Journal of Environmental Radioactivity 102 (2011) 987–994

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Inventory and vertical migration of ^{90}Sr fallout and ^{137}Cs/^{90}Sr ratio in Spanish mainland soils

M. Herranz [a], L.M. Romero [b], R. Idoeta [a], C. Olondo [a,*], F. Valiño [b], F. Legarda [a]

[a] Department Nuclear Engineering and Fluid Mechanics, University of the Basque, Country (UPV/EHU), Alda Urquijo s/n, E-48013 Bilbao, Spain
[b] CIEMAT, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Av. Complutense 22, 28040 Madrid, Spain

ARTICLE INFO

Article history:
Received 30 March 2011
Received in revised form
7 July 2011
Accepted 16 July 2011
Available online 6 August 2011

Keywords:
^{90}Sr inventory in soils
^{90}Sr vertical migration in soils
Radioactivity in soil
Undisturbed soil
Fallout

ABSTRACT

In this paper the inventory of ^{90}Sr in 34 points distributed along the Spanish peninsular territory is presented. Obtained values range between 173 Bq/m^2 and 2047 Bq/m^2. From these data set and those ^{137}Cs data obtained in a previous work the ^{137}Cs/^{90}Sr activity ratio has been established, laying this value between 0.9 and 3.6. Also the migration depth of both radionuclides has been analysed obtaining for ^{137}Cs an average value 57% lower than that obtained for ^{90}Sr.

Additionally, this paper presents the results obtained in 11 sampling points in which the activity vertical profile has been measured. These profiles have been analysed to state the behaviour of strontium in soils and after, by using a convective–diffusive model, the parameters of the model which governs the vertical migration of ^{90}Sr in the soil, *v* (apparent convection velocity) and *D* (apparent diffusion coefficient) have been evaluated. Mean values obtained are 0.20 cm/year and 3.67 cm^2/year, respectively.

1. Introduction

As a result of the nuclear era, which began in 1945, different radioactive pollutants releases were produced all over the Earth. The main sources of these releases were nuclear weapon testing, occurring between 1950 and 1980, and the accident which occurred in the Chernobyl's nuclear power plant in 1986 (UNSCEAR, 1982, 1988, 2000a, b). Obviously, all radioactive discharges have produced radioactive deposits in soils whose deposition distribution is different along the Earth's surface.

Among the different released fission radionuclides, ^{90}Sr and ^{137}Cs are the most significant ones as they contribute to long term doses to population (UNSCEAR, 1982; NCRP, 2007; Francis, 1978) due to their radioactive long life. This fact causes the necessity of having a detailed knowledge of their distribution and behaviour in soils.

The knowledge of the activity per unit area of both radionuclides, or inventory, that soils present at a given date, allows the evaluation of the impact that any new potential release of these radionuclides would produce. The knowledge of their behaviour in soils, such as their vertical migration, would help develop remediation actions and to have more dose estimations.

The amount of knowledge of these two radionuclides is very different. Whereas the presence of ^{137}Cs has been intensively analysed all over the world (NCRP, 2007), the number of studies that focus on ^{90}Sr is much lower and the vast majority of these studies have analysed ^{90}Sr content only near the soil surface and, as a result, not all ^{90}Sr content present in soil has been obtained (Bossew et al., 2007; Ehlken and Kirchner, 1996; Forsberg et al., 2000; Kim et al., 1998; Pröhl et al., 2006; Schimmack et al., 2003; Quang et al., 2004). It must be taken into account that ^{90}Sr can reach a depth near 120 cm. A reasonable motive why these two radionuclides present such a difference in the number of realized studies lies firstly in the fact that it is well known that the major contributor to the total dose to be received from soil is ^{137}Cs (UNSCEAR, 1972), and secondly, ^{90}Sr needs a laborious and expensive radiochemical separation procedure to quantify its activity content, as Schimmack et al. (2003) also points out, whereas ^{137}Cs is measured directly by gamma spectrometry.

In Spain, as well as in other countries, only a few studies were carried out to know the amount of ^{90}Sr in soils and, except for that developed by Gil-García et al. (2008), the rest of them have all focused their respective studies in just one of the different Spanish regions (Baeza et al., 1993; Gómez et al., 1997; Herranz et al., 2001). Additionally, their sampling programmes were developed only to obtain samples between 3 and 20 cm soil depth, not enough to obtain all the ^{90}Sr present in soil.

---

[*] Corresponding author. Tel.: +34 94 601 3964; fax: +34 94 601 4159.
E-mail address: kontxi.olondo@ehu.es (C. Olondo).

0265-931X/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2011.07.004

To establish the inventory of these two radionuclides in soils of the peninsular territory of Spain, a research project has been carried out with the support of the Spanish Nuclear Safety Council (CSN). To deal with it, a sampling programme covering all the territory under study was accomplished and, as a result, thirty-four undisturbed soil samples have been collected, five of them located in the surroundings of the five Spanish nuclear power plants. The soil depth sampled was established as 125 cm in order to be able to take all existing content of ^{137}Cs and ^{90}Sr. Details about the sampling strategy and programme, as well as sampling methodology, are presented in Legarda et al. (2011).

In all these samples, the ^{90}Sr activity inventory was obtained by means of the radiochemical isolation of the Sr and its further measurement with a gas-flow proportional counter. In twelve of these samples and in order to get information about the strontium vertical migration, the vertical activity profiles were obtained following the same analytical procedure.

As it was concluded in a previous study about ^{137}Cs contents in the Spanish mainland (Legarda et al., 2011), the radioactive impact of the Chernobyl nuclear power plant accident has had a negligible contribution to the ^{90}Sr radioactive inventory on the Iberian peninsula (Appleby et al., 2002; Baeza et al., 1991; Ferrero et al., 1987; González et al., 2004; Llaurado et al., 1994; Navas et al., 2007; De Cort et al., 1998; UNSCEAR, 2000a) and so, results obtained in this work are regarded as coming only from the fallout of nuclear weapon testing.

This paper presents the ^{90}Sr inventory in the Spanish mainland soils. The earlier knowledge of ^{137}Cs inventory in these soils allows calculating the ^{137}Cs/^{90}Sr activity contents ratio and by means of this ratio average value and the Spanish map of ^{137}Cs (Legarda et al., 2011), ^{90}Sr inventory in Spanish mainland can be established.

