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DIRECT DETERMINATION OF GEOCHRONOLOGICALLY-USEFUL RADIONUCLIDES IN SEDIMENTS BY LOW-ENERGY PHOTON ANALYSIS

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• Environment Canada

DIRECT DETERMINATION OF GEOCHRONOLOGICALLY-USEFUL RADIONUCLIDES IN SEDIMENTS BY LOW-ENERGY PHOTON ANALYSIS

DÉTERMINATION DIRECTE PAR L'ANALYSE DE PHOTONS DE FAIBLE ÉNERGIE DES RADIOÉLÉMENTS UTILISÉS EN GÉOCHRONOLOGIE DANS LES SÉDIMENTS

S.R. Joshi et A. Mudroch

MANAGEMENT PERSPECTIVE/EXECUTIVE SUMMARY

The levels of Pb-210 and Am-241 in sediments are normally measured by using complex chemical procedures which are both time consuming and require considerable dexterity in separation chemistry.The two radionuclides,however,also emit very low energy gamma rays which can be measured on a properly calibrated hyperpure germanium detector.The present paper describes the development of such a gamma-ray spectrometric technique.The technique is applied to assign dates to two sediment cores from the Great Slave Lake,NWT.The results are compatible with the dates derived from Cs-137 measurements.

PERSPECTIVE-GESTION/SOMMAIRE

En général, on mesure les niveaux de Pb-210 et Am-241 dans les sédiments en faisant appel à des processus chimiques complexes qui demandent du temps et une dextérité considérable en manipulation chimique de séparation. Cependant, les deux radioéléments émettent également des rayons gamma de très faible énergie que l'on peut mesurer grâce à un détecteur à germanium extrêmement pur et adéquatement étalonné. Le présent document décrit la mise au point de cette technique de spectrométrie gamma. On a utilisé cette méthode pour dater deux carottes de sédiments prélevées dans le Grand lac des Esclaves, T.N.-O. Les résultats sont compatibles avec les données sur les mesures de Cs-137.

Low-energy gamma-ray spectrometry is used for the non-destructive determination of ^{210}Pb, ^{226}Ra and ^{241}Am in sediments. The self-absorption of the low-energy gamma-rays is corrected using a technique involving direct gamma transmission measurements on sample and efficiency calibration standard. Several reference materials when analyzed by the described technique gave values similar to those obtained by other techniques. The utility of the technique is illustrated through the analysis of excess ^{210}Pb and fallout ^{241}Am profiles in two sediment cores from Great Slave Lake, NWT, Canada. It is proposed that simultaneous ^{241}Am measurements provide an attractive alternative to the conventional measurement of fallout ^{137}Cs profiles.

On utilise la spectrométrie gamma à faible énergie pour déterminer de façon non destructive la teneur des sédiments en Pb-210, en Ra-226 et en Am-241. On corrige le facteur d'auto-absorption des rayons gamma de faible énergie en employant une technique de mesure directe des rayons gamma transmis sur les échantillons et une norme d'étalonnage pour mesurer l'efficacité du détecteur. L'analyse de plusieurs matériaux de référence par la technique décrite a permis d'obtenir des résultats semblables à ceux d'autres techniques. On se rend compte de l'utilité de cette technique en analysant les courbes de surplus de Pb-210 et des retombées d'Am-241 dans deux carottes de sédiments provenant du Grand lac des Esclaves, T.N.-O., Canada. On croit que les mesures simultanées de l'Am-241 peuvent avantageusement remplacer les techniques courantes de mesure des courbes de retombées de Cs-137.

1. Introduction

Since their introduction as convenient tools for establishing the chronology of recently-deposited sediments, both atmospherically-derived ^{210}Pb (half-life, 22.3 y) and ^{137}Cs (half-life, 30.1 y) have been extensively used by researchers for various purposes. Some of the environmental applications include studies of the persistence of organotoxics [1, 2], eutrophication in lakes [3], and the recent history of heavy metal pollution [4].

