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Recent Evolution of the Multi-Isotopic Radioactive Content in Ice of Livingston Island, Antarctica

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The temporal arrangement of the ice layers that are produced in ecosystems with perpetual snows form situations that greatly favour the study of the temporal evolution of the radioactive fallout that occurs in the said zones, whether this fallout is natural or artificial in origin. This allows one to investigate the causes of the fallout and the mechanisms transporting the radionuclides involved from their source point to the study zone, as well as their subsequent behaviour in that zone. There are special difficulties involved in this type of study in Antarctica. Some are of a general character deriving from the conditions of extreme climate and isolation which complicate the processes of extraction and pre-treatment of the samples. Others are directly related to the detection of the temporal evolution of the radionuclides of interest, since these generally possess very low activity levels, so that one has to collect large volumes of samples and use refined techniques of extraction and selective radiochemical concentration prior to assay. In particular, the primordial radionuclides are usually detected in Antarctic ice at lower activity levels than are found for similar latitudes in the northern hemisphere (Nijampurkar and Rao 1993), mainly because of the greater proportion of land to sea in the northern hemisphere. With respect to the deposition of artificial radionuclides, save for those from the re-entry into the atmosphere of the SNAP-9A satellite and from the French atmospheric tests in the Mururoa Atoll between 1966 and 1974, the main events responsible for global artificial radioactive contamination occurred in the northern hemisphere (Kathren 1984), with the resulting observed progressive fall in artificial radioactivity, levels from northern to southern latitudes (Baeza et al. 1996, Holm et al. 1991).

In this sense, have have been carrying out a variety of studies aimed initially at quantifying the radioactive content of some of the most characteristic receptor media of the Antarctic ecosystem (Baeza et al. 1994a, Baeza et al. 1994b), specifically on Livingston Island of the South Shetland archipelago, site of the Spanish Antarctic Base, Juan Carlos I, and at determining the external irradiation dosimetry in the said zone. We subsequently studied the temporal evolution of the overall radioactive indices in the upper layers of the ice, down to a depth of 8.32 metres, extracted from the Hurd Glacier on Livingston Island. For this purpose we dated the ice samples using two procedures. On the one hand we used the the method of ^{210}Pb excess, in which one evaluates the disequilibrium between ^{226}Ra and its radioactive descendant ^{210}Pb, and on the other hand we used the depth of the volcanic ash deposits in the cited glacier as a consequence of the eruptions that took place on the 12 and 13 August, 1970, on the nearby Deception Island (Baker and McReath 1971). The two methods led to coincident values for the mean annual mass balance of the snow deposited

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Correspondence to: A. Baeza, Department of Physics, Faculty of Veterinary Science, Avda. de la Universidad, s/n, 10071 Cáceres, Spain in the zone of the glacier under study, 0.24 kg m^{-2} yr^{-1}, allowing us to deduce that the ice layers sampled were formed in the years between 1969 and 1993.

The objective of the present work is, on the basis of the aforementioned dating process, to contribute information concerning the temporal evolution undergone by the radioactive levels deposited on the ice of the Hurd Glacier during a relatively recent time period (1969-93), for two natural radionuclides (^{234}U, ^{238}U), a radionuclide of mixed natural/artificial origin (^{3}H): and artificial radionuclides (^{137}Cs ^{90}Sr, ^{338}Pu, $^{239+240}$Pu, and ^{241}Am).

MATERIALS AND METHODS

The zone chosen for sampling is in a flat elevated region at 333 m above sea level on the Hurd Glacier, Livingston Island. The GPS coordinates are: 62 40' 50.6416" S, 60 22' 07.1263" W. The collection of snow/ice samples was performed between December 1993 and February 1994. We took a total of 19 samples, with thicknesses between 0.4 and 0.5 m, down to a depth of 8.32 m (Baeza et al. 1996). The samples were transported to the laboratory of the Spanish Antarctic Base, were they were melted by heating to 80° C, followed by filtering through 0.45µm pore membranes prior to their radiochemical pre-treatment and/or separation.