This paper also presents the results obtained for the ^{90}Sr vertical activity profiles. These profiles have been analysed with a convective–diffusive migration model which governs the strontium vertical migration through soils. The obtention of these profiles has provided values for the parameters of the model for strontium. Differences in behaviour between caesium and strontium have been found and some explanation has been provided.

2. Material and experimental methods

2.1. Sampling points

The Spanish Peninsular mainland has an area of about 500,000 km^2. The sampling has been carried out using a 150 × 150 km mesh grid which allows obtaining 29 square regions. In each of these regions one sampling point has been selected in the neighbourhoods of a meteorological station, which had recorded rainfall data for all of the fallout period, from 1950 to 1980. These sampling points were selected considering that they were located on a soil which has not been disturbed from the beginning of nuclear weapons tests and that they should belong to one of the principal types of soils of the Spanish Mainland.

Another five sampling points have been sampled in the surroundings of five Spanish nuclear power plants. In this case, each one of these five sampling points has to fulfil the same standards requirements as the rest of the sampling points. So, the total number of sampling points was 34.

2.2. Sample collection and preparation

At each point two cylindrical sub-samples of 125 cm depth and 6 cm diameter were taken.

The sub-samples of 22 sampling points were cut into 30, 20 and 10 cm long slices. The rest of the sub-samples, 12, were cut to form 6 cm long slices. The samples were made by mixing the corresponding slices of the two sub-samples. After its preparation, fine powder test samples were obtained.

More details about sample collection and preparation are provided in (Legarda et al., 2011).

2.3. Strontium radiochemical isolation

The radiochemical procedure followed to obtain the ^{90}Sr activity in soil has been the following:

Firstly, the soil test sample has been calcinated to eliminate the organic material in a mufla oven at least for 12 h at 600 °C and 30 g of the remaining is taken. 8M HNO_3 are slowly added to the sample and, when effervescence is finished, Sr (Sr^{2+} 60 mg) carrier is added in order to get a balance between the two strontiums. After that, 100 ml of 8M HNO_3 are added again and, at least for 8 h, the sample is left for digestion over a hot plate.

After filtering and adjusting the pH to 4.8 with NH_4OH, the remaining sample dissolution is poured through a Dowex 50W-X8 cationic exchange column in order to retain the strontium. After that, and using Na_2EDTA the yttrium is eluted from the column and that moment is recorded as the strontium–yttrium separation time. The strontium is eluted from the column by means of NaCl and the obtained dissolution, after its pH is adjusted to 8 with NH_4OH, is precipitated with Na_2CO_3. After filtering and drying, the obtained precipitate is dissolved in HNO_3 and its pH is adjusted to 8 with NH_4OH. Later, barium acetate tampon and sodium chromate are added to the dissolution to obtain a barium chromate precipitate which, in theory, should be free from the existing alpha emitters in the dissolution. After filtering, Na_2CO_3 is added to the remained solution to obtain S_rCO_3 as a precipitate. That precipitate is deposited on a Whatman filter by filtration, constituting the test sample to be analysed in a gas-flow proportional counter.

The chemical yield is obtained by emission in an Atomic Absorption Spectrometer once the sample has been measured in a gas-flow proportional counter.

2.4. ^{90}Sr activity determination

The test sample filters were measured by a gas-flow proportional counter for at least 1000 min in order to obtain detection limits lower than 0.1 Bq/kg. Measurements were made in ^{90}Sr–^{90}Y equilibrium conditions that is, at least after 21 days from strontium–yttrium separation time.

The equipment used was a Canberra proportional counter, model HT1000, with 4 independent detectors of 2 inches in diameter, provided with mylar windows, whose nominal background is lower than 1 cpm and its detection efficiency is around 40% for ^{90}Sr + ^{90}Y mixed emissions.

Specific calibrations were carried out using a calibration source, prepared following the same radiochemical procedure over distilled water spiked with a certified reference solution, from NIST, containing ^{90}Sr and ^{90}Y in equilibrium.

To take into account self-attenuation effects due to the different samples thicknesses, a curve that provides the self-attenuation factor (S_F) as a function of the sample mass-thickness (gr/cm^2) was determined experimentally. This curve was obtained by fitting the results from the measurements of different samples, prepared with different and known mass-thicknesses, from a ^{90}Sr + ^{90}Y standard solution.

The result of the measurements were analysed using AlphaBeta, a self-developed software, which uses the following expression for activity concentration, uncertainties and detection limits calculations.

Table 1
^{90}Sr and ^{137}Cs inventory measured at each sampling point (Reference date = January 1, 2006) in Bq/m^2 with its expanded uncertainty (coverage factor $k = 2$) and ^{137}Cs and ^{90}Sr inventory ratio obtained (^{137}Cs/^{90}Sr). The code of sampling points is a C with a number corresponding to one of the 29 square regions in which the Spanish Peninsular mainland has been divided for this study. Sampling points with a CN denote a sampling point near a nuclear power plant.

Sampling point	^{90}Sr Inventory $\pm u$ (Bq/m^2)	^{137}Cs Inventory $\pm u$ (Bq/m^2)	^{137}Cs and ^{90}Sr inventory ratio
C2	1017 ± 29	2771 ± 64	2.7
C3	720 ± 42	1473 ± 111	2.0
C4	971 ± 26	2619 ± 85	2.7
C5	692 ± 32	947 ± 38	1.4
C7	523 ± 22	1539 ± 53	2.9
C8	531 ± 17	1097 ± 56	2.1
C9	857 ± 35	1113 ± 40	1.3
C10	781 ± 44	2626 ± 165	3.4
C12	1475 ± 77	2434 ± 139	1.7
C13	984 ± 33	2653 ± 79	2.7
C14	587 ± 56	817 ± 63	1.4
C15	666 ± 51	1120 ± 166	1.7
C16	180 ± 44	251 ± 38	1.4
C17	749 ± 82	1035 ± 68	1.4
C18	2022 ± 118	2457 ± 148	1.2
C19	440 ± 12	1557 ± 53	3.5
C20	910 ± 59	1415 ± 123	1.6
C21	567 ± 19	1157 ± 32	2.0
C22	173 ± 42	619 ± 86	3.6
C23	619 ± 21	1108 ± 38	1.8
C24	908 ± 77	1462 ± 124	1.6
C25	1059 ± 30	2153 ± 44	2.0
C26	679 ± 85	1972 ± 172	2.9
C28	778 ± 23	2669 ± 86	3.4
C29	424 ± 16	934 ± 18	2.2
C30	763 ± 63	657 ± 54	0.9
CN 04	198 ± 26	623 ± 67	3.1
CN 14	394 ± 35	821 ± 113	2.1
CN 15	798 ± 67	1552 ± 107	1.9
CN 17	1069 ± 43	979 ± 61	0.9
CN 21	2047 ± 119	4219 ± 232	2.1

^{90}Sr activity concentration is calculated by equation (1):

$$A = \frac{r_g - r_0}{2 \cdot m \cdot R \cdot (\varepsilon / S_F)} \quad [Bq/kg] \tag{1}$$

Where:

r_g is the gross count rate, in per second (s^{-1}),
r_0 is the background count rate, in per second (s^{-1}),
m is the sample mass (kg),
R is the chemical yield,
S_F is the self-attenuation factor and ε is the counting efficiency for the ^{90}Sr + ^{90}Y calibration source.

