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Radiological Survey in Soil of South America

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1. Introduction

When the Earth was formed, the crust and consequently the soil and water were conformed by a wide variety of chemical elements with different concentrations; being some of these radioactives. There are different activity levels of natural radionuclides, as those of the ^{238}U and ^{232}Th decay chains, ^{40}K, ^{7}Be and ^{14}C, etc. along the planet [Cooper et al., 2003]. Among the 80 nuclides found in the environment, the more relevant concerning the radiobiological significance are ^{40}K, and the nuclides belonging to the ^{238}U and ^{232}Th decay chains. The human activities can strongly modify the natural concentrations due to the presence of residues or accumulation of elements caused by the release of effluents to the environment. In the 60´s the nuclear power production and nuclear weapon testing discharge to the environment anthropogenic nuclides. In particular, the Southern Hemisphere was mainly polluted by the debris originated in the South Pacific and middle Atlantic nuclear weapon tests [UNSCEAR, 2008]. Along with the class of anthropogenic gamma emitter nuclides releases, the ^{137}Cs is the most prominent isotope in the Earth crust originated by fission process. It is considered as one of the hazardous environmental contaminant due to the contribution to the external irradiation exposure and its incorporation to the human food chain [Singh et al., 2009].

Regardless, both natural and man-made nuclides have radiobiological implication because they significantly contribute to human external radiation dose and to the internal dose by inhalation and ingestion [Cooper et al., 2003; UNSCEAR, 2008]. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has estimated that exposure to natural sources is approximately 98% of the total radiation dose (excluding medical exposure) [UNSCEAR, 2000; UNSCEAR, 2008]. The dose arising from natural nuclides varies worldwide depending upon factors such as height above sea level, the amount and type of radionuclides in the air, food and water, as well as the concentration of the natural nuclides in the soil and rocks, which in turn depend on the local geology of each region, etc.

The information about the presence and migration anthropogenic radionuclides is crucial to fully understand the long-term behaviour in the environment, the uptake by flora and fauna including the human food chain, as well as potential contribution to groundwater. In consequence, before assessing the radiation dose to the population, a precise knowledge of the activity of a number of radionuclides is required [UNSCEAR, 2000;

UNSCEAR, 2008]. The mobility of the radionuclide in the ecosystem involves a number of complex mechanisms [Velasco et al., 2006; IAEA, 2010; Salbu, 2009; Cooper et al., 2003; Sawhney, 1972; Cornell, 1993; Staunton et al. , 2002; Bellenguer et al., 2008], and their transfer through the environmental compartments implies multiple interactions between the biotic and abiotic components of the ecosystem, as well as human interferences like the use of fertilizer [Tomazini da Conceic & Bonotto, 2006] or the overexploitation of the natural resources. For the identification of these interactions it is necessary to develop and test predictive models describing the radionuclide fluxes from the environment to the man.

In South America, the soil resource is extensively used in agriculture, stockbreeding and for building materials. Baselines of natural and anthropogenic activity nuclides in several countries are not established ye, as well regulations concerning the natural and anthropogenic activity and chemical restrictions in freshwater and food accordingly to the local situations. These facts and the scattered of the activity dataset put in relevance the present review on nuclide activity determinations in soils of South America, that could be considered as the first attempt in this direction.

A systematic compilation of radionuclide activity data of soil of Argentina, Brazil, Chile, Venezuela and Uruguay are presented. Radionuclide activity data concern to the natural ^{40}K, ^{238}U, and ^{232}Th and to the anthropogenic ^{137}Cs nuclides. These different pieces of information are put together, the quality of the environmental compartments is provided and the impact on the population is evaluated throughout the exposure dose. The migration of ^{137}Cs in soil is also analysed in the frame of different approaches [Kirchner, 1998; Schuller et al., 1997], and the transport parameters are discussed. Moreover, the caesium inventories are compared with the latitudinal UNSCEAR predictions [UNSCEAR 2000, UNSCEAR 2008].

2. Radionuclides in the environment

The man is continuously exposed to natural radiation since radioactive material is present in throughout nature. It occurs naturally in the soil, rocks, water, air, and vegetation. The components of the natural radioactive background are the cosmic radiation and the natural radioactivity of ground, atmosphere and water. Natural environmental radioactivity arises mainly from primordial radionuclides, such as ^{40}K and the nuclides from the ^{232}Th and ^{238}U series, which are at trace levels in all ground formations. Natural environmental radioactivity and the associated external exposure due to gamma radiation are primarily up to the geological and geographical conditions [UNSCEAR, 2000]. The specific concentrations of terrestrial environmental radiation are related to the composition of each lithologically separated area, and to the type of parental material from which the soils originate.

The high geochemical mobility of radionuclides in the environment allows them to move easily throughout the environmental matrixes. Rivers erode soil which contains radionuclides, and they reach lakes and oceans; atmospheric depositions can also occur on their surfaces; and groundwater containing some radionuclides can reach them.

Concerning the presence of artificial nuclides in the environment, after bombarding Hiroshima and Nagasaki in 1945, USA, USSR, France, England and Chine deserved to be a nuclear potency. In this frame, 543 underground and atmospheric nuclear weapon essays were carried from 1945 to 1980 in different regions of the globe. URSS, Chine and USA performed the tests in the North Hemisphere, while England and France in the South Hemisphere. The

underground essays were the more numerous; however, the global environmental impact resulted small because the radioactive material remains in the essay area. On the contrary, the atmospheric ones delivered to the atmosphere huge amounts of radioactive detritus causing a big impact on the environment [UNSCEAR, 2008; Valkovic, 2000]. It is worth to mention that because of the atmospheric circulation, approximately the 82 % of the debris remain in the hemisphere of injection [UNSCEAR, 2008; Valkovic, 2000]. The relevant nuclides originate in the essays were ^{3}H, ^{14}C, ^{54}Mn, ^{55}Fe, ^{85}Kr, ^{89}Sr, ^{90}Sr, ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{131}I, ^{137}Cs, ^{131}Ce and ^{144}Ce, among others [UNSCEAR, 1982]. Due to ^{90}Sr and ^{137}Cs are volatiles and have large half-life (28.6 years and 30.2 years, respectively) they are dispersed in the atmosphere, comprising the stratospheric global fallout, contributing to the residual background.
When analyzing the total annual effective dose received by human from natural sources, the dose received by the cosmic ray, terrestrial exposure, ingestion and inhalation of long-lived natural radionuclides needs consideration. Each environmental matrix, e.g. soil, air and water, has several associated pathways. These three environmental media cannot be thought as isolated and so, nuclide transfers are produced from one to the other. The different pathways exposure routes are schematized in the Fig. 1. The importance of these paths depends upon the particular radionuclide or radionuclides present in each compartment. The starting point to evaluate the people doses is to determine the nuclide concentrations in the environmental matrixes [USNCEAR, 2000; UNSCEAR, 2008].

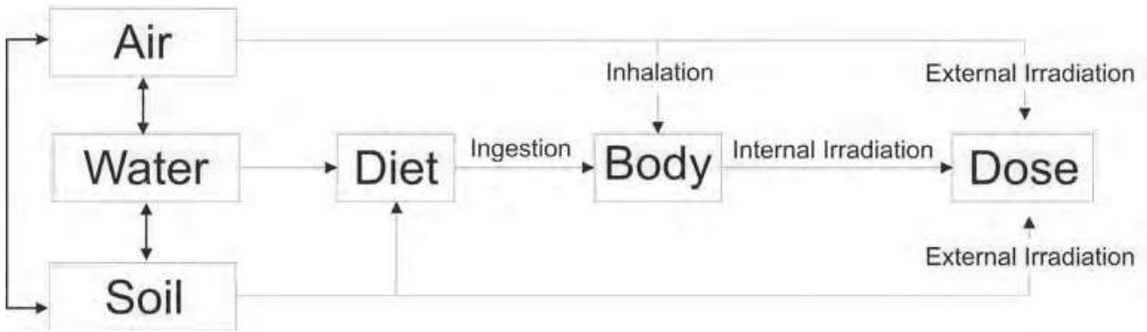

Fig. 1. Schematic terrestrial pathways of nuclide transfers and dose to humans.