For the determination of the ^{3}H concentration, the liquid samples were distilled in the presence of $KMnO_4$, in order to eliminate interfering salts, and then concentrated by electrolysis in a basic medium from an initial volume of 250 mL to a final volume of 50 mL. The resulting samples were neutralized with $PbCl_2$ and redistilled, this time without additives, to eliminate the salts which might produce inhomogeneities in the sample/scintillator mixture. The proportion used in the said mixture was 8/12. The samples were put into 20 mL capacity teflonated polyethylene vials and stored for 24 hours in cold and dark conditions before assay. The assay was performed using a low background liquid scintillation spectrometer, Quantalus 1220 from LKB, with 1000 minute counting times. Since the detection efficiency for the ^{3}H energy region in a sample with no extinction is 22%, and the background count rate is 1.2 c.p.m., with this procedure we obtained a detection limit of around 0.4 Bq/L (Baeza et al. 1997, Baeza et al. 1998).

The determination of the activities of ^{137}Cs, ^{234}U, 238U, ^{238}Pu, $^{239+240}$Pu and ^{241}Am was performed using 50 L of liquid sample for each of the profiles collected. These aliquots were conditioned with HNO_3 at pH=1, followed by addition of the corresponding tracers, ^{232}U, ^{232}Pu, and ^{243}Am, and the caesium carrier, CsCl. The mixtue was then stirred for 24 hr to guarantee ionic equilibrium. Following the procedure refined and described by Nakaoka et al. (1980), ammonium molybdophosphate, AMP, was then added, and the stirring was continued for at least one hour more. The AMP-Cs precipitate was allowed to form, and filtered out onto a 9 cm diameter cellulose filter for its later determination by gamma spectrometry. We determined the yield of the AMP-Cs extraction, which is close to 100%, by atomic absorption on the stable Cs content of a solution of an aliquot of the AMP-Cs precipitate in H_2SO_4 We then used the supernatant for the extraction by precipitation of the uranium and the transuranides with ferric hydroxide in a basic medium, filtering the precipitate obtained before its transport to Spain, The radiochemical separation continued by dissolving the precipitate in 8M HCl, extracting the iron with di-isopropyl ether (DIE). The plutonium valences were then adjusted (Gibb 1975), for its improved absorption in the ion-exchange column (Dowex 1x8). This column retains the

Table 1. Activity levels detected in the 19 ice layers, whose datings are specified, collected on the Hurd Glacier (Livingston Island, Antarctica) between 12/07/93 and 02/16/94.

SAMPLE	YEAR	^{3}H (Bq/L)	^{234}U (μBq/L)	^{238}U (μBq/L)	^{238}Pu (μBq/L)	$^{239+240}$Pu (μBq/L)	^{241}Am (μBq/L)
1	1993	0.99±0.22	850±130	780±120	3.2±0.8	30±4	12.1±1.5
2	1992	1.78±0.08	97±14	79±12	3.5±0.8	18.0±2.0	18.1±2.6
3	1991	0.48±0.15	67±9	61±8	4.6±0.6	19.4±1.4	21.3±1.8
4	1990	1.27±0.17	48±6	38±5	1.6±0.7	19±3	17.2±2.4
5	1989	1.05±0.07	140±30	131±24	NO DATA	NO DATA	NO DATA
6	1988	0.74±0.08	138±23	104±19	3.0±1.0	16±4	32±5
7	1987	1.02±0.10	58±7	44±6	NO DATA	NO DATA	NO DATA
8	1985	0.71±0.09	93±14	71±11	2.7±0.9	25±3	25±5
9	1984	0.91±0.11	620±90	760±110	4.4±1.1	18.0±2.3	59±14
10	1982	0.43±0.22	140±18	85±12	0.76±0.19	22±3	18.9±1.7
11	1981	0.63±0.13	560±100	320±70	10.7±2.4	25±4	53±8
12	1980	1.01±0.12	56±7	44±6	2.9±0.5	19.9±1.6	32.0±2.0
13	1978	<0.7 (*)	132±16	98±13	16±4	22±5	43±4
14	1977	<0.7 (*)	55±7	37±5	2.4±0.5	18.5±1.6	70±11
15	1975	0.76±0.22	33±5	12.0±1.7	11.2±2.0	55±5	21±4
16	1974	<1.0 (*)	59±9	48±8	2.1±0.6	25.3±2.2	46±15
17	1972	0.80±0.07	400±80	220±60	13±9	43±6	32±19
18	1970	<0.4 (*)	618±19	516±17	59±9	30±6	51±3
19	1969	1.14±0.10	600±30	488±25	16±3	45±5	35±4