The combined relative uncertainty u_{rel} is obtained following the GUM methodology (ISO, 1993; Herranz et al., 2008) as equation (2) shows:

$$u^2_{\mathrm{rel}}(A) = \frac{(r_g/t_g + r_0/t_0)}{(r_g - r_0)^2} + u^2_{\mathrm{rel}}(m) + u^2_{\mathrm{rel}}(R) + u^2_{\mathrm{rel}}(\varepsilon) + u^2_{\mathrm{rel}}(S_F) \tag{2}$$

where t_g and t_0 are the sample and background counting time, respectively, in second (s) and u_{rel} (x) is the relative standard uncertainty of the variable x.

The detection limit (L_D) is calculated according to ISO standards (ISO, 2005; Herranz et al., 2008) and established previously so that it would have a maximum value of 0.1 Bq/kg. Equation (3) is used to calculate it:

$$L_D = \frac{2L_C + k^2/(2 \cdot t_g \cdot R \cdot m \cdot \varepsilon / S_F)}{1 - k^2 \cdot \left[u^2_{\mathrm{rel}}(m) + u^2_{\mathrm{rel}}(R) + u^2_{\mathrm{rel}}(\varepsilon) + u^2_{\mathrm{rel}}(S_F)\right]} \tag{3}$$

where L_C is the critical level, which is calculated as follows:

$$L_C = k \cdot \frac{S_F \cdot \sqrt{r_0(1/t_g + 1/t_0)}}{2 \cdot m \cdot R \cdot \varepsilon} \tag{4}$$

Coefficient k is the quantile of the standard normal distribution for the probability $1 - \alpha$, and it has been assumed a value of 1.65 which implies a confidence level of 95%.

3. Results and discussion

3.1. Inventory

The inventory of ^{90}Sr at each sampling point, expressed as the activity deposited per unit area of ground, has been obtained by adding up the activity per square metre contained in each slice of soil. Obtained values, in units of Bq/m^2, are shown in Table 1, and the reference date for these values is the 1st January 2006. These values are reported together with their corresponding relative expanded uncertainties u_{rel}, with a coverage factor $k = 2$. Reported u_{rel} values range from 2% to 24%, with a mean value of 7%. The relative uncertainty corresponding to sampling and preparation has been estimated to be about 5%.

Three sampling points have been removed from the data set, C01, C06 and C11, because they present an anomalous activity vertical profile as a result of floods and snowfalls.

Table 1 also contains the results for those five points that had been sampled in the vicinity of the nuclear power plants. These points are coded with CNX, where X represents the number of the square region that contains the corresponding sampling point.

Table 2
Depth reached by ^{137}Cs and ^{90}Sr in the sampling points. For points in which only the inventory has been measured, this value represents the depth in which 95% of ^{137}Cs and ^{90}Sr is contained.

	Sampling point	^{137}Cs reached depth (cm)	^{90}Sr reached depth (cm)
Only for inventory	C3	50	60
	C10	60	80
	C12	30	60
	C14	30	30
	C15	30	50
	C16	30	30
	C17	30	50
	C18	50	100
	C20	50	80
	C22	30	90
	C24	30	30
	C26	30	30
	C29	45	54
	C30	30	50
	CN 04	50	70
	CN 14	30	70
	CN 15	30	70
	CN 17	30	40
	CN 21	60	80
For inventory and activity vertical profile	C2	27	42
	C4	39	84
	C5	15	24
	C7	15	42
	C8	12	36
	C9	12	42
	C13	21	96
	C19	30	66
	C21	24	30
	C23	24	36
	C25	33	42
	C28	24	54

As seen in Table 1, ^{90}Sr inventory values range from 173 Bq/m^2 to 2047 Bq/m^2, with a mean value of 793 Bq/m^2 and with 436 Bq/m^2 as the standard deviation of the ^{90}Sr inventory data set.

The obtained inventory values of sampling points near the nuclear power plants match those from the rest of sampling points, as can be observed in Table 1. These values have been the expected ones taking into account the results of the developed surveillance programs in the Spanish nuclear power plants surroundings (González et al., 2004).

In addition, the inventory mean value matches the expected value presented in UNSCEAR (2000a), of about 900 Bq/m^2, for the studied latitude and longitude.

3.2. *^{137}Cs and ^{90}Sr activity inventory ratio*

In the same Table 1 the inventory obtained for ^{137}Cs with its expanded relative uncertainty value and the ^{137}Cs and ^{90}Sr inventory ratio are also presented for each sampling point.

The obtained ^{137}Cs/^{90}Sr inventory ratio mean value is 2.1, with a standard deviation of 0.8. This ratio was established to be of about 1.6 (NATO, 2000; UNSCEAR, 1969; UNSCEAR, 1988), with a maximum of 1.85 in the period from 1963 to 1964 (SCOPE, 2000). These last values are average values obtained from air, rain and dry deposit measurements, and if corrected to this work activity reference date, 1st January 2006, they have values of about 1.7 and 2 respectively. The obtained ^{137}Cs/^{90}Sr inventory ratio value is coherent with those values.

Other studies (Baeza et al., 1993; Bossew et al., 2007; Gómez et al., 1997; Quang et al., 2004) show different ^{137}Cs/^{90}Sr inventory ratios, which are higher than those obtained in this work, but the vast majority of them had measured the ^{90}Sr activities only in the upper soil layer and so, these ratios would be the result of an underestimated ^{90}Sr inventory. Some of them (Baeza et al., 1993; Gómez et al., 1997) even state that ^{90}Sr must have migrated deeper into soil.

