems to appear in a fairly constant ratio in media such as soil and cryoconite, but due to low activity concentration it requires long measuring times and careful background correction.

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Appendix. A simulation method for evaluating depletion constants

The various uncertainties in the variables involved are modelled as follows.

The deposition time for global fallout is assumed to be between 1963 and 1965; Chernobyl is set to 1986.5; sampling between 1996.5 and 2004.5. For qSrCs(g) and qSrCs(C) we use the lower and upper margins of the estimates, Tables 2 to 4. The probability distributions of all variables were set to be uniform between the margins. Therefore, in each simulation, we sample a random value for each variable, say z :

$$z = z_{\text{lower}} + (z_{\text{upper}} - z_{\text{lower}})r, \quad r \sim U([0, 1]) \Rightarrow z \in [z_{\text{lower}}, z_{\text{upper}}] \text{ uniformly.}$$

For the qSrCs(C) etc., in order to account better for small values, we use instead

$$z = z_{\text{lower}} \left(z_{\text{upper}} / z_{\text{lower}} \right)^r \Rightarrow \log z \in [\log z_{\text{lower}}, \log z_{\text{upper}}] \text{ uniformly.}$$

With variables generated in this way, β is calculated according to Eq. (4). This is repeated $N = 10\,000$ times, and the statistics for β are calculated. A simple QBasic program was used for the simulation.

The resulting frequency distribution for $\beta(\text{soil})$ is shown in Fig. A.1. The estimates of β are probably conservative, since assuming uniform distributions exaggerates the uncertainties. The method can certainly be refined by using more realistic uncertainty modelling.

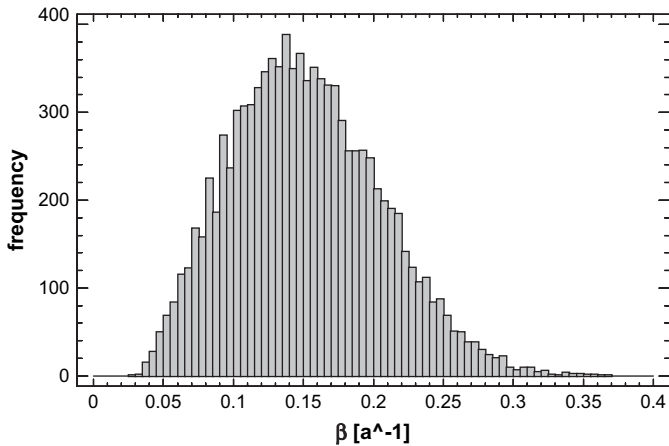


Fig. A.1. Frequency distribution of β (soil; Sr vs. Cs) resulting from 10 000 simulations.

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