(*) The detection limits are calculated for the sample measurement date. The net activities are corrected for decay to the sampling date.

plutonium and americium isotopes when the sample is dissolved in 1M HNO_3 and 90% CH_3OH (Crespo et al. 1992). The plutonium was recovered from the column using a solution of 10M HCl - 0.1M HI. Each of the three fractions were dried and conditioned for their respective electrodeposition onto stainless steel planchets (Hallstadius 1984). The chemical yields that were normally obtained in these extraction processes were approximately 75%, providing alpha spectrometry sources of 25-30 keV resolution, whose spectra were determined with silicon detectors of 450 mm active area and 18 keV nominal resolution.

To extract the strontium, we used the ion exchange method (Gascó and Alvarez 1988), starting from a volume of 15 L for each of the snow/ice layers to be assayed. This volume was pre-concentrated by evaporation to a final volume of 1 L, to which we added the corresponding stable strontium carrier in acid medium. Subsequently with the use of ethylenediaminetetraacetic acid, EDTA, and adjusting the solution to pH=4.8, we retained the Sr in a Dowex 50Wx8 Na^{+} cationic resin. The strontium was extracted from the resin by flushing with a NaCl solution, and precipitated onto striated stainless steel planchets in the form of $SrCO_3$ by adding Na_2CO_3 in a basic medium. Measurements were made in a proportional gas flow counter, with a zero thickness efficiency of 32.9% and an average background of 0.12 c.p.m.

RESULTS AND DISCUSSION

Table 1 lists the activity levels obtained for each of the radioisotopes assayed, corrected for decay to the date of sample collection, which was from 7 December 1993 for sample 1 to 16 February 1994 for sample 19. The second column of the table gives the year of formation of the respective snow/ice layer assigned to each sample, according to the previously performed dating using the two methods described above (Baeza et al. 1996). We then analyzed, for each of the systematically assayed radionuclides, both the values of the radioactive contaminations obtained and their temporal evolution.

From an observation of the tritium activity levels detected in the ice layers analyzed (see Table 1), one notes that they are distributed apparently at random in the interval (0.43. 1.78) Bq/L, with a mean above the detection limit of (0.91±0.34 (S.D)) Bq/L. These values are slightly greater than, though of the same order of magnitude as, those we obtained in a previous work (Baeza et al. 1994b) for freshwater collected during 1991 on Livingston Island. (0.280±0.015) Bq/L, the water coming mainly from the Hurd Glacier. Furthermore, they are wholly comparable to those obtained by other workers for seawater at different points of Antarctica, which values lie in the range (0.03, 2.0) Bq/L (Michel et al. 1979, Sakanoue 1987, Chen et al. 1994, Wedekind et al. 1997).

When we corrected the tritium activities for the dates at which the successive snow/ice layers sampled were deposited, using $T_{12}(^{3}H)=12.35$ yr and the dating performed previously (Baeza et al. 1996), we obtained the temporal evolution of the activity levels shown in Figure (1). One sees that the temporal evolution of the tritium activities presents two clearly identifiable behaviours. Firstly the values decline from the maximum value detected in 1969 to the quasi-constant values measured for the ice layers belonging to the years from 1982 to 1993 around a mean value of (1.2±0.4) Bq/L, which we represent by a dashed line on the said figure. We attribute these latter values to the deposition of ^{3}H of purely cosmogenic origin existing in the study zone. They are, indeed, fully comparable to the activity levels of tritium measured by Koide et al. (1979) for the ice layers extracted from the Ross Sea (Antarctica) corresponding to the early 1950’s, whose values vary between 1.2 and 2.4 Bq/L, and which this author indicates are very close to the natural cosmogenic background value existing in the ices of the southern hemisphere.