Taking into account the ^{137}Cs/^{90}Sr inventory ratio mean value and the map which shows the ^{137}Cs inventory in the Spanish mainland territory (Legarda et al., 2011), it could be possible to determine the distribution of ^{90}Sr in the territory under study.

3.3. *Chemical yield*

Strontium measurements (297) were made and the mean average yield lies on 52% with 17% standard deviation of obtained yield data set.

This yield is obtained by emission in an Atomic Absorption Spectrometer which works with a precision lower than 1%. These

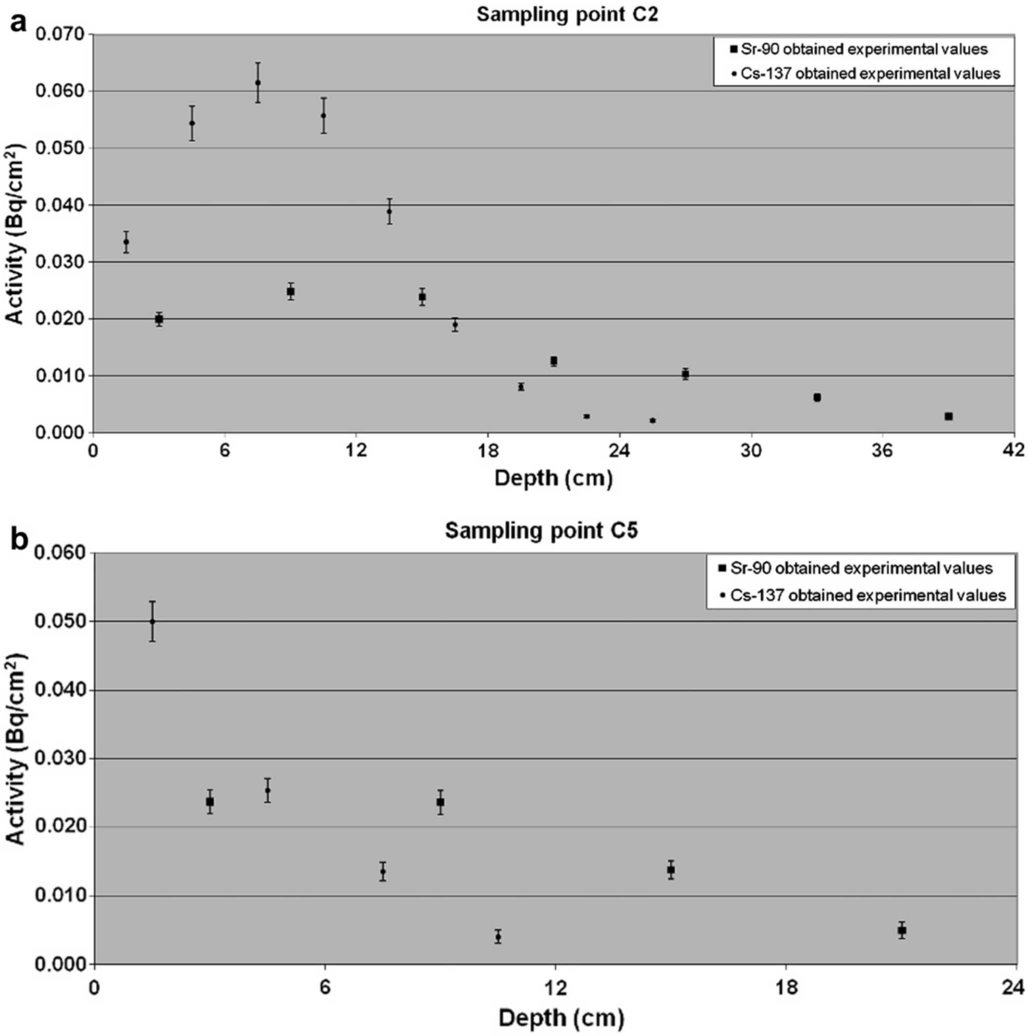

Fig. 1. ^{90}Sr and ^{137}Cs vertical activity profiles of sampling points C2 (a) and C5 (b). Their uncertainties are also shown (coverage factor = 2).

measurements were made destroying the sample after being measured.

As expected, the lowest chemical yields are obtained in those soils with high calcium contents. As calcium and strontium have a similar chemical behaviour, they compete in the extraction process and so, when the amount of calcium is much higher than that of the strontium, the strontium chemical yield is reduced.

3.4. ^{90}Sr and ^{137}Cs migration capacity in soils

In Table 2, a comparison between the depth attained by ^{137}Cs and ^{90}Sr in the soils analysed is shown. Depth attained has been defined as the depth at which more than 95% of radionuclide inventory is present. As seen in this Table 2, whereas strontium has reached a maximum of 100 cm depth, caesium has only reached a maximum of 60 cm.

Considering the mean value of the reached depths for both radionuclides it can be stated that the ^{137}Cs migrates 57% less than ^{90}Sr.

To obtain this value, only depths reached in the sampling points with the vertical profile obtained have been taken into account. In the other sampling points, as the length of slices has been longer, the precision of the depth reached has been lower and they have not been considered. Nevertheless, if those points are also taken into account, it can be observed that ^{137}Cs migrates 64% less than ^{90}Sr, which is a value similar to the first one.

The greater migration capacity shown by strontium compared to that shown by caesium can be explained by the fact that, whereas caesium is retained in soil by fixation processes, strontium is retained as exchangeable form. So, it will migrate deeper than caesium even if pH, cation exchange capacity and exchangeable calcium presence affect its vertical migration. This fact has been also pointed out in others studies developed not only in Spain (Baeza et al., 1995) but also in Europe, for example Forsberg et al. (2000) and Konshin (1992), and in Asia (Quang et al., 2004).

3.5. ^{90}Sr vertical migration analysis

The eleven ^{90}Sr activity vertical profiles (plot of activity deposited per unit area as function of depth) obtained show a similar behaviour, unlike those shown by ^{137}Cs with two different behaviours (Legarda et al., 2011). As an example, the activity vertical profiles of two of these 11 points, points C2 and C5, are shown in Fig. 1 for ^{137}Cs and ^{90}Sr.

Sampling point C13 has not been taken into account to perform the analysis of the ^{90}Sr vertical distribution. This point has been removed from this analysis because its vertical activity profile might be severely altered at deep depths, of about 55 cm–60 cm deep, for being very close to an irrigated land.