3. Monitored regions and dataset

Two kinds of surveys have been performed, some of them deal with the determination of nuclide activity concentrations in depth, while others only reported single values of surface activity concentrations. Argentina, Brazil and Chile are the most studied countries, while there are reported a few data of Venezuela and Uruguay. The location of the monitored places, type of survey and monitored nuclides are summarized in the Table 1. Regarding the natural nuclides in South America, the reports of UNSCEAR only account values of the activity concentration of the natural nuclide ^{40}K for Argentina [UNSCEAR, 2000; UNSCEAR, 2008]. In San Luis Province, Argentina, two sites have been studied [Juri Ayub et al., 2008]. Recently, the first systematic studies to establish baseline activities for the naturally occurring radionuclides in unperturbed soils around La Plata city, Province of Buenos Aires, have been settled on samples taken from the surface down to a depth of 50 cm [Montes et al. 2010a, 2010b]. Moreover, in four superficial soils in the Ezeiza region, Argentina, the

activities of ^{40}K and of natural chains of ^{238}U and ^{232}Th have been determined [Montes et al. 2011]. In the Brazilian State of Rio Grande do Norte the average concentrations of ^{226}Ra, ^{232}Th and ^{40}K in unperturbed soils have been determined [Malanka et al., 1996]. Samples of soils were also studied in different departments of Uruguay since 2004 to determine the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th up to 5 cm of depth [Odino Moure, 2010]. Regarding the anthropogenic nuclides, in Argentina ^{137}Cs reference activity profile was determined in the Pampa Ondulada region [Bujan et al., 2000, 2003] and in the central part of the country in natural and semi-natural grassland regions [Juri Ayub et al., 2007, 2008]. Beside the natural chains values, the profiles of ^{137}Cs in the region of Buenos Aires Province have been settled [Montes et al. 2010a, 2010b]. Some studies have been performed in Brazil, dealing with the determination of the activity of the ^{137}Cs globally presented on the soil because of nuclear weapon tests [Correchel et al., 2005; Handl et al., 2008]. Total inventories and depth distributions of ^{137}Cs were established in agricultural and sheep-farming regions of Chile [Schuller et al., 1997, 2002, 2004]. In Uruguay, surface soil ^{137}Cs activity has been determined in different regions since 2004 [Odino Moure, 2010]. In Venezuela, the ^{137}Cs concentration at two different depth (0 cm -20 cm and 20 cm - 40cm) were measured [Sajó-Bohus et al., 1999].

3.1 Natural radionuclides

According to the UNSCEAR [UNSCEAR, 2000], in South America only the activity concentration of ^{40}K in unperturbed soils has been measured in Argentina (UN in Table 1 and 2), being the activity concentration range 540 Bq/kg -750 Bq/kg. Later, data profiles of ^{226}Ra and ^{40}K of semi-natural grassland soils of the central part of the country, Province of San Luis (AS23 and AS24) have been reported down to 25 cm depth [Juri Ayub, 2008]. The activity concentrations of ^{40}K were determined to vary from 720 Bq/kg to 750 Bq/kg very close to the upper limit of the values reported by UNSCEAR [UNSCEAR, 2000; UNSCEAR, 2008] , while ^{226}Ra activities were in the range 64 Bq/kg to 73 Bq/kg, as observed in Fig. 2. The profiles recorded down to 22.5 cm indicated that both nuclides activity concentrations are constant in depth (see Fig.2). Activity concentrations down to 50 cm of natural nuclides (^{238}U and ^{232}Th chains and ^{40}K) have been determined in soil samples collected from inland (AS1 and AS2) and coastal (AS3 and AS4) areas of the La Plata River, located in the North eastern region of the Province of Buenos Aires, Argentina [Montes et al., 2010a; Montes et al., 2010b]. The main observed activity resulted originated from the decay of the ^{40}K with following in importance those of the natural ^{238}U (obtained from the ^{226}Ra activity) and ^{232}Th (obtained from the ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl activities) chains, as shown in Fig. 2. While the activity of ^{235}U was, in all the cases, lower than the detection limit (L_D= 0.02Bq/kg), the activity values of the ^{238}U and ^{232}Th chains lay in the intervals 52 Bq/kg – 104 Bq/kg and 32 Bq/kg - 50 Bq/kg, respectively. In the case of the ^{238}U, the activities resulted to some extent high when comparing with data from Uruguay [Odino Moure, 2010]. It was also observed that the coastal soils without magnetite and lower hematite relative fraction presented a higher U probably related to the geological origin of the soils [Montes et al., 2010a; Montes et al., 2010b]. The ^{40}K activity profiles were quite different when comparing the monitored soils, ranging the surface activities values from 531 Bq/kg to 873 Bq/kg as observed in Fig. 2. In the inland profiles, the activity increased with depth and the depletion of the activity was detected in the approximately first 20 cm of the inland soils. The Fe^{3+} relative fractions

determined from Mössbauer spectroscopy [Vandenberghe, 1991] and the ^{40}K distribution had quite similar behaviour. This correlation could be ascribed to the soil pedogenic and edaphic properties [Montes et al, 2010b], as well as to the presence of plant roots that use both ions as nutrients.

The other studied region of the Buenos Aires Province is located in the neighbourhood of the Centro Atómico Ezeiza [Valdés et al, 2011]. In this case the monitoring dealt with surface samples and ^{238}U, ^{232}Th and ^{40}K activities were determined down to 10 cm (AS5- AS11) [Montes, 2011]. The activities, quoted in Table 2 and Fig. 3, ranged from 52 Bq/kg to 65 Bq/kg, 24 Bq/kg to 35 Bq/kg and from 470 Bq/kg to 644 Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively.

In a frame of a survey program to study the environmental radioactivity in the Brazilian State of Rio Grande do Norte (BS1), the average concentrations of ^{226}Ra (29.2 Bq/kg), ^{232}Th (47.8 Bq/kg) and ^{40}K (704 Bq/kg) in fifty-two soil samples down 20 cm in areas with homogeneous lithology of the eastern and central regions of this states were determined. These values were higher than the world average and consistent with the predominance of granites and other Precambrian igneous rocks in the region [Malanca et al., 1996].

In Uruguay, the surface (down to 5 cm) ^{40}K activity values (US1-US8) ranged from 89.9 Bq/kg up to 1054 Bq/kg while activity values of ^{226}Ra and ^{232}Th were from 7.2 Bq/kg to 23.2 Bq/kg and 5.5 Bq/kg to 75.4 Bq/kg, respectively [Odino Moure, 2010].

The data of the all determined surface activity concentrations are compiled in Table 2 and Fig. 3 together with the worldwide average data reported by the UNSCEAR [UNSCEAR, 2008]. The mean and range worldwide values have been included by completeness [UNSCEAR, 2000; UNSCEAR, 2008]. It is clear that the reported data for ^{238}U for Brazil and Argentina are higher than the worldwide mean values. The observed ^{232}Th activities of Argentina are close to the worldwide mean values, while the Brazilian ones are quite higher than the worldwide average values. Due to the scattering and the scarcity of the data of Uruguay, it is not possible yet to extract a general conclusion. Finally, the ^{40}K data are higher than the mean values in most of the cases, and fit into the worldwide range with some exceptions.

Location	*Code*	^{238}U	^{232}Th	^{40}K	^{137}Cs	*Reference*
Argentina						
34°54.45' S; 58° 8.37' W	AS1	P	P	P	P	Montes et al., 2010b
35° 3.26' S; 57°51.21' W	AS2	P	P	P	P	
34°54.14' S; 57°55.10' W	AS3	P	P	P	P	
34°48.46' S; 58° 5.25' W	AS4	P	P	P	P	
34°48.08' S; 58° 5.04' W	AS5	S	S	S	S	Valdés, M. E. et al., 2011 Montes, 2011
35° 0.70' S; 57°44.29' W	AS6	S	S	S	S	
34°57.85'S; 57°45.66' W	AS7	S	S	S	S	
34°49.67' S; 58°35.14 W	AS8	S	S	S	S	

Location	*Code*	^{238}U	^{232}Th	^{40}K	^{137}Cs	*Reference*
34°49.30' S; 58°35.14' W	AS9	S	S	S	S	
34°50.69' S; 58°34.73' W	AS10	S	S	S	S	
34°50.46' S; 58°45.13' W	AS11	S	S	S	S	
33° 50.00′ S; 59°52.00′ W	AS12				P	Bujan et al., 2000; 2003;
33° 50.00′ S; 59°52.00′ W	AS13				P	
33° 50.00′ S; 59°52.00 W	AS14				P	
33°50.00′ S; 59°52.00′ W	AS15				P	
33°40.17' S; 65°23.45' W	AS16				P	Jury Ayub et al., 2007
33°40.17' S; 65°23.45' W	AS17				P	
33°40.17' S; 65°23.45' W	AS18				P	
33°40.17' S; 65°23.45' W	AS19				P	
33°39.93' S; 65°23.27' W	AS20				P	
33°39.93' S; 65°23.27' W	AS21				P	
33°39.93' S; 65°23.27' W	AS22				P	
33°40.17' S; 65°23.45' W	AS23	P	P	P	P	Jury Ayub et al., 2008
33°39.93' S; 65°23.27' W	AS24	P	P	P	P	
UNSCEAR	UN			S		UNSCEAR, 2008
Brazil						
Rio Grande do Norte	BS1	S	S	S		Malanca et al, 1996
22°42.00′ S; 47°38.00′ W	BS2				P	Corrochel et al, 2005
22°47.00′ S; 47°19.00′ W	BS3				P	
22°09.00′S; 47°01.00′ W	BS4				P	
22°40.00′ S; 48°10.00′ W	BS5				P	