The levels of tritium deposition that we measured in the Hurd Glacier ice in the years 1980 back to 1969 lie in the range (2.1, 4.4) Bq/L. These are of the same order of magnitude as, though somewhat lower than, the levels detected by Koide in ice samples taken from different localities corresponding to analogous time periods. In particular, he obtained values between 6 and 18 Bq/L for ice layers from the Ross Sea (82 22' S; 168 40' W), with formation dates between 1979 and 1969 (Koide et al. 1979), while for ice from Greenland (65 11' N; 43 50' W), originating between 1980 and 1970, the activity levels measured were between 6 and 36 Bq/L (Koide et al. 1982). We think that the differences with our results must be associated with the different locations from which the samples were taken. Whatever the reason, the behaviour exhibited by the temporal evolution of the tritium levels in ice samples dating from before 1980 leads us to deduce the existence in them of a non-negligible contribution of tritium of man-made origin, probably originating

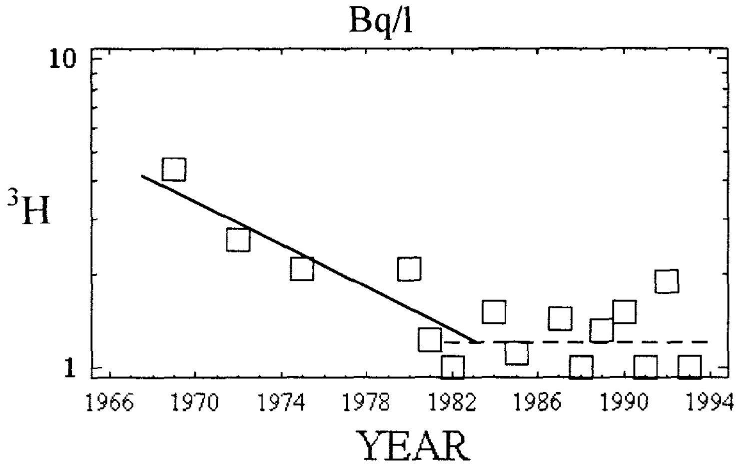

Figure 1. Temporal evolution of the ^{3}H levels present in the Hurd Glacier ice, corrected for decay to the date of formation of each of the ice layers analyzed. Superposed on the measured ^{3}H levels, by means of a continuous and a dashed line, are the observed trends.

in the atmospheric atomic tests carried out basically in the mid-1960's. The steady decline in importance of this man-made contribution is readily appreciated in Figure (1). Indeed, we were able to establish a linear regression of the temporal evolution of the logarithms of the tritium activities measured for the years 1969-1982, which we represent by a continuous line in the cited Figure (1):

$$\text{Ln } A(^3\text{H, Bq/L})=(180\pm40)+(-0.092\pm0.020)*\text{YEAR} \qquad r=-0.919$$

The slope found for the straight line fit allow us to estimate the effect half-life of tritium in the ice samples collected for the time period 1969-80. This is (7.5±16) yr. This value coincides with what we deduced from the plot given by Koide et al. (1979) which show the temporal evolution of the tritium levels detected for the Ross Ice Shelf. One observes in that plot that for the interval from 1969 to 1976 the levels fall to practically one half in a period of approximately 7 yr. That the effective residence half-life of tritium in the ice is less than its physical half-life may be due to a number of causes, amongst which we identify the following two. On the one hand, there exists a certain mobility of tritium towards the lower layers of ice, which differs from year to year as a consequence of the different percolation rates as the ice partially thaws during the warmest periods of each year. On the other hand, there is a contribution from the tritium in the seawater in the immediate vicinity (0.5 to 2 km), in this case diluting that present in the glacier under stud! given the comparatively lower concentration of this radionuclide in the seawater.