With the rest of the vertical activity profiles, and in order to perform the analysis of them, their normalised profiles (NP) have been obtained plotting, for each point, the fractional accumulated activity deposited per unit area a_i, in percentage, as a function of the fractional reached depth z_i, in percentage, as well.

Where a_i and z_i are calculated as follows:

$$a_i = 100\frac{\sum_{j=0}^{i} A_j}{A} \tag{5}$$

Where i is the index of the depth slice used in activity determination (1 corresponds to the soil slice nearest to ground surface), A_j is the contribution to the activity deposited per unit area from slice j, and A is the total activity deposited per unit area at the considered sampling point.

$$z_i = 100\frac{\sum_{j=0}^{i} Z_j}{Z_m} \tag{6}$$

Where Z_j is the depth of slice j, which is the bottom depth of each slice and Z_m is the depth of the last soil slice containing strontium, also taken as its bottom level, so that this value represents the depth reached by the ^{90}Sr in its migration into soil.

The 11 normalised profiles (NPs) obtained are shown in Fig. 2.

In Fig. 2, the soils whose texture is clay or loam are represented by solid lines and sandy soils, by dashed lines. As it can be observed in the figure, almost all normalised profiles describe a unique tendency in their behaviour, irrespective to the differences in the texture of soil they belong to, in contrast to the case of ^{137}Cs, which shows two different behaviours depending on the texture of the corresponding analysed soil (Legarda et al., 2011).

Only one of the analysed soils differentiates itself from the rest, C9. At this sampling point ^{90}Sr has the lowest retention in the upper soil depth and thus, the greatest mobility in this depth span (between 0 cm and 18 cm). At deeper soil depths, this behaviour is similar to that presented by the rest of the soils. This low retention could be explained by the low amount of organic matter that soil from this sampling point presents in this depth range, which is the lowest and thus, diminishes the hydrological retention capacity that this soil would present compared to soils of similar rainfall values and textures. Therefore, and as a result of this lower hydrological retention, rainfall would not be retained in the upper surface and it would penetrate deeper in the soil and wash off deposited strontium vertically more efficiently than in other soils.

3.6. ^{90}Sr migration model

The diffusion–convection model of radionuclide transport has been used to evaluate the vertical migration of strontium in soils

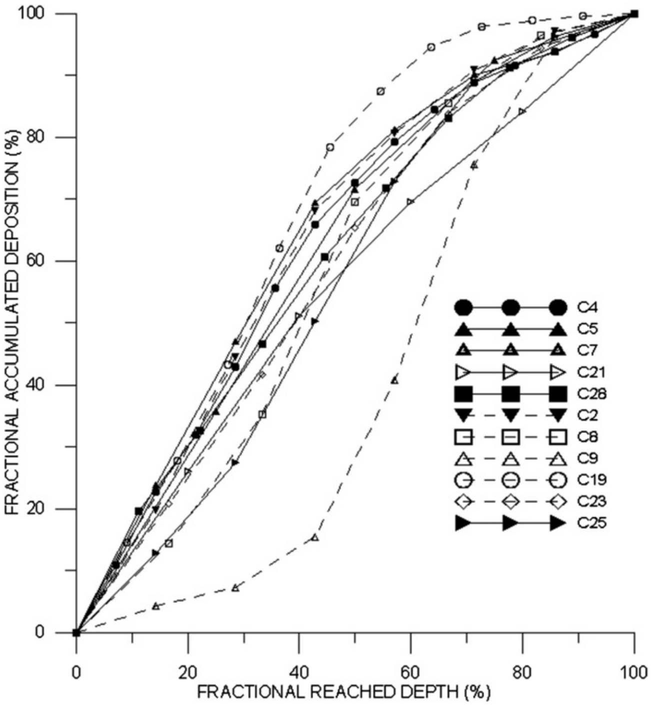

Fig. 2. ^{90}Sr normalised profiles (NP) of those sampling points whose activity vertical profile has been analysed.