Location	*Code*	^{238}U	^{232}Th	^{40}K	^{137}Cs	*Reference*
01°57.00′ S; 54°12.00′ W	BS6				P	
03°08.00′ S; 60°01.00′ W	BS7				P	
03°08.00′ S; 60°01.00′ W	BS8				P	
08°10.00′ S; 34°54.00′ W	BS9				P	
09°26.00′ S; 38°08.00′ W	BS10				P	
09°26.00′ S; 38°08.00′ W	BS11				P	
15°58.00′ S; 47°59.00′ W	BS12				P	
16°42.00′ S; 47°40.00′ W	BS13				P	
19°29.00′ S; 57°25.00′ W	BS14				P	
20°43.00′ S; 54°31.00′ W	BS15				P	Handl et al, 2008
20°22.00′ S; 43°24.00′ W	BS16				P	
20°21.00′ S; 43°29.00′ W	BS17				P	
22°20.00′ S; 43°37.00′ W	BS18				P	
22°30.00′ S; 44°30.00′ W	BS19				P	Handl et al, 2008
22°20.00′ S; 44°40.00′ W	BS20				P	
23°10.00′ S; 44°11.00′ W	BS21				P	
23°07.00′ S; 44°10.00′ W	BS22				P	
25°17.00′ S; 48°55.00′ W	BS23				P	
26°39.00′ S; 48°41.00′ W	BS24				P	
29°21.00′ S; 50°51.00′ W	BS25				P	
30°05.00′ S; 51°36.00′ W	BS26				P	

Location	*Code*	^{238}U	^{232}Th	^{40}K	^{137}Cs	*Reference*
Uruguay						
32°26.00' S; 54°19.00' W	US1	S			S	Odino Moure, 2010
31°18.00' S; 57°02.00' W	US2	S			S	
34°20.00' S; 56°43.00' W	US3	S			S	
34°10.00' S; 57°41.00' W (2004)	US4	S			S	
34°10.00' S; 57°41.00' W (2005)	US5	S			S	
34°10.00' S; 57°41.00' W (2006)	US6	S			S	
34°10.00' S; 57°41.00' W (2007)	US7	S			S	
34°10.00' S; 57°41.00' W (2009)	US8	S			S	
Venezuela						
Guaña	VS1				S	Sajó-Bohus et al, 1999
Chile						
39º44.00′ S; 73º22.80′ W	CS1				P	Schuller et al., 1997
38º41.50′ S; 72º53.00′ W	CS2				P	
39º41.30′ S; 72º57.10′ W	CS3				P	
40º23.00′ S; 72º57.50′ W	CS4				P	
1	CS5				P	Schuller et al., 2002
2	CS6				P	
3	CS7				P	
4	CS8				P	
5	CS9				P	
6	CS10				P	
7	CS11				P	
8	CS12				P	
9	CS13				P	
10	CS14				P	
11	CS15				P	
12	CS16				P	
13	CS17				P	

Location	*Code*	^{238}U	^{232}Th	^{40}K	^{137}Cs	*Reference*
14	CS18				P	
15	CS19				P	
16	CS20				P	
17	CS21				P	
18	CS22				P	
19	CS23				P	
20	CS24				P	
21	CS25				P	
22	CS26				P	
23	CS27				P	
24	CS28				P	
25	CS29				P	
26	CS30				P	
27	CS31				P	
28	CS32				P	
29	CS33				P	
50º53.00′ S; 72º40.00′ W	CS34				P	
51º08.00′ S; 53º10.00′ W	CS35				P	
51º10.00′ S; 73º05.00′ W	CS36				P	
51º12.00′ S; 73º00.00′ W	CS37				P	
52º20.00′ S; 68º25.00′ W	CS38				P	
52º16.00′ S; 68º50.00′ W	CS39				P	
52º35.00′ S; 69º50.00′ W	CS40				P	
52º38.00′ S; 70º15.00′ W	CS41				P	
52º40.00′ S; 70º50.00′ W	CS42				P	
52º25.00′ S; 71º25.00′ W	CS43				P	
52º35.00′ S; 71º33.00′ W	CS44				P	
51º55.00′ S; 72º00.00′ W	CS45				P	
52º35.00′ S; 71º42.00′ W	CS46				P	
53º36.00′ S; 70º50.00′ W	CS47				P	

Schuller et al., 2004

Table 1. Location, type of survey and monitored nuclides in soils of South America. S: surface activity determinations and P: profile activity determination.

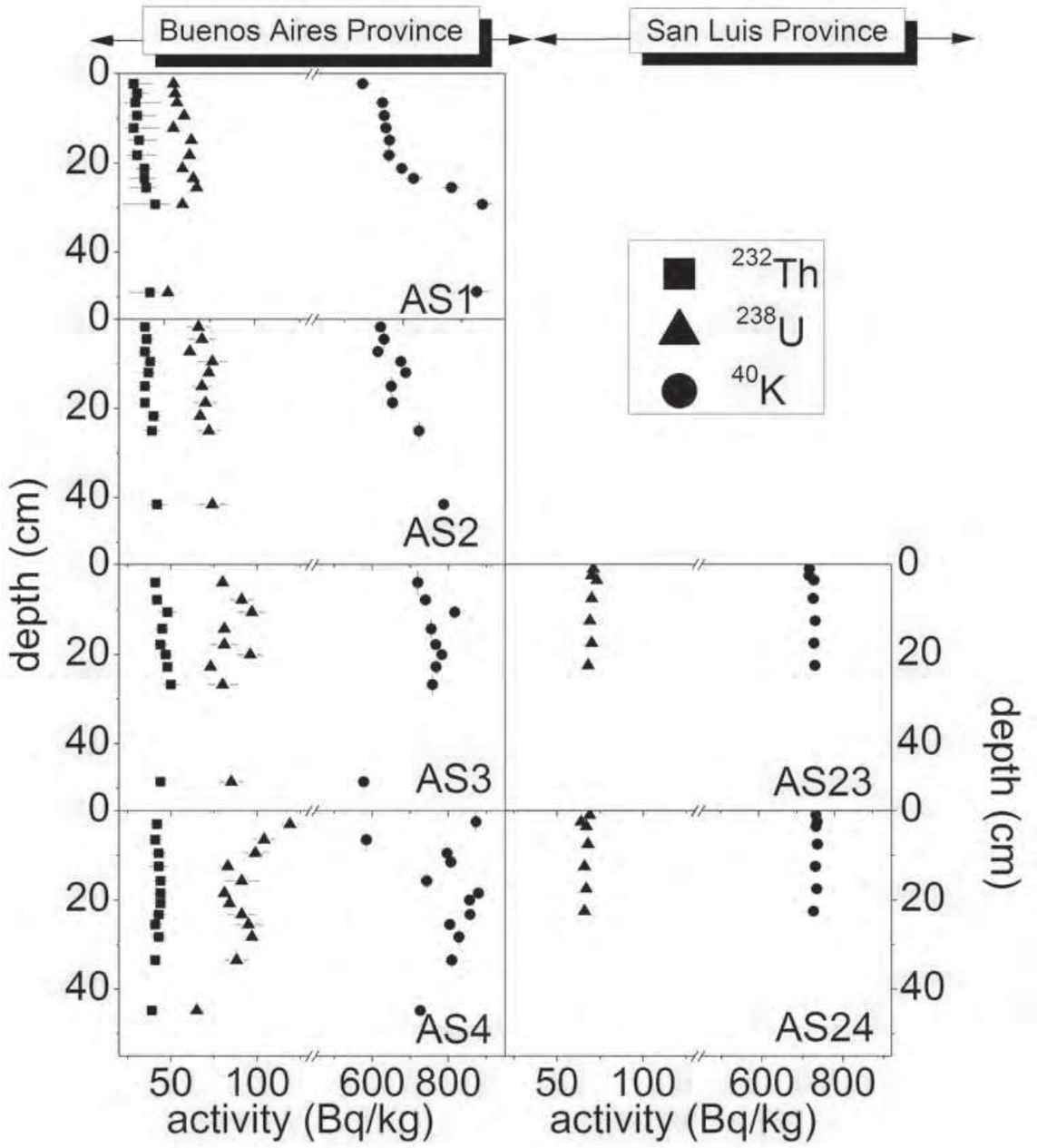

Fig. 2. Depth distribution activity of ^{232}The, ^{238}U and ^{40}K in Argentina, labelled with the sample code.

code	^{238}U *(Bq/kg)*	^{232}Th *(Bq/kg)*	^{40}K *(Bq/kg)*	*Code*	^{238}U *(Bq/kg)*	^{232}Th *(Bq/kg)*	^{40}K *(Bq/kg)*
AS1	55±6	33±4	531±13	BS1	10-136.7	12-191	56-1972
AS2	66±7	35±4	622±15	US1	19±2	75±7	1054±100
AS3	80±4	41±2	720±14	US2	7.2±0.5	11±1	90±5
AS4	119±5	42±4	717±15	US3	23±2	51±5	440±40
AS5	106±10	43±4	873±18	US4	19±2	8.6±0.5	492±45
AS6	61±8	35±2	576±15	US5	22±2	36±31	560±51
AS7	52±9	30±4	658±17	US6	21±2	35±30	495±45
AS8	65±18	35±17	644±29	US7	7.7±0.5	9.4±0.5	255±21
AS9	57±11	27±16	498±25	US8	14±1	19±2	340±31
AS10	53±13	24±12	470±22	**WA**	**35**	**30**	**400**
AS11	52±10	32±7	547±23	**WR**	**16-110**	**11-64**	**140-850**
AS23	71±4	-	733±11				
AS24	69±4	-	734±20				
UN	-	-	540-750				

Table 2. Natural surface activity concentrations in soils. WA: worldwide average and WR: worldwide range [UNSCEAR, 2000; UNSCEAR, 2008].