With reference to the activity levels measured for ^{234}U and ^{238}U, we can see in Table 1 that these vary respectively, in the ranges [33, 850] µBq/L - with an average value (250±260(SD)) µBq/L - and [12, 780] µBq/L - with an average value (210±250(SD)) µBq/L. The great variability of the uranium levels obtained has to be attributed on the one hand to the different contributions coming from the surrounding land around the glacier during each of the layers of the study and on the other hand, for the years 1970 and 1969, to the possible contribution from the ashes deposited on the glacier as a consequence of the volcanic eruption that occurred in 1970 on Deception Island, as well as its subsequent percolation through the ice to deeper layers.

On the basis of the activity levels measured for each of the ice layers sampled, we may calculate the ratio between the $^{234}U/^{238}U$ activities from the linear regression:

$$A(^{234}U,\quad \mu Bq/L)=(39\pm23)+(1.01\pm0.08)*A(^{238}U,\ \mu Bq/L)\qquad r=0.956$$

In this case, the value of the slope of the straight line fit provides an excellent estimate of the secular equilibrium existing between the two radioisotopes (Ivanovich and Harmon 1982) in the ice samples.

We were not able to find any data in the literature concerning uranium levels in polar region ice with which to contrast the values obtained in the present work. We can state, however, that the values are consistent with those that we obtained (Baeza et al. 1994b) for the freshwater of Livingston Island collected in 1991. Those values were (440±40) and (320±30) µBq/L, respectively for ^{234}U and ^{238}U. These values are greater than those recorded for the glacier ice layer formed in 1991 (see Table 1), and even greater than the average activity values calculated for the 19 years covered by the present study. This is because the cited freshwater not only contains uranium from the ice of the Hurd Glacier, but also that leached from the soil over the glacier’s relatively short fluvial course (about 1.5 km). Applying the equivalence that exists between the activic of ^{238}U and the mass of the cited isotope, we can estimate both the average concentration of natural uranium in the ice of the Hurd Glacier and the value corresponding to the year 1991 in the freshwater, the results being 0.017 and 0.026 µg/L, respectively. Both values lie at the lower level of the range of activities normally reported for freshwater in the literature [0.02, 6.6] µg/L (Ivanovich and Harmon 1982).

With respect to the ^{90}Sr and ^{137}Cs content in the ice layers sampled, these surpassed the procedure’s detection limit only in the case of ^{90}Sr in the sample corresponding to the year 1974, for which the specific activity obtained was (0.6±0.4) mBq/L. In the other ice layers analyzed, the levels measured were systematically lower than the respective detection limits, which are of the order of: 0.5 and 1.8 mBq/L for ^{90}Sr and ^{137}Cs, respectively. These results are consistent with having detected (Baeza et al. 1994b) the following net activity levels for the 1991 Livingston Island freshwater sample: (1.2±0.5) mBq/L for ^{90}Sr and (0.33±0.11) mBq/L for ^{137}Cs. There are two causes that explain the differences in the results of the two sampling campaigns. Firstly, the sample sizes were different: 50 L for each of the ice layers extracted from the glacier, and 450 L for the freshwater sample, so that the detection limit was notably lower for the latter. Secondly, the ^{90}Sr and ^{137}Cs concentrations were predictably greater in the freshwater sample, since this includes not only the concentration of these radionuclides in the ice of the glacier formed after 1969, which is the object of the present study, but also that existing in the ice formed in the previous years during which man-made depositions were far more intense, together with the incorporation of these radionuclides into the freshwater by being leached from the soil along the fluvial final section of the glacier.

The ranges and average values of the activities detected for the different transuranide isotopes that were systematically analyzed, and whose itemized values for each of the ice layers sampled are listed in Table 1, these are:[12.1, 70] µBq/L and average value