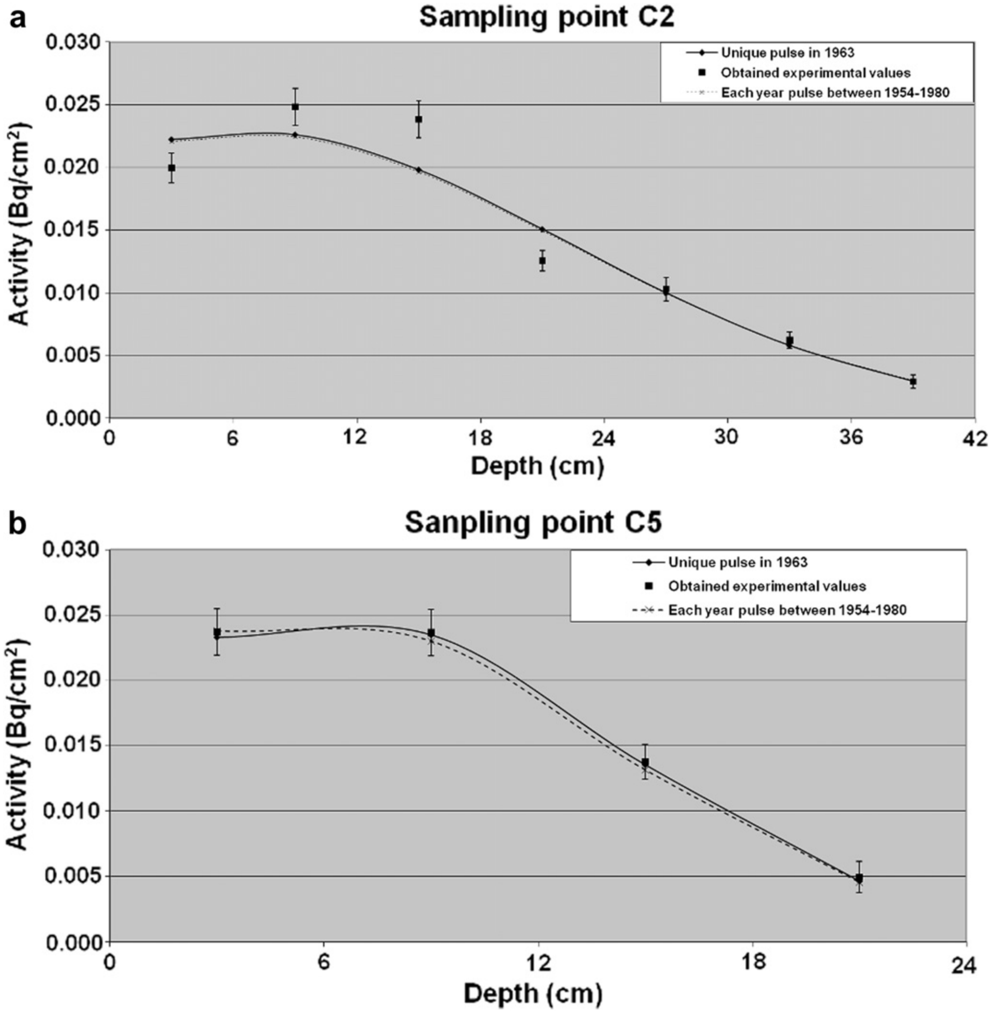

Fig. 3. Experimental and calculated ^{90}Sr activity vertical profiles for sampling points C2 (a) and C5 (b) using the convective–diffusive mass transport equation, showing the two different approaches studied: a pulse of ^{90}Sr every year during the fallout period and a unique pulse of fallout radioactivity in 1963.

(Bachhuber et al., 1981, 1982; Luckner and Schestakow, 1991; Van Genutchen and Cleary, 1979). This transport model is based on the unsteady-state, one dimensional convective–diffusive mass transport equation:

$$\frac{\partial C(z,t)}{\partial t} = D\frac{\partial C^2(z,t)}{\partial z^2} - v\frac{\partial C(z,t)}{\partial z} - \lambda C(z,t) \tag{7}$$

Where:

$C(z,t)$ is ^{90}Sr activity concentration in the soil at depth z and time t [$\text{L}^{-3}\text{T}^{-1}$].

$D = D_r/R$ is the apparent diffusion coefficient [L^2T^{-1}],

where D_r is the diffusion coefficient of ^{90}Sr in the soil solution and takes into account not only the molecular diffusion but also the hydrodynamic dispersion.

R is the retardation factor, a dimensionless coefficient, and calculated by $R = 1 + (\rho/\theta)K_d$ when a linear absorption isotherm is considered. ρ is the bulk density of the soil, θ is the soil porosity and K_d is the strontium distribution coefficient between both phases, solid and liquid.

$v = v_w/R$ is the apparent convection velocity [LT^{-1}], being v_w the pore water velocity.

λ, is the radioactive decay constant of ^{90}Sr [T^{-1}].

Whilst different solutions for this equation have been used to model ^{137}Cs vertical migration in soils with different results, this model has hardly ever been used for other radionuclides, including ^{90}Sr.

Bossew and Kirchner (2004) obtained a solution for this equation (7) considering: D, v and θ are constants for each soil; the soil as a half infinite space and the following boundary conditions: is 0 at z infinite, in order to obtain a real solution, and at $t = 0$ and $z = 0$, a pulse of value [$\text{L}^{-2}\text{T}^{-1}$] represents the input of ^{90}Sr in the soil.

$$C(z,t) = J_0 e^{-\lambda t}\left(\frac{1}{\sqrt{\pi D t}} e^{-(z-vt)^2/4Dt} - \frac{v}{2D} e^{vz/D} \mathit{erfc}\left(\frac{v}{2}\sqrt{\frac{t}{D}} + \frac{z}{2\sqrt{Dt}}\right)\right) \tag{8}$$

In order to obtain the theoretical contribution to the deposited activity per unit area that each soil slice (A_i^T) presents, this expression has been integrated over each slice, being the solution for this integration:

$$A_i^T = \int_{z_1}^{z_2} C(z,t)\mathrm{d}z = J_0 e^{-\lambda t}\left[1 + erf\left(\frac{z-\upsilon t}{2\sqrt{Dt}}\right) - e^{\frac{\upsilon z}{D}} erfc\left(\frac{z+\upsilon t}{2\sqrt{Dt}}\right)\right]_{z_1}^{z_2} \quad (9)$$

Where *erf* is the error function and *erfc* is the complementary error function.

In order to obtain a value for J_0, two different approaches have been compared:

- In the first one, a pulse of ^{90}Sr is considered every year during the fallout period, between 1954 and 1980. This pulse is calculated from experimental values of A in this work, which is the total activity deposited per unit area, and the fallout temporal pattern obtained from Milford Haven (UK) (Cambray et al., 1985) record station data. This is the nearest station to Spain with recorded data. The fallout temporal pattern used is the same as in the ^{137}Cs case because both radionuclides have shown a fairly constant ratio in deposition over a long time (UNSCEAR, 1982).
- In the second approach a unique pulse of value *A*, occurred in 1963, is considered. This approach has also been used by the Almgren and Isaksson (2006).

Both approaches have been used to obtain J_o values in all sampling points. Using both approaches in equation (9), with the same *v* and *D* tentative values, results obtained for A_i^T are almost identical (see Fig. 3). So, it is concluded that both approaches work properly, the second one being easier and this is finally used to obtain the velocity *v* and the diffusion coefficient *D* by fitting A_i^T to A_i using a least square method.

With these *v* and *D* fitted values the match between theoretical predictions (A_i^T) and experimental values (A_i) can be shown in Fig. 3.

The obtained values for *v* and *D* are presented in Table 3.

The obtained values of *v* range from 0.02 to 0.59 cm/year, with a mean value of 0.20 cm/year. For the diffusion coefficient *D* these values range from 0.53 to 11.51 cm^2/year with a mean value of 3.67 cm^2/year.

However, it must be considered that one of the soil parameters that characterises the radionuclide transport in soils, *v*, depends on the soil water content or rainfall. In order to remove this vertical migration dependence and to modelize the ^{90}Sr migration with parameters that depend only on soil characteristics, *v* has been divided by the mean annual rainfall, *r*. This new factor is called *f*.

Table 3
The velocity *v* and the diffusion coefficients *D* values for the sampling points whose vertical ^{90}Sr migration has been studied.

Sampling point	*v* (cm/year)	Mean *v* (cm/year)	*D* (cm^2/year)	Mean *D* (cm^2/year)
C2	0.10	0.20	3.18	3.67
C4	0.20		11.51	
C5	0.10		0.83	
C7	0.02		3.79	
C8	0.25		1.30	
C9	0.59		0.53	
C19	0.28		3.23	
C21	0.10		3.33	
C23	0.18		2.22	
C25	0.31		1.89	
C28	0.08		8.55	

Table 4
Factor *f* values for the sampling points whose vertical ^{90}Sr migration has been studied.