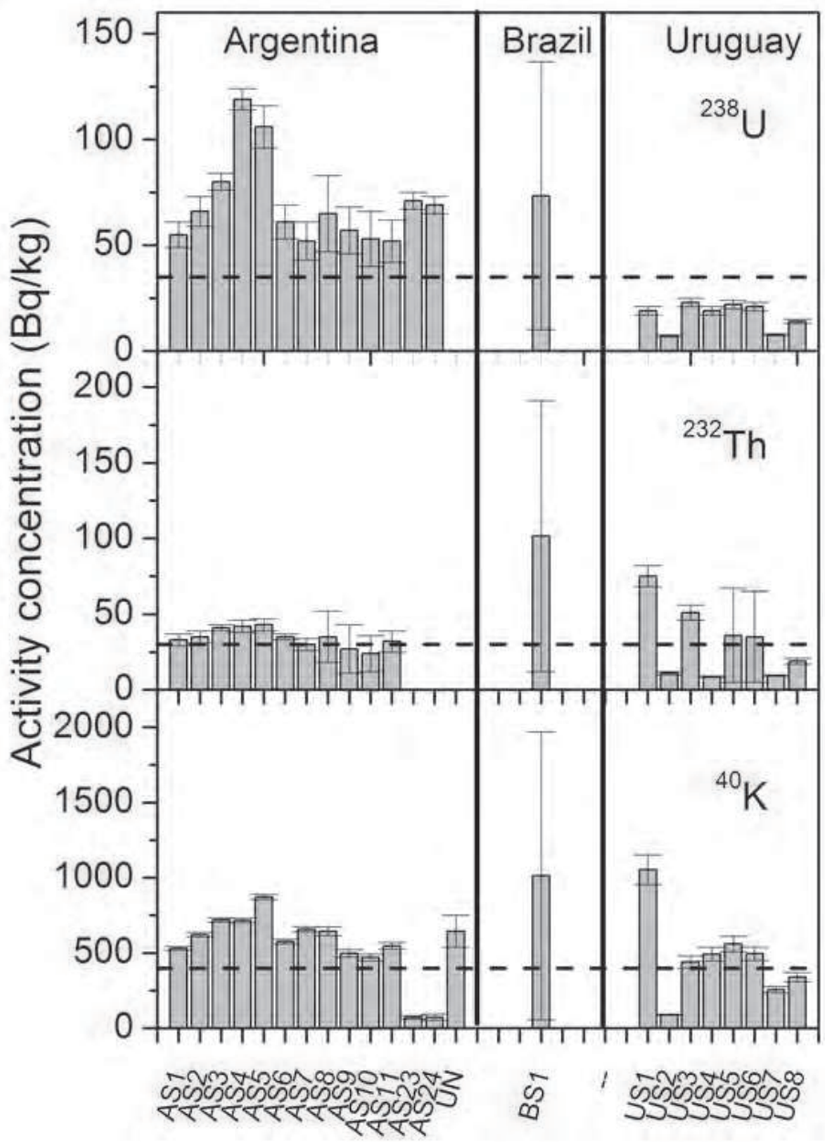

Fig. 3. Natural activity of ^{40}K, ^{232}Th and ^{238}U in soil surface. The dash line corresponds to the UNSCEAR average worldwide values [UNSCEAR, 2000]. The vertical bars correspond to the experimental errors in the case of Argentina and Uruguay, and to the standard deviation of a set of determinations in the case of Brazil.

3.2 Annual committed effective dose by external irradiation calculations

The contribution of natural nuclides to the absorbed dose rate at 1m above the ground depends on the concentration of radionuclides in the soil. There is a direct relationship between terrestrial gamma radiation dose and radionuclide natural concentrations in soils. The exposure dose rate can be evaluated accounting for the activity values of the nuclides (A_i) and the conversion factors (f_i). These coefficients are reported in Table 3 [UNSCEAR, 2000; UNSCEAR, 2008]. Based in the analysis of the UNSCEAR 1982 report [UNSCEAR, 1982], the International Committee of Radiation Protection (ICRP) used a coefficient (C_i) to

convert the absorbed dose in air to annual committed effective dose (*aced*). Monte Carlo Calculations radiation- transport codes indicate that higher values should be used for infant and children. These values are quoted in the Table 3. To calculate the annual effective dose it has also considered that the spent time outdoors is 20% of total time [USNCEAR, 2008], i.e.:

$$\text{aced (Sv)} = 10^{-9} \times 24 \times 365 \times C_c \times 0.2 \times \sum_i f_i A_i C_i \quad (1)$$

nuclide	f_i *(nGyh^{-1}/Bqkg^{-1})*	C_i *(Sv/Gy)*		
		infants	children	adults
^{40}K	0.0417	0.926	0.803	0.709
^{232}Th	0.604	0.907	0.798	0.695
^{238}U	0.462	0.899	0.766	0.672
Average		0.91	0.79	0.69

Table 3. Conversion factors (f_i) and absorbed dose to effective dose equivalent conversion coefficients (C_i) [UNSCEAR 2008, UNSCEAR 2000].

	aced (mSv)			*aced (mSv)*			
code	*infants*	*children*	*adults*	*code*	*infants*	*children*	*adults*
AS1	0.108±0.006	0.093±0.005	0.082±0.005	BS1	0.03-0.42	0.02-0.36	0.02-0.32
AS2	0.134±0.007	0.107±0.006	0.094±0.005	US1	0.16±0.01	0.137±0.009	0.120±0.008
AS3	0.146±0.004	0.126±0.003	0.111±0.003	US2	0.022±0.001	0.019±0.001	0.0667±0.0009
AS4	0.175±0.006	0.151±0.005	0.133±0.004	US3	0.095±0.006	0.083±0.005	0.073±0.005
AS5	0.177±0.009	0.153±0.007	0.135±0.007	US4	0.055±0.004	0.048±0.003	0.042±0.003
AS6	0.117±0.007	0.101±0.006	0.089±0.005	US5	0.09±0.03	0.08±0.03	0.07±0.03
AS7	0.111±0.008	0.096±0.007	0.084±0.006	US6	0.08±0.03	0.07±0.03	0.06±0.02
AS8	0.12±0.02	0.11±0.02	0.09±0.02	US7	0.032±0.002	0.028±0.002	0.024±0.001
AS9	0.10±0.02	0.09±0.02	0.08±0.02	US8	0.051±0.003	0.0447±0.003	0.039±0.003
AS10	0.09±0.02	0.08±0.01	0.07±0.01				
AS11	0.11±0.01	0.091±0.009	0.080±0.008				

Table 4. Calculated annual committed effective terrestrial exposure dose for infants, children and adults.

It is worth to mention that in the case of adults, the calculated annual committed effective doses due to terrestrial external exposure resulted slightly higher than the UNSCEAR reported values [UNSCEAR, 2000], as observed in Fig. 4.