(35±16(SD)) µBq/L for ^{241}Am: [16,55] µBq/L and (26±11(SD)) µBq/L for $^{239+240}$Pu; and [0.76, 59] µBq/L and (9±14(SD)) µBq/L for ^{338}Pu. In the case of this last radionuclide, its activity levels corrected for decay back to the dates of formation of the different ice layers analyzed lie in the range [0.83, 71] µBq/L, with an average value of (11±17(SD)) µBq/L. It can be seen that the maximum levels detected are comparable to, though somewhat less than, the values obtained by Cutter et al. (1979) in Antarctic ice layers dated to between 1970 and 1979 whose ranges are: [170, 800] µBq/L for $^{239+240}$Pu, and [17, 300] µBq/L for ^{241}Am. The same conclusion can be drawn on making the comparison with the concentrations measured by Koide et al. (1982) in Greenland ice between 1970 and 1980, whose ranges are: [5, 22] µBq/L for ^{238}Pu, [55, 280] µBq/L for $^{239+240}$Pu, and [30, 55] µBq/L for ^{241}Am. Our values are also greater than the levels detected in seawater at Antarctic latitudes, such as those reported by Holm et al. (1991) of 1.5 µBq/L for $^{239+240}$Pu and 0.3 µBq/L for ^{338}Pu, or in Arctic waters with ranges of [0.65, 2.43] µBq/L for ^{238}Pu, [11.5, 18.1]µBq/L for $^{239+240}$Pu, and [2.3, 4.7] µBq/L for ^{241}Am (Wedekind et al. 1997). This last finding is coherent with the effect of dilution produced by the oceans on the radioactive depositions in the zone (Koide et al. 1979).

As can be appreciated from the values given in the cited Table 1, one again observes a relative variability in the levels detected for the actinides in the different ice layers analyzed. This variability is also manifest in the activity levels reported in other studies (Koide et al. 1979, Koide et al. 1982) for transuranide elements present in ice after the late 1970's. The causes of the said behaviour are twofold. On the one hand, the annual mixing of part of the ice layers, produced by the melting commented on above, for the latitude of our sampling point occurs during the warmest months to a more or less significant degree depending on the year. On the other hand, the extremely low radioactive concentrations existing in the ice that was sampled means that there may be a feedback process affecting the radioactive content of the uppermost layers of the glacier drawing radioisotopes from the deposits existing in the uppermost layers of the soils in the vicinity.

Based on the foregoing remarks, we can indicate that the activity levels that we detected for plutonium and americium oscillate around a constant average value practically after the beginning of the 1980's, undergoing at most a very slight decay. We show this behaviour in Figure (2), in which we present the temporal evolution of the measured activity levels, subsequently smoothed by means of a three-term moving average. One sees in this figure that there is good concordance between the evolution of the three radionuclides represented. This indicates, at least partially a possible common origin for the said radioisotopes. This aspect was brought out in a cross-correlation analysis between the smoothed levels for the cited radionuclides together with tritium, obtaining the value for the latter also by means of a three-term moving average and correcting the data for decay back to the deposition date. The respective resulting correlation coefficients are: ^{338}Pu vs $^{239+240}$Pu (r=0.785), ^{241}Am vs $^{239+240}$Pu (r=0.503) and ^{3}H vs ^{238}Pu (r=0.943).

One also observes in Figure (2) a steady but different decay of the activities detected for ^{238}Pu and $^{239+240}$Pu from the oldest samples collected to those of the early 1980's. This aspect is brought out in Figure (3), in which we show the temporal evolution for the n-hole period sampled of the ratios of the ^{238}Pu activities - corrected for decay to the deposition date - and of $^{239+240}$Pu. As can be seen, the proportion of ^{238}Pu varies between 90% and 20% of $^{239+240}$Pu during the 1970's, after which it remains practically constant at around 20% up until the most recent ice layer. This behaviour practically coincides with those