Most researchers measure the atmospherically-delivered or excess ^{210}Pb (i.e. total ^{210}Pb in the sediment less that supported by ^{226}Ra) by measuring the alpha-emission of its 138-d half-life granddaughter, ^{210}Po, with [5] or without [6] regard to the efficiency of the analytical protocol. Measurements ignoring the yield factor could result in serious error since the levels of ^{226}Ra-supported ^{210}Pb in various sections of the sediment cores can vary by a factor of two or more [7, 8]. Furthermore, unless stored for long periods of time, the status of equilibrium between ^{210}Pb and ^{210}Po in top section(s) is not known. An alterantive, and generally superior, approach involves beta-counting of the 5.01-d half-life daughter (^{210}Bi) of ^{210}Pb [9]. This method is relatively complex and requires considerable dexterity in radiochemical separation techniques though it allows a simultaneous assay for ^{137}Cs as well which is normally measured on a Ge(Li) or a hyperpure coaxial Ge detector.

The levels of excess ^{210}Pb present in the sediments may also be ascertained via direct counting of its 46.5-keV (4%) gamma-emission [10, 11]. A generally unrecognized advantage of this non-destructive approach is that a simultaneous determination for ^{226}Ra (half-life, 1620 y) can also be carried out. This facilitates derivation of reliable excess ^{210}Pb values. Another major, but relatively unexploited, advantage of direct photon analysis technique lies in the possibility of deriving ^{241}Am (half-life, 432.2 y) concentrations via its 59.6-keV (35.9%) gamma-emission [12, 13]. This eliminates the need for doing separate assays for ^{137}Cs which is normally measured for checking the results afforded by the ^{210}Pb technique. The present article, therefore, seeks to validate the reliability of the direct photon analysis technique and illustrates its utility through an analysis of the excess ^{210}Pb and fallout ^{241}Am profiles in the recent sediments of Back Bay and Yellowknife Bay (Great Slave Lake), NWT, Canada.

2. Experimental

2.1 Sediment samples

Sediment cores were retrieved in 1984, from locations shown in fig. 1, as part of a project designed to study the effect of gold mining activities on the quality of area sediments. Cores were stored

at 4°C until sectioned. Precise sample thicknesses were obtained by dividing the wet mass of the sediment aliquot by its bulk density and area. The sediment porosity was measured from the wet and dry weights and the density data.

2.2 Nuclear spectrometry

Low-energy gamma-ray intensities were measured with an Aptec hyperpure Ge planar detector with an active area and depth of 1500 mm^2 and 10 mm, respectively. The detector resolution was 720 eV for 122 keV. The 661.6-keV γ-ray of ^{137}Cs was measured in a large volume, closed-end coaxial detector. The detectors were shielded by 10 cm of pre-World War II lead on all sides. Amplified and shaped pulses from the two detectors were analyzed by a Nuclear Data 6620 nuclear spectrometer. The efficiencies of the two detectors were measuared at four different sample thicknesses as described earlier [14].

2.3 Analytical procedure

The dried, powdered and weighed (5-10 g) sample was stored sealed in a 55-mm diameter polystyrene counting vial for over 25 days to ensure equilibrium between ^{226}Ra and its daughters which grow in with the 3.85-d half-life of ^{222}Rn. The gamma-spectrum was accumulated for 2.5×10^5 s by placing the counting vial directly on the detector. The

events in the respective photopeaks were summed and Compton background subtracted. The net sample count rate was obtained by subtracting background count rate, measured with an empty vial, from the photopeak count rate. The principal difficulty in applying low-energy (up to about 300 keV) γ-ray analysis to ^{210}Pb and ^{241}Am measurements is that self-absorption of the radiation by the sample is relatively difficult to correct because the attenuation coefficient for low-energy γ-ray is highly dependent on sample composition. Since the samples to be analyzed were of composition and density different from that for the Lake Ontario sediment used for deriving the efficiency data, a normalization technique was used to eliminate the self-absorption effects. The technique, following the work reported by Cutshall and coworkers [11], relates the net count rate for the sample (S) to that expected for material identical to that used for efficiency calibration (C) in similar geometry by the expression