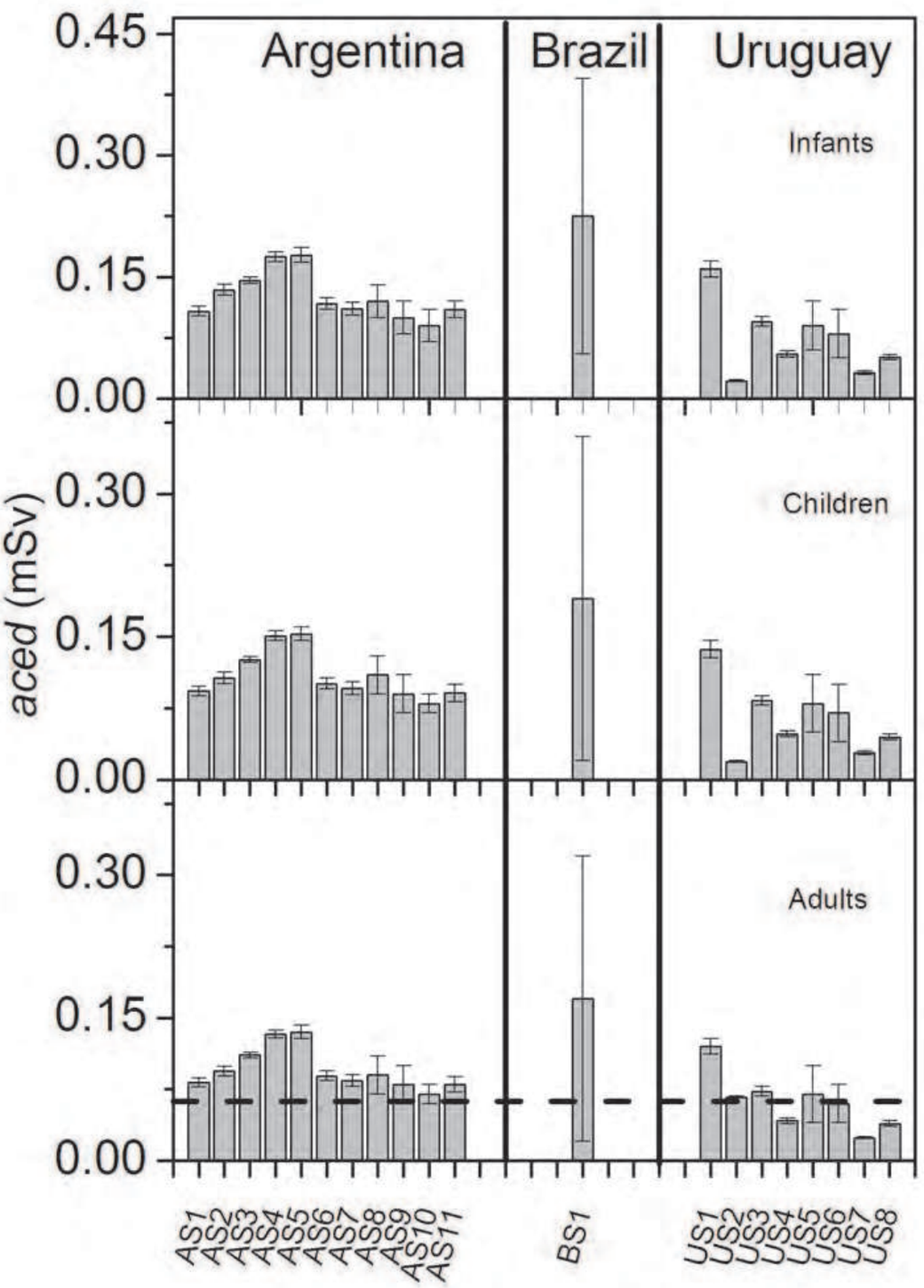

Fig. 4. Calculated annual committed effective dose for infants, children and adults. The dash line corresponds to the UNSCEAR reported values [UNSCEAR, 2000]. The vertical bars correspond to the experimental errors in the case of Argentina and Uruguay, and to the standard deviation of a set of determinations in the case of Brazil.

3.3 Anthropogenic nuclides

In the last decades, ^{137}Cs was the only monitored anthropogenic nuclide (gamma emitter) in the Southern Hemisphere. Aimed in the study of soil erosion, ^{137}Cs reference activity profiles (Fig. 5) were determined in the Pampa Ondulada of the Buenos Aires Province region, Argentina (AS12-AS15) [Bujan et al., 2000; Bujan et al., 2003]. The ^{137}Cs activities determined down to 90 cm declined sharply from the surface to the first 20 cm, the maxima activity was observed at the top layer. An average value of 1108 Bq/m^2 was obtained for the local inventory. In the La Plata city region, in spite that the ^{137}Cs integrated activities of the profiles obtained down to 50 cm were similar in all soils, differences in the ^{137}Cs depth distributions were detected (Fig. 5). The profiles of AS1 and AS3 sites followed a Gaussian-type feature, typical of a convective-diffusive process [Likar et al., 2001; Bossen & Kirchner, 2004]. The profile of AS2 was quite different since a Gaussian-shape was established down to 7 cm in depth. In the case of the AS4 soil, placed at 5 km from the La Plata river coast, the activity values were high at the surface and then suddenly decreased. Both facts, the high values at the surface and the deviation from the Gaussian shape [Likar et al., 2001;Bossen & Kirchner, 2004], could be explained considering the fine texture and the flat relief of the region which induce water-logging, i.e., this area shows a low permeability of the underlying horizons, and the phreatic water affects the deepest horizons [Imbellone, 2009]. It has been claimed that Cs is sorbed by Fe_3O_4 [Singh et al., 2009; Catallette et al., 1998; Marnier & Fromage, 2000]. However, by comparing the ^{137}Cs profiles and the Mössbauer relative fraction of Fe_3O_4 as well as with the other iron species [Montes et al.; 2010b], it was not observed an apparent correlation. A series of surface studies were also performed in the Buenos Aires Province in the neighbourhood of the Centro Atómico Ezeiza (AS5-AS11) showing that the activity concentration values down to 10 cm ranged between 0.9 Bq/kg and 2.6 Bq/kg [Vadés et al., 2011]. These values are consistent with the top layer activity data obtained from the profiles AS1-AS4 [Montes et al.; 2010b]. Vertical migration of ^{137}Cs was studied in soils of natural and semi-natural grassland areas of San Luis Province (AS16- AS21, AS23 and AS24) [Juri Ayub et al., 2007; Juri Ayub et al., 2008]. The inventories ranged from 330 Bq/m^2 to 730 Bq/m^2, while depth profiles had different shapes (see Fig. 5).

As observed in Fig. 6, differences in the patterns of ^{137}Cs depth distribution in the soil profiles of the different regions were found in the four studied sites of the South-Central region of Brazil (BS2-BS5), ascribed to chemical, physical, mineralogical and biological differences of the soils [Correchel et al., 2005]. The variability of the soil characteristics was not able to explain the spatial variability of the profiles. The average inventories of the four studied sites were 268 Bq/m^2, and the maximum activity value was detected at the top layer. The spatial distribution and behaviour of the ^{137}Cs in tropical, subtropical and equatorial unperturbed Brazilian soils have been investigated up to 40 cm (BS6-BS26) [Handl et al., 2008]. The shape of all 23 sampled sites depth profiles varied between the two ones showed in Fig. 6. The majority of the Cs content was observed in the 10-15 cm top layer while minor quantities were detected down to 35 cm. Low deposition densities were observed at the Amazon region where ascendant convection of water vapour is intense, while the south area exhibited considerable large concentrations. No correlation was observed between altitude and ^{137}Cs concentration. On the contrary, the results were correlated with the climatic de Martonne index, suggesting that the process can not be explained with single meteorological parameters [Handl et al., 2008].

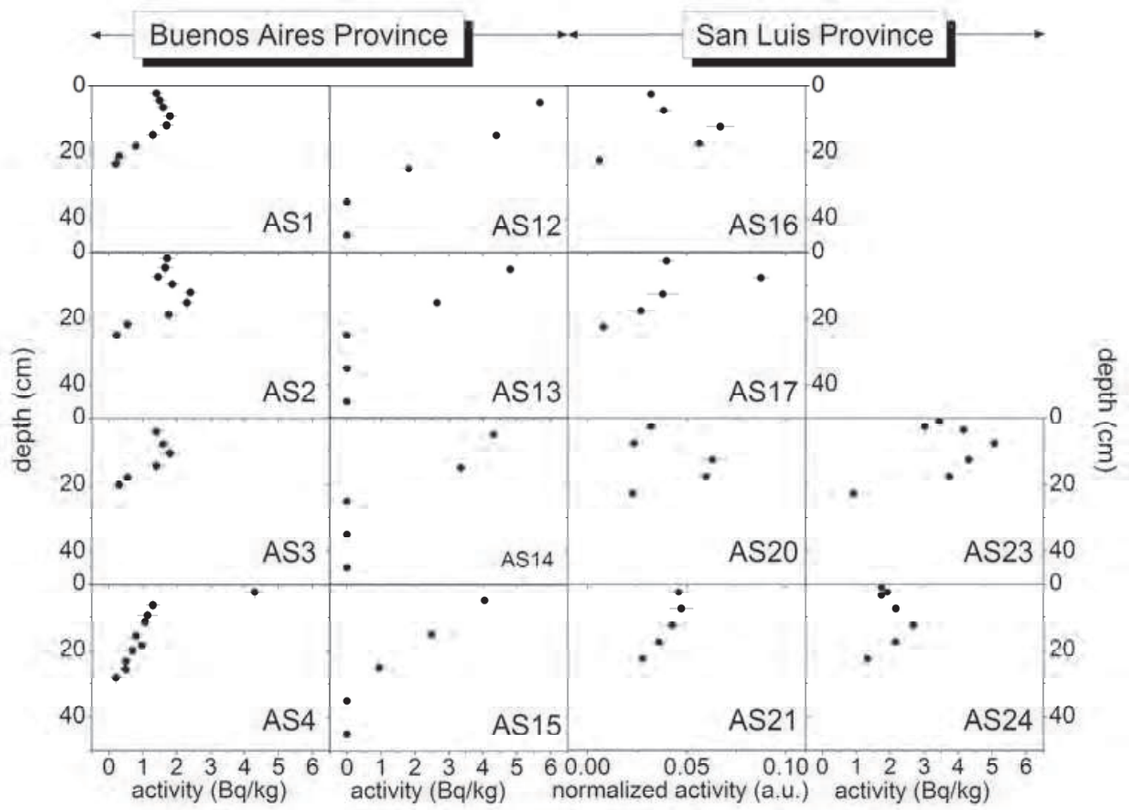

Fig. 5. ^{137}Cs depth profiles recorded in Argentina, labelled with the site code.