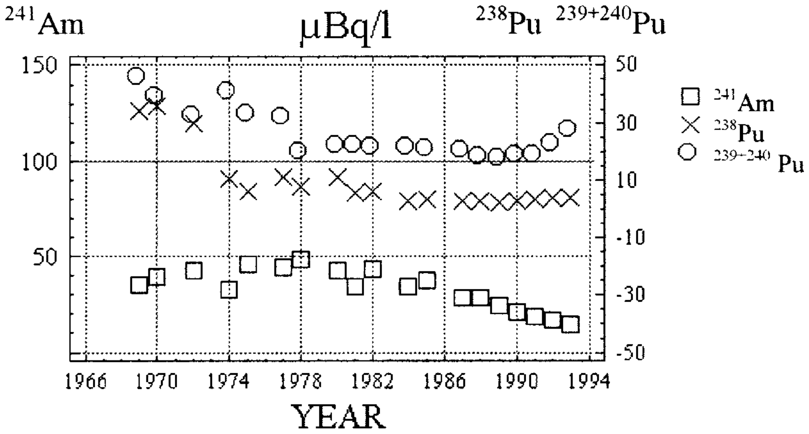

Figure 2. Temporal evolution of the activity levels of ^{241}Am, ^{238}Pu, and $^{239+240}$Pu, smoothed by means of a three-term moving average.

described in a number of studies in which (Koide et al. 1979) the ^{238}Pu percentage declines in Antarctic ice samples from 100% to 10% for the period 1970-77, and between 30% and 10% for the period 1970-75 in Greenland ice samples (Koide et al. 1977), or varies between 14-22% in 1989 for Antarctic seawater samples (Holm et al. 1991) with percentages of 27% in algae and 24% in carpets of mosses and lichens. We have quantified this behaviour, performing logarithmic regressions of the temporal evolution between 1969 and 1982 of the activity levels of each of the plutonium isotopes analyzed, in order to determine their residence half-lives. The effective half-lives obtained were, respectively: T_{12} (^{238}Pu) = (4.8±2.1) yr (r=-0.839) and T_{12} ($^{239+240}$Pu)= (11.4±2.3) yr (r=-0.878). These half-lives are of the order of what may be deduced from the graphs published in various studies (Cutter et al. 1979, Koide et al 1977, Koide et al 1979), as well as of the (7-8) yr established by Holm et al. (1991) for the waters analyzed in his study of the South Atlantic. From the mean annual mass balance that we calculated (Baeza et al 1996) for the snow/ice of the Hurd Glacier zone used in the present study (0.24 $m_w$eq yr^{-1}) and the activity levels that we found for each of the radionuclides analyzed in this work, we shall determine and evaluate the importance pertaining to the fallout of each radionuclides during the time period 1969-93.

For the case of tritium, assuming average contributions of 2.2 and 1.2 Bq/L for the two periods considered separately above in the analysis of the temporal evolution of the activity levels, 1969-82 and 1983-93, and correcting for decay back to their deposition date, we are able to determine their respective mean annual tritium fallout rates. The results are 528 and 288 Bq $m^{-2} yr^{-1}$ From these we can obtain the overall fallout values, 7.4 and 3.2 kBq m^{-2}, for the aforementioned two periods (1969-82 and 1983-93). As is known, 75-80% of the total tritium inventory produced by the atmospheric atomic tests corresponds to the fallout that took place in the northern hemisphere (Kathren 1984), with there being a marked negative gradient from the said hemisphere towards the south. Specifically, the total tritium inventory measured in glacier ice for the period 1944-80 is estimated by Koide et al. (1982) as 811 and 40 kBq m^{-2} for Arctic and Antarctic latitudes, respectively. One can

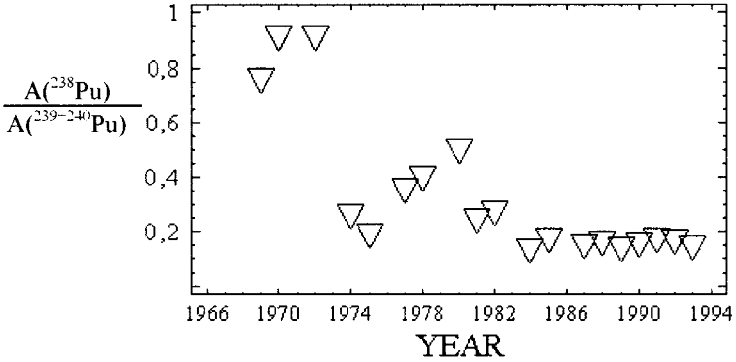

Figure 3. Temporal evolution of the ^{238}Pu/$^{239+240}$Pu activity ratio, smoothed by means of a three-term moving average, as a function of the dating of each of the ice layers analyzed.

hence deduce that the recent man-made contribution of tritium in the zone under study does not surpass 10% of the typical deposition that occurred in Antarctica.