Sampling point	*f*	Sampling point	*f*	Mean *f*
C2	8.93E-04	C19	4.57E-03	3.39E-03
C4	1.51E-03	C21	2.43E-03	
C5	2.24E-03	C23	2.34E-03	
C7	4.85E-04	C25	5.26E-03	
C8	5.46E-03	C28	1.32E-03	
C9	1.08E-02			

$$f = \frac{\upsilon}{r}$$

As *r* can be calculated as $r = \theta \cdot \upsilon_W$, *f* has been obtained by means of the apparent velocity definition, so that $f = 1/R \cdot \theta$.

That is, *f* factor depends exclusively on soil and strontium properties. The *f* values obtained range from 1.08E-02 to 4.85E-04 with 3.39E-03 mean value, as it can be seen in Table 4.

The values obtained for *v*, *D* and *f* not differ very much between the two soil texture groups, as it was observed in the case of caesium. This is why a mean value of *f* factor and *D* diffusion apparent coefficient has been obtained, which would represent strontium behaviour in soils. With these values of *D* and *f* and knowing the porosity that a soil presents, strontium transport action would be predictable.

Anyway, the range of the values obtained support the fact that, as strontium mobility is greater than that shown by caesium in soils, the soil typology will affect less the former than the latter. This is the reason why only one trend has been observed and only one value of *D* and *f* represent its movement in soils.

4. Conclusions

As a result of the research program ^{90}Sr inventory in Spanish mainland soils has been obtained. These inventory values range from 173 Bq/m^2 to 2047 Bq/m^2, with a mean value of 793 Bq/m^2, which is in accordance with the expected value for this hemisphere and latitude based on formers deposits.

The ^{90}Sr values obtained in the Spanish nuclear power plants surroundings confirm what surveillance programs have been pointing out since their beginning: that Spanish nuclear power plants have not generated any relevant ^{90}Sr deposits during their operational life.

Taking into account the inventory obtained for ^{137}Cs in a previous work, the ^{137}Cs/^{90}Sr activity ratio has been obtained, showing a ratio that varies from 0.9 to 3.6 with a mean value of 2.1, which is coherent with those shown in previous literature. With the help of the ^{137}Cs map for Spanish mainland soils and by means of this ^{137}Cs/^{90}Sr inventory ratio it could be possible to establish the ^{90}Sr inventory map of Spanish mainland soils and so, after quantifying the impact that a future release would have, take the appropriate countermeasures.

A comparison between the migration capacities of ^{90}Sr and ^{137}Cs is also made and shows that ^{137}Cs migrates around 57% of the depth migrated by ^{90}Sr.

The analysis of vertical migration of strontium in the main soils of the Spanish territory has shown that ^{90}Sr has a unique behaviour irrespective of soil classification based on texture. This situation does not occur in the case of caesium in which two different behaviours were observed depending on soil texture.

The activity deposited per unit area at different depth values match the convective–diffusive model and the parameters that govern this vertical migration model through soils have been obtained by fitting. In the case of ^{90}Sr, these parameters are the same for all the soil types and have the following values: factor $f = 3.390\text{E-}03$ and apparent diffusion coefficient $D = 3.67\ \text{cm}^2/\text{year}$.

Acknowledgements

The work has been financially supported by the Spanish Nuclear Safety Council.

References

Almgren, S., Isaksson, M., 2006. Vertical migration studies of ^{137}Cs from nuclear weapons fallout and the chernobyl accident. Journal of Environmental Radioactivity 91, 90–102.

Appleby, P.G., Koulikov, A.O., Camarero, L., Ventura, M., 2002. The input and transmission of fallout radionuclides through Redó, a high mountain lake in the Spanish Pyrenees. Water, Air and Soil Pollution 2 (2), 19–31.

Bachhuber, H., Bunzl, K., Dietl, F., Kretner, R., Schimmarck, W., Schultz, W., 1981. Ausbreitung von Radionukliden im oberflächennaben Boden. GSF-Bericht S-788, Neuherberg.

Bachhuber, H., Bunzl, K., Schimmarck, W., 1982. The migration of ^{137}Cs and ^{90}Sr in multilayered soils: results from batch, column and fallout investigations. Nuclear Technology 59, 291–301.

Baeza, A., Del Río, L.M., Miró, C., Navarro, E., Paniagua, J.M., 1991. Radiological impact of the chernobyl nuclear power plant accident on two regions of Spain – Extremadura and Valencia. In: García-León, M., Madurga, G. (Eds.), Low-Level Measurements of Man-Made Radionuclides in the Environment. World Scientific Publishing Co. Pte. Ltd., Singapore, pp. 416–424.

Baeza, A., Del Río, L.M., Miró, C., Paniagua, J.M., 1993. Surface and depth fallout distribution of Cs-137 and Sr-90 in soils of Caceres (Spain) - dose commitments due to external irradiation. Journal of Radioanalytical and Nuclear Chemistry-Letters 175, 297–316.

Baeza, A., Del Río, M., Jiménez, A., Miró, C., Paniagua, J., 1995. Relative sorption of ^{137}Cs and ^{90}Sr in soil: influence of particle size, organic matter content and pH. Radiochimica Acta 68, 135–140.

Bossew, P., Kirchner, G., 2004. Modelling the vertical distribution of radionuclides in soil. Part 1: the convection–dispersion equation revisited. Journal of Environmental Radioactivity 73, 127–150.

Bossew, P., Lettner, H., Hubmer, A., Erlinger, C., Gastberger, M., 2007. Activity ratios of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in environmental samples. Journal of Environmental Radioactivity 97, 5–19.

Cambray, R.S., Playford, K., Lewis, G.N.J., 1985. Radioactive Fallout in Air and Rain: Results to the End of 1984. United Kingdom Atomic Energy Authority, Harwell, Oxon, UK.

De Cort, M., Dubois, G., Fridman Sh, D., Germenchuk, M.G., Izrael, Y.A., Janssens, A., Jones, A.R., Kelly, G.N., Knaviskova, E.V., Matveenko, I.I., Nazarov, I.M., Pokumeiko, Y.M., Sitak, V.A., Stukin, E.D., Tabachny, L.Y., Tsaturov, Y.S., Avdyushin, S.I., 1998. Atlas of Caesium Deposition on Europe After the Chernobyl Accident. EUR report nr. 16733. Office for Official Publications of the European Communities, Luxembourg.