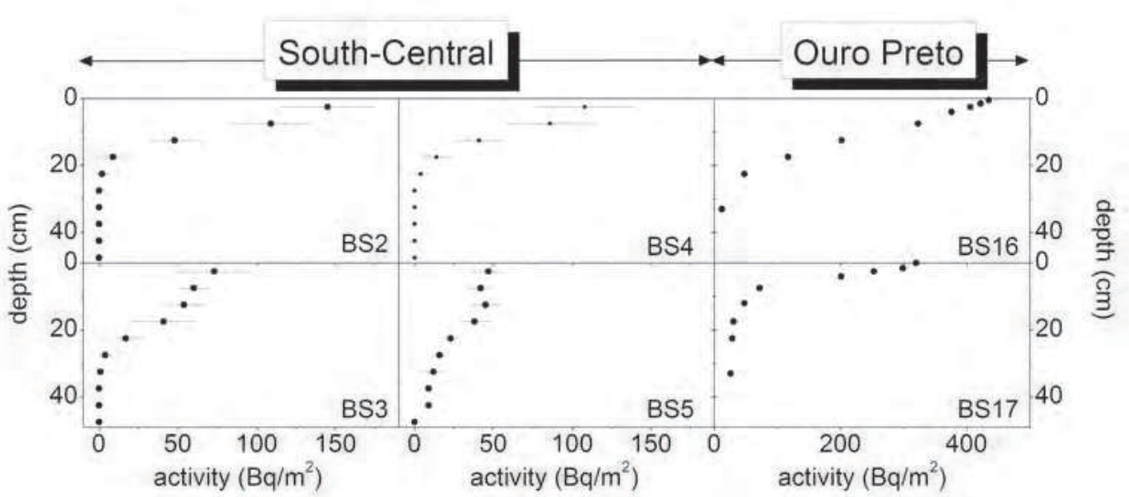

Fig. 6. ^{137}Cs depth profiles recorded in Brazil, labelled with the site code.

In Chile, total inventories and depth distributions of ^{137}Cs (Fig. 7) were determined at four sites of two agriculturally used soil types (CS1-CS4). The inventories were always higher than previously estimated for the Southern Hemisphere and depend on annual rainfall [Schuller et al, (1997)]. The depth distribution of ^{137}Cs in well-developed agricultural soil at 28 sites in different southern regions (CS5-CS33) was also studied [Schuller et al, 2002]. The profiles in most of the sites followed no systematic pattern in the upper few centimetres (Fig. 7), but below this depth an exponential behaviour was observed. The calculated relaxation depth [Schuller et al, 2002] ranged from 4.4 cm in Palehumults to 8.4 cm and 9.7 cm in Hapludands and Psamments soil types, respectively. The relaxation depth increased with decreasing clay content and increasing volume of coarse pores. Activity densities ranged from 450 Bq/m^2 to 5410 Bq/m^2, correlating with the mean annual rainfall rate of the sampling sites. The South Patagonia sheep-farming region (CS34-CS47) was studied (Fig. 7). The areal activity density varied from 222 Bq/m^2 to 858 Bq/m^2, positively correlated with the mean annual precipitation rate [Schuller et al., 2004].
In Venezuela, the ^{137}Cs concentration at two different depths, 0 cm - 20 cm and 20 cm – 40 cm, (VS1) was measured being the activity concentration around 0.5 Bq/kg and 10 Bq/kg at 20 cm in depth for the littoral and central regions, respectively [Sajó-Bohus et al., 1999].
Finally, the obtained values of ^{137}Cs surface activity concentration in different Uruguayan departments varied from 1.2 Bq/kg to 2.3 Bq/kg in the 2004 to 2009 period (US1-US8) [Odino Moure, 2010]. A spatial variation was also observed.

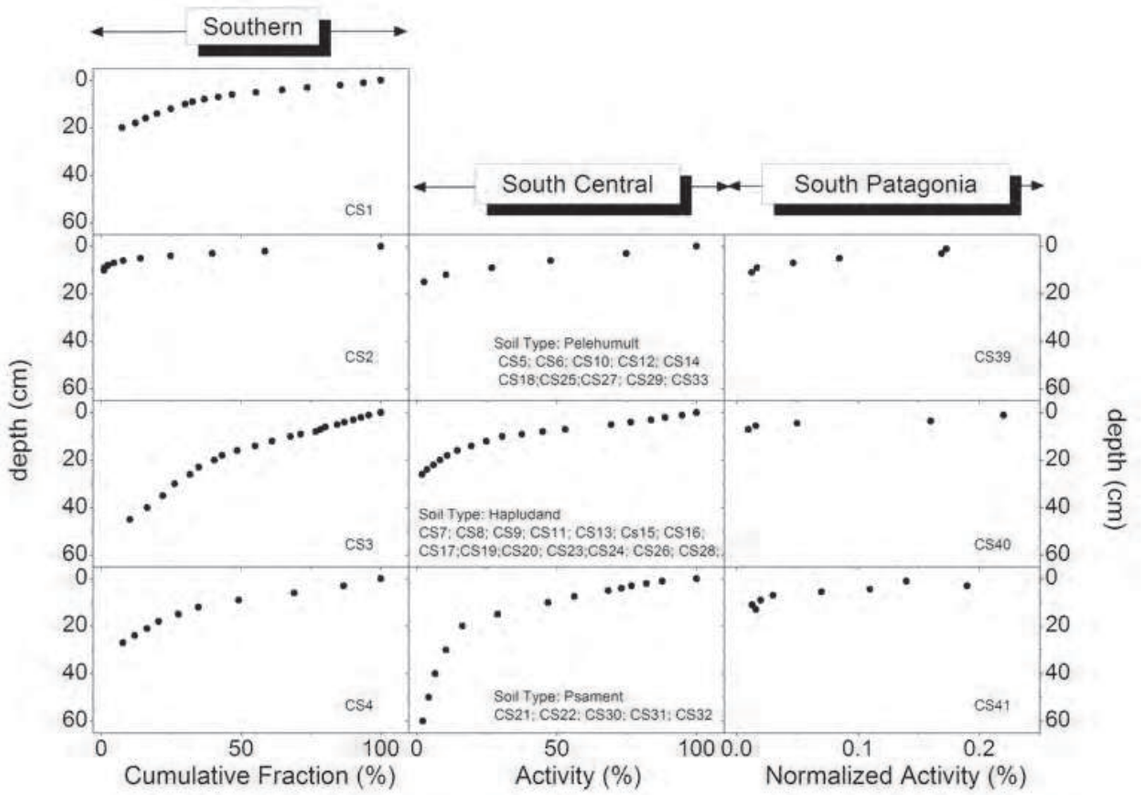

Fig. 7. ^{137}Cs depth profiles recorded in Chile, labelled with the site code.

3.4 Cs inventories analysis

The cumulated annual deposition of ^{90}Sr was compiled by UNSCEAR [UNSCEAR 2000; UNSCEAR 2008]. It is worth to mention that data of ^{137}Cs are not available due to the technological limitations on the detection of gamma emitters of the survey period. However, there is experimental evidence that the $^{90}Sr/^{137}Cs$ activity release ratio is constant and equal to 1.5, allowing using the global determination of ^{90}Sr to estimate the ^{137}Cs one. Through it should be considered that once the nuclides are incorporated in the soil, the migration rates are different due to the dissimilar soil-nuclide interaction process. Since the wind circulation is presented in latitudinal bands, this is the assumption used to evaluate the transport and deposition of nuclides [UNSCEAR, 1982; UNSCEAR, 2000; UNSCEAR, 2008].

In order to compare the inventory data with the UNSCEAR predictions [UNSCEAR, 2000; UNSCEAR, 2008], in the Table 5 are presented the inventory of ^{137}Cs data corrected by nuclide decay using the time of determination (a single input at 1965 is considered). It is observed that the experimental data do not follow the UNSCEAR prediction. The Fig. 8 shows the inventories of ^{137}Cs, the average annual precipitation vs. latitude and the inventory vs. precipitation. Globally, it seems that the inventory depends on the annual precipitation and the Andes Cordillera plays a very important role on the inventory due to the generation of higher annual precipitations, hence more ^{137}Cs deposition. It is also observed that mountains act as barrier for Argentina.

Code	*^{137}Cs inventory (Bq/m^2)*	*Latitudinal band (degree)*	*Integrated deposit (Bq/m^2)*
BS6	945±110	0-10	720
BS7	0.8±0.1		
BS8	5.15±0.07		
BS9	99±13		
BS10	83±11		
BS11	188±27		
BS12	558±71	10-20	630
BS13	654±212		
BS14	16±2		
BS17	1120±76	20-30	1050
BS16	596±143		
BS15	1691±216		
BS4	621±46		
BS20	3494±477		
BS18	1333±79		
BS19	479±61		
BS5	594±37		
BS2	771±34		
BS3	614±77		

Code	*^{137}Cs inventory (Bq/m^2)*	*Latitudinal band (degree)*	*Integrated deposit (Bq/m^2)*
BS22	1355±173		
BS21	1407±180		
BS23	1375±176		
BS24	320±41		
BS25	3629±463		
BS26	1344±172	30-40	1140
AS20	1877		
AS21	1285		
AS22	848		
AS16	1311		
AS17	1645		
AS18	1427		
AS19	745		
AS12	2512		
AS13	2041		
AS14	2350		
AS15	2050		
AS4	689±36		
AS3	849±38		
AS1	950±34		
AS2	1366±38		
CS2	1438		
CS3	2821		
CS1	2202		
CS5	1329		
CS6	910		
CS7	874		
CS9	110		
CS13	1565		
CS16	1511		
CS24	2111		
CS25	2020		
CS26	2584		
CS27	2894		
CS28	1893		

Code	*^{137}Cs inventory (Bq/m^2)*	*Latitudinal band (degree)*	*Integrated deposit (Bq/m^2)*
CS29	3913		
CS4	1420	40-50	1335
CS8	819		
CS10	1128		
CS11	1292		
CS12	1492		
CS14	1165		
CS15	1438		
CS17	892		
CS18	2002		
CS19	1238		
CS20	2038		
CS21	3130		
CS22	1711		
CS23	1674		
CS30	2621		
CS31	9045		
CS32	4641		
CS33	9846		
CS34	1162±89	50-60	705
CS35	1464±113		
CS36	1457±112		
CS37	1334±103		
CS45	796±61		
CS39	647±50		
CS38	630±48		
CS43	511±39		
CS40	527±41		
CS44	564±43		
CS46	991±76		
CS41	433±33		
CS42	546±42		
CS47	1673±129		

Table 5. Determined ^{137}Cs inventory together with the UNSCEAR predictions [UNSCEAR, 2008], ordered by latitudinal band.