The estimates of the mean annual fallout of ^{234}U and ^{238}U are 0.065 Bq $m^{-2} yr^{-1}$, and 0.050 Bq $m^{-2} yr^{-1}$, respectively, taking 0.25 and 0.21 mBq/L as being the average activities. The result is an overall integrated fallout for the total period of our study, 1969-93, of 1.63 Bq m^{-2} and 1.25 Bq m^{-2}, for the two uranium isotopes, respectively. This represents 100 µg m^{-2} expressed in terms of natural uranium deposition. These values are listed in Table 2 together with the deposition of the gross alpha and diverse alpha-emitting radionuclides. One sees from the table that the uranium deposition is 12% of the deposition of alpha-emitting radionuclides in the ice of the Hurd Glacier (Baeza et al. 1996). The rest of the radionuclides contributing to the overall natural alpha deposition consists fundamentally of other descendants of the natural series, such as ^{210}Po or ^{226}Ra. Indeed, as might have been expected in principle, given the excess of ^{210}Po detected with respect to ^{216}Ra, the former radionuclide is the main contributor to the cited inventory, resulting in the specific case of Table 2 an overall activity even greater than that predicted from the gross alpha count rate. This apparent contradiction is a consequence of the cited overall count rate having only a qualitative value, since the proportional counter used to perform the said determination is calibrated with a single alpha emitter, in our case ^{241}Am.

For the case of the man-made radionuclides, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am, the mean annual fallout obtained was 0.0026, 0.0064 and 0.0083 Bq $m^{-2} yr^{-1}$ respectively, taking as averages of their respective activities 11, 26 and 35 µBq/L. The values of the overall integrated fallout between 1969 and 1993 for these same radionuclides are therefore 0.065, 0.16 and 0.21 Bq m^{-2} respectively. As can be seen comparatively in Table 2, these esclusively man-made depositions represent a minimal part of the total alpha deposition, specifically 0.9% for the case of plutonium and 0.8% for americium. The values that we obtained for the actinide fallout are clearly less than those found by other workers in their respective inventories. By way of example, Koide et al. (1977), in a study of ice layers in

Table 2. Average and integrated deposition in the ice samples from the Hurd Glacier for characteristic alpha-emitting radionuclides. We indicate by α_T the gross alpha activity.

ISOTOPE	MEAN ANNUAL FALLOUT ($Bq\ m^{-2}\ yr^{-1}$)	INTEGRATED FALLOUT (1969-1993) ($Bq\ m^{-2}$)
^{234}U	0.065	1.63
^{238}U	0.050	1.25
^{238}Pu	0.0026	0.065
$^{239+240}Pu$	0.0064	0.16
^{241}Am	0.0083	0.21
^{226}Ra	0.046(*)	1.14
^{210}Po	1.9(*)	47.5
α_T	1.0(*)	25.0

(*) Deposition values calculated from (Baeza et al. 1996).

South Greenland, report an ice deposition rate of 0.6 m_w eq yr^{-1}, and a mean annual fallout for $^{239+240}Pu$, of 0.1 Bq $m^{-2}\,yr^{-1}$ as measured for the period 1944-75. Cutter et al. (1979) for Antarctic ice layers dated between 1955 and 1977, obtains a fallout rate for $^{239+240}Pu$ of 1.5 Bq $m^{-2}\,yr^{-1}$, and for ^{238}Pu of 0.24 Bq $m^{-2}\,yr^{-1}$. In comparing our results with the aforementioned results of other workers, it has to be borne in mind that the time period considered by those authors to calculate the mean annual fallout includes the dates for which the maxima in transuranide fallout activities were observed, as a consequence of the atmospheric atomic tests that took place between 1945 and 1970. This leads to their values for the integrated fallout being significantly greater than those that we estimated for the period 1969-93.

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