Ehlken, S., Kirchner, G., 1996. Seasonal variations in soil-to-grass transfer of fallout stromtium and cesium and of potassium in north German soils. Journal of Environmental Radioactivity 33 (2), 147–181.

Ferrero, J.L., Jordá, M.L., Milió, J., Monforte, L., Moreno, A., Navarro, E., Senent, F., Soriano, A., 1987. Atmospheric radioactivity in Valencia, Spain, due to the chernobyl reactor accident. Health Physics 53, 519–524.

Forsberg, S., Rosén, K., Fernandez, V., Juhan, H., 2000. Migration of ^{137}Cs and ^{90}Sr in undisturbed soil profiles under controlled and close-to-real conditions. Journal of Environmental Radioactivity 50, 235–252.

Francis, C.W., 1978. Radiostrontium Movement in Soils and Uptake in Plants. Technical Information Center U.S. Department of Energy, Virginia.

Gómez, E., Garcías, F., Casas, M., Cerdá, V., 1997. Determination of ^{137}Cs and ^{90}Sr in calcareous soils: geographical distribution on the Island of Majorca. Applied Radiation and Isotopes 58, 699–704.

Van Genuchten, M.Th., Cleary, R.W., 1979. Movement of solutes in soil: computer – simulated and laboratory results. In: Bolt, G.H. (Ed.), Soil Chemistry, Part B Physico-Chemical Models. Elsevier, Amsterdam, pp. 349–386.

Gil-García, C.J., Rigol, A., Rauret, G., Vidal, M., 2008. Radionuclide sorption–desorption pattern in solis from Spain. Applied Radiation and Isotopes 66, 126–138.

González, A., Marugán, I., Rey del Castillo, C., Ramos, L.M., Salas, R., Sterling, A., 2004. Programas de vigilancia radiológica ambiental. Resultados 2002. Colección Informes Técnicos. INT-04.08. Consejo de Seguridad Nuclear, Madrid.

Herranz, M., Elejalde, C., Legarda, F., Romero, F., 2001. Sr-90 content of soils from Biscay (Spain). Applied Radiation and Isotopes 55, 521–525.

Herranz, M., Idoeta, R., Legarda, F., 2008. Evaluation of uncertainty and detection limits in radioactivity measurements. Nuclear Instruments and Methods in Physics Research Section A 595, 526–534.

ISO, 1993. Guide to the Expression of Uncertainty in Measurement (GUM). Guide 98. International Organization for Standardization, Géneve.

ISO, 2005. Determination of the Detection Limit and Decision Threshold for Ionizing Radiation Measurements – Part 7: Fundamentals and General Applications. Standard 11929–7. International Organization for Standardization, Géneve.

Kim, C.S., Lee, M.H., Kim, C.K., 1998. ^{90}Sr, ^{137}Cs, $^{239+240}$Pu concentrations in surface soils of Korea. Journal of Environmental Radioactivity 40, 75–88.

Konshin, O.V., 1992. Applicability of the convection–diffusion mechanism for modeling migration of ^{137}Cs and ^{90}Sr in the soil. Health Physics 63 (3), 291–300.

Legarda, F., Romero, L.M., Herranz, M., Barrera, M., Idoeta, R., Valiño, F., Olondo, C., Caro, A., 2011. Inventory and vertical migration of ^{137}Cs in Spanish mainland soils. Journal of Environmental Radioactivity 102, 589–597.

Llaurado, M., Vidal, M., Rauret, G., Roca, C., Fons, J., Vallejo, V.R., 1994. Radiocesium behaviour in Mediterranean conditions. Journal of Environmental Radioactivity 23, 81–100.

Luckner, L., Schestakow, W.M., 1991. Migration Proceses in the Soil and Groundwater Zone. Lewis Publishers, Michigan.

NATO, 2000. Atmospheric Nuclear Test. Environmental and Human Consequences. NATO ASI Series, Partnership Sub-Series:2. Environment. Springer.

Navas, A., Walling, D.E., Quine, T., Machín, J., Soto, J., Domenech, S., López-Vicente, M., 2007. Variability in ^{137}Cs inventories and potential climatic and lithological controls in the central Ebro valley, Spain. Journal of Radioanalytical and Nuclear Chemistry 274 (2), 331–339.

NCRP, 2007. Cesium-137 in the Environment: Radioecology and Approaches to Assessment and Management. NCRP REPORT No. 154. National Council on Radiation Protection and Measurements, Bethesda.

Pröhl, G., Ehlken, S., Fiedler, I., Kirchner, G., Klemt, E., Zibold, G., 2006. Ecological half-lives of ^{90}Sr and ^{137}Cs in terrestrial and aquatic ecosystems. Journal of Environmental Radioactivity 91, 41–72.

Quang, N.H., Long, N.Q., Lieu, D.B., Mai, T.T., Ha, N.T., Nhan, D.D., Hien, P.D., 2004. $^{239+240}$Pu, ^{90}Sr and ^{137}Cs inventories in surface soils of Vietnam. Journal of Environmental Radioactivity 75, 329–337.

Schimmack, W., Kracke, W., Sommer, M., 2003. Spatial variability of fallo-^{90}Sr and vegetation of an Alpine pasture. Journal of Environmental Radioactivity 65, 281–296.

SCOPE, 2000. Nuclear Test Explosions. Environmental and Human Impacts. In: Scientitfic Committee on Problems of the Environment, Series: SCOPE Report, vol. 59. John Wiley & Sons, Ltd.

UNSCEAR, 1969. Report of the United Nations Scientific Committee on the Effect of Atomic Radiation. Twenty-fourth Session Supplement No 13 (A/7613). United Nations, New York.

UNSCEAR, 1972. Ionizing Radiation: Levels and Effects. United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations Publication E.72.IX.17, New York.

UNSCEAR, 1982. Ionizing Radiation: Sources and Biological Effects. United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations Publication E.82.IX.8, New York.

UNSCEAR, 1988. Sources, Effects and Risks of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations. United Nations Publication E.82.IX.8, New York.

UNSCEAR, 2000a. Sources and Effects of Ionizing Radiation. Vol. I: Sources. United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations Publication E.00.IX.4, United Nations, New York.

UNSCEAR, 2000b. Sources and Effects of Ionizing Radiation. Vol. II: Effects. United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations Publication E.00.IX.4, United Nations, New York.