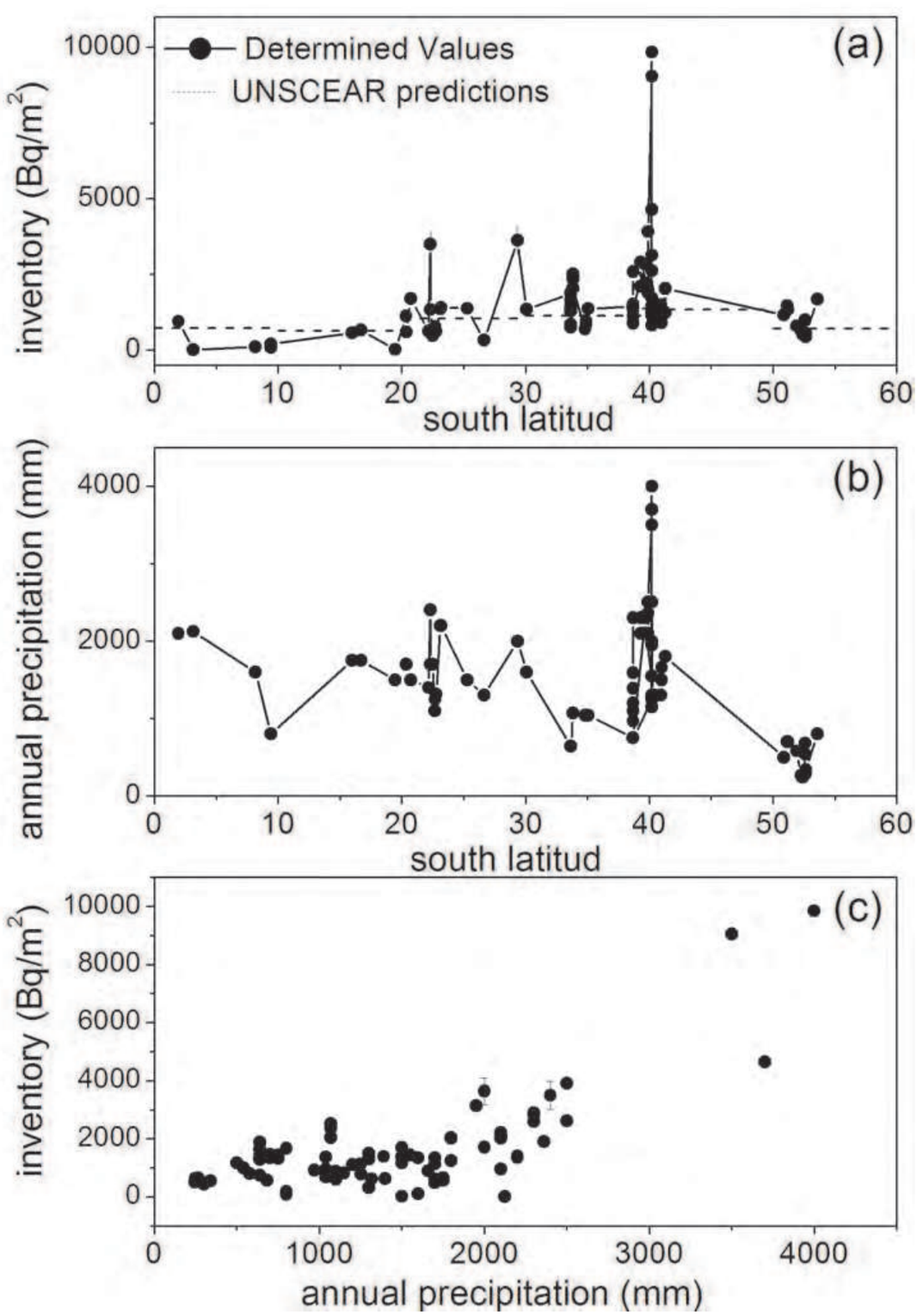

Fig. 8. a) ^{137}Cs inventory vs. annual precipitation; b) Annual precipitation vs. south latitude and c) ^{137}Cs inventory vs. annual precipitation.

3.5 Soil profile analysis

The basic processes controlling mobility of anthropogenic nuclides in soil include convective transport by water, dispersion caused by spatial variations of convection velocities, diffusive movement within the fluid, and physicochemical interaction with soil matrix. Because the slow migration velocities of Cs in soils, generally the models do not take into account the soil moisture changes in the unsaturated zone but assume mean constant water content. The spatial uniformity of the deposition rates is also considered which may be defensible for the weapon tests and, at least, on a local scale in nuclear accidents. In this frame, the two trendy for modelling the migration of radionuclides in soils are the one dimensional convection-dispersion equation (ODCDE) with constant parameters [Likar et al., 2001; Bossen & Kirchner, 2004] and the serial compartmental approach (CA) [Kirchner, 1998; Schuller et al., 1997].

The ODCDE model is based on the diffusive-convective transport, the mass conservation and the Cs-soil matrix interaction. The equation describing the migration process is usually known as the Fokker Planck equation:

$$\frac{\partial C(x,t)}{\partial t} = D_e \frac{\partial^2 C(x,t)}{\partial^2 x} - v_e \frac{\partial C(x,t)}{\partial x} - \lambda C(x,t) \tag{2}$$

where $C(x, t)$ is the ^{137}Cs concentration in the soil (mobile and sorbed), λ is the decay constant, D_e is the effective diffusion coefficient of caesium in soil, v_e is a convective velocity, x is the soil depth with respect to the soil surface and t is the time from the deposition. Assuming that all sorbed Cs is exchangeable and that the exchange process is in equilibrium, D_e is an effective coefficient that depends on the porosity ε, the bulk density ρ and the solid aqueous partitioning coefficient K_d:

$$D_e = \frac{D_w}{\left(1 + \rho K_d / \varepsilon\right)} \qquad v_e = \frac{v_w}{\left(1 + \rho K_d / \varepsilon\right)} \tag{3}$$

and where D_w *and* v_w are the diffusion coefficient of Cs in soil water and the convection velocity of water in pores of soil, respectively. The factor between parentheses in eqs. 3 is called the retardation factor. As boundary conditions, a half-infinite space-time is assumed and the considered initial condition is a pulse-like deposit at t=0 with deposition density J_0. With all these assumptions, it is obtained the well known solution:

$$. C(x,t) = J_0 e^{-\lambda t}\left[\frac{1}{\sqrt{\pi D_e t}} e^{-(x-v_e t)^2 / 4D_e t} - \frac{v_e}{2D_e} e^{-v_e / D_e} erfc\left(\frac{v_e}{2}\sqrt{\frac{t}{D_e} + \frac{x}{2\sqrt{D_e t}}}\right)\right]. \tag{4}$$

The irreversible fixation of Cs to soil has been also accounted for, however in this case, no analytical solution of the transport equations are able to obtain, so numerical methods are needed to obtain the concentration profiles [Antonopoulus-Domis et al., 1995; Toso & Velasco, 2001]. The fitting of experimental data with the ODCDE model allows determining D_e and v_e.

The CA has been used to analyse the depth layered profiles without detailed information about the site-specific processes that influence radionuclide´s mobility. Usually the soil profile is split into a series of horizontal layers (compartments) which are connected by

radionuclide downward transport rates and the migration dynamic is described by a system of lineal first-order differential equations with constant coefficients. This model is applicable only if the transport of the radionuclide is dominated by convection [Kirchner et al., 2009]. It is worth to mention that neither the presence of micro-organisms nor the root intake is considered by the models.

In the following, data compilation of the transport parameters of soils of South America is presented in Fig. 9. To facilitate the comparison, the worldwide average values corresponding to weapons fallout have been also included [IAEA, 2010]. In the La Plata city area of the Buenos Aires Province, the two profiles (AS1 and AS3) that clearly have a Gaussian shape were fitted using the ODCDE [Montes et al., 2010a; Montes et al., 2010b]. For the AS1, the D_e and v_e resulted equal to 0.728 cm²/y and 0.23 cm/y, respectively. while for AS3 the values were 0.5 cm²/y and 0.22 cm/y, correspondingly. Since in the case of profile of AS2 site, the Gaussian-shape was established down to 7 cm in depth, the data of the top layer were disregarded in the analysis leading to diffusion coefficient and convection velocity values of 0.39 cm²/y and 0.34 cm/y, respectively. These set of transport parameter values agrees well with the South American values [Juri Ayub et al., 2007; Juri Ayub et al., 2008; Schuller et al., 1997; Schuller et al., 2004], as observed in Fig. 6, but is slightly larger than the average values reported by IAEA [IAEA, 2010].

In the semi-natural and natural central area of Argentina (AS16-AS24), the diffusion coefficients obtained using the ODCDE varied from 0.43 cm²/y to 2.27 cm²/y, and the convection velocity varied from 0.13 cm/y to 0.39 cm/y. The D_e values were in the range reported in the bibliography for some Chilean and European soils, while the v_e values were one order of magnitude higher than those reported for Chilean soils and of the same order of magnitude than the European sandy ones [IAEA, 2010]. The great penetration in these soils was ascribed to the high sand and low fine materials content, i.e., high porosity facilitating water passage to deeper layers.

The Chilean Southern soil profiles of these soils were analysed using both, the CA and the ODCDE. The results of the fits were not good. In the case of the CA, the variation of the migration rates did not improve appreciably the fits. Moreover, the CS3 profile cannot be reproduced with this model. The determined migration rates resulted always low, between 0.1 cm/y and 0.3 cm/y, in the lower range of the reported data obtained for nuclear weapons and Chernobyl fallout [Schuller et al., 1997]. On the other side, the analysis of the data using the ODCDE indicated that the transport was dominated by diffusion process in agreement with the high silt and clay content of the studied soils. The agreement of D_e and v_e with reported data was better than in the CA case. However some misfit was observed at large depth, probably due to preferential transport through macropores, migration of suspended particles, spatial variability or agricultural land disturbance. Least-square fits of semi-natural and natural South Patagonia profiles using the ODCDE with constant parameters and improved by assuming a logarithmic distribution of D_e and v_e or depth dependence of both parameters were also tried. In all attempts, the mean obtained parameters were the same. The convection velocity was found to be negative (upward migration) in the CS43 site, because of the yearly flooded lowland, and at CS45 soil, where the upper soil layer was possibly disturbed by animal hoof prints. In the CS43 site, the D_e values were considerably larger than the determined for the other profiles. In the other sites, the determined median convection velocity and the diffusion coefficient values were 0.056 cm/y and 0.048 cm²/y, respectively. The

convection velocities resulted rather higher when compared with the data of temperate regions from Chile [Schuller et al., 1997], while the diffusion coefficient was close to those obtained in the Antarctic region [Schuller et al., 2002].

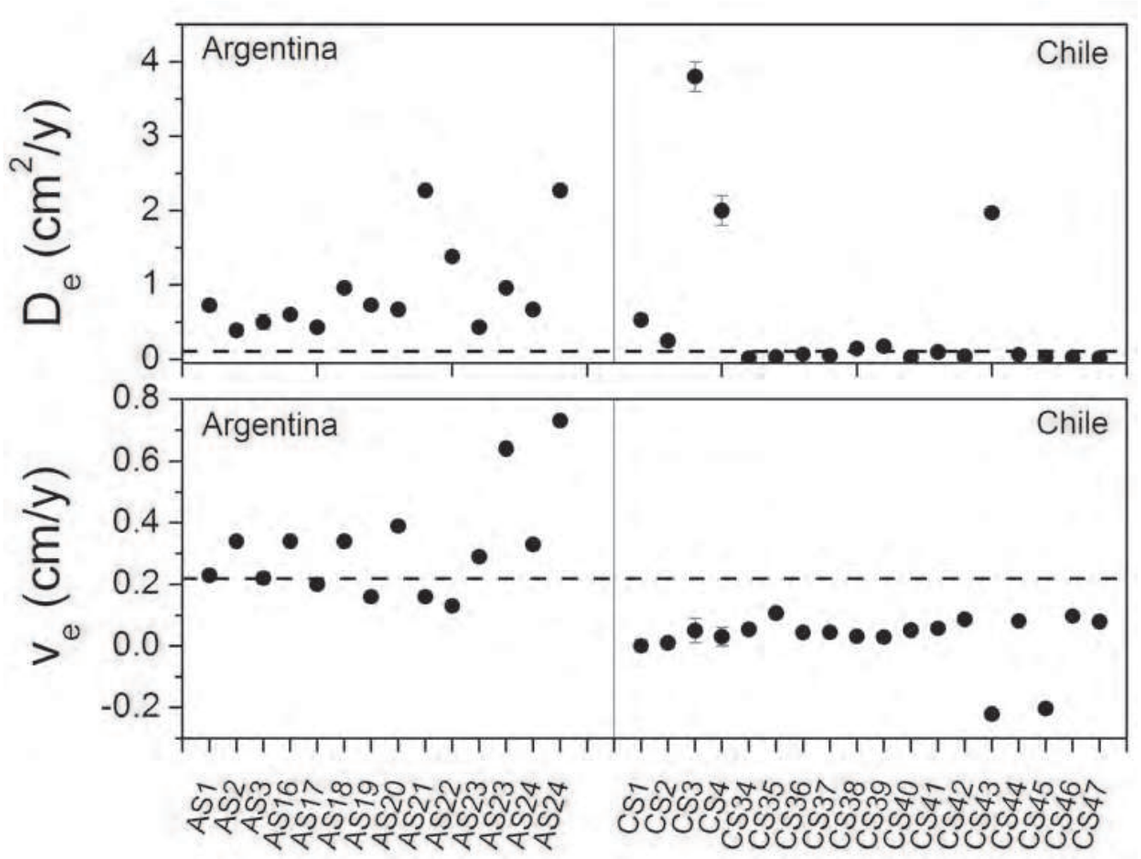

Fig. 9. Diffusion coefficient (D_e) and convection velocity (v_e) parameters together with the IAEA (dash line) [IAEA, 2010].

The present set of transport parameters of ^{137}Cs in soils is presented in Fig. 9. A scatter is observed in the data. Most of Argentinean diffusion coefficient data are slightly larger than the average values reported by IAEA for weapon test fallout [IAEA, 2010] while the Chilean data fit quite well with the average values with the exception of CS3, CS4 and CS43. Concerning the convection velocity parameter, the determined values from Provincia de Buenos Aires-Argentina soils resulted close to the worldwide average ones [IAEA, 2010], while those determined in the San Luis Province-Argentina are higher than the IAEA data [IAEA, 2010]. In Chile, the values were lower than the worldwide average [IAEA, 2010], being some of them negative, probably related to the periodic flooded lowland. Several mechanisms, such as bioturbation, horizontal transport, transport through macropores, migration of suspended particles, etc., have been used to explain the deviations from the convection-dispersion predictions, putting the ODCDE model under consideration and suggesting that the model is an oversimplification of such a complex process. However the ODCDE model is very useful to estimate the transport parameters.

4. Conclusions

A systematic compilation of radionuclide activity data in soil of South America has been completed. Radionuclide activity data concern to the natural ^{40}K, ^{238}U and ^{232}Th chains, and to the anthropogenic ^{137}Cs nuclides.

The surface activity concentrations for ^{238}U for Brazil and Argentina are higher that the worldwide mean values. The ^{232}Th activity data of Argentina are closer to the worldwide values while the Brazilian ones are quite higher than the worldwide values. In the case of Uruguay, it is not possible to extract conclusions yet due to the insufficiency and dispersion of data. The ^{40}K data are higher than the mean values in most of the cases, and fit into the worldwide range with some exceptions. The annual committed effective terrestrial exposure dose for infants, children and adults have been calculated, resulting the values slightly higher than the UNSCEAR value in the case of adults. The analysis of the ^{137}Cs inventories allows concluding that the experimental data do not follow the latitudinal band deposition predictions proposed by UNSCEAR. It is worth to mention that the analysis of the whole set of information in South America allows to establish a correlation between the inventory and the annual precipitations. Different shape type profiles have been determined for Argentina, Brazil and Chile. In several cases it was possible to reproduce the ^{137}Cs profiles with models accounting diffusion and convection process. The transport parameters agree well with the average worldwide values due to nuclear weapon test fallout. Some discrepancies were detected when bioturbation and floodedland are present, indicating that efforts to include these processes should be done to fully reproduce the caesium profiles, hence to be able to make predictions of migration in case of possible pollution.

The present set of data contributes to the establishment of regional baselines as well as help in the development of local regulations concerning to permitted activity limits to people health protection.

5. Acknowledgements

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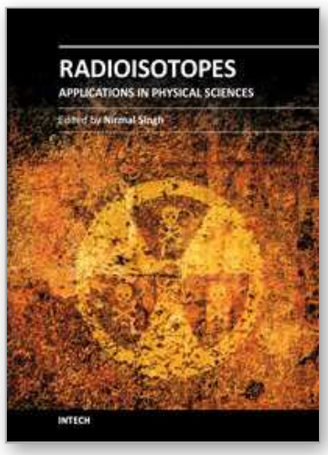

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The book Radioisotopes - Applications in Physical Sciences is divided into three sections namely: Radioisotopes and Some Physical Aspects, Radioisotopes in Environment and Radioisotopes in Power System Space Applications. Section I contains nine chapters on radioisotopes and production and their various applications in some physical and chemical processes. In Section II, ten chapters on the applications of radioisotopes in environment have been added. The interesting articles related to soil, water, environmental dosimetry/tracer and composition analyzer etc. are worth reading. Section III has three chapters on the use of radioisotopes in power systems which generate electrical power by converting heat released from the nuclear decay of radioactive isotopes. The system has to be flown in space for space exploration and radioisotopes can be a good alternative for heat-to-electrical energy conversion. The reader will very much benefit from the chapters presented in this section.

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