$$C = S \cdot \frac{\ell n\ (C'/S')}{1 - S'C'} \quad ,$$

where C' and S' are the photon emission rates of ^{210}Pb or ^{241}Am disc source through the unspiked calibrating material and sample, respectively. The respective net sample count rate, normalized to the self-absorption characteristics of the calibrating material, was then converted to disintegration rate by using the detector efficiency data.

2.4 Sedimentation rates

The sedimentation rates were computed from the following expressions derived earlier [7]:

$$A(x) = \frac{P}{S_0(1 - \phi_0)\rho_s} \exp\left[-(\lambda/S_0)\, f(x)\right] , \qquad (1)$$

where

$$f(x) = \frac{1}{(1 - \phi_0)} \left[x - \frac{\phi_0}{\beta}(1 - e^{-\beta x})\right] \qquad (2)$$

with A(x) being the concentration, in mBq/g, of excess ^{210}Pb at depth x cm below the sediment/water interface, P is the flux of excess ^{210}Pb (mBq cm^{-2} y^{-1}) at the interface, ϕ_0 is the porosity at the interface, ρ_s is the density of dry solids, β is a constant, and S_0 the surface sedimentation rate (cm y^{-1}). Linear regression analysis of the log transform of eqn. (1) gives values of S_0, P and ω, the mass sedimentation rate ($=S_0(1-\phi_0)\rho_s$). The errors in the sedimentation rate measurements are also evaluated by linear regression analysis of the core data as described earlier [15]. The correspondence between the age (t) of a sediment core section and its depth is given by

$$t = \frac{x}{S_0(1 - \phi_0)} \left[1 - \phi_0 \left\{\frac{(1 - e^{-\beta x})}{\beta x}\right\}\right] \qquad (3)$$

3. Results and discussion

The low-energy gamma-ray spectrum of the U.S. National Bureau of Standards (NBS) 'Rocky Flats Soil' (SRM-4353), obtained at about 0.21 keV/channel, is shown in fig. 2. An inspection of the spectrum shows that the photopeaks due to ^{210}Pb, ^{241}Am and ^{226}Ra (^{214}Pb) are well resolved from major potential intefering contributions from naturally-occurring radionuclides. According to the decay scheme adopted in some compilations [16, 17], a 59-keV gamma-ray should be emitted in the disintegration of ^{232}Th with an emission probability of about 0.2%. If present, this photoemission would cause a potential interference, especially for samples relatively richer in ^{232}Th, in the gamma-ray spectrometric determination of ^{241}Am. This gamma-ray, however, has never been observed experimentally. The most recent investigation on the gamma-rays from the ^{232}Th decay using a planar detector [18] also failed to detect the 59-keV gamma-ray. Thus, it is reasonable to infer that no correction is needed for ^{232}Th contribution to the 59.6-keV ^{241}Am photopeak.

Since most sediment samples contain ^{235}U at natural levels, the 58.6 keV gamma-emission from its 25.5-h half-life daughter, ^{231}Th, may constitute another potential interference. This photoemission is listed in some compilations [16, 17, 19] but has not been referred to in other investigations on the low-energy gamma-ray spectra of normal uranyl acetate [20] and uranium ore [21]. Even where listed,

different values are given for the absolute intensity of this photoemission. These range from about 0.02 to 0.56% [16, 17, 19]. A recent reevaluation of the gamma-ray emission probabilities in the decay of ^{235}U, while confirming the occurrence of the 58.6-keV gamma-ray, unfortunately, does not list the emission probability [22]. In most cases, however, the contribution will be very small and may be ignored. For example, if both ^{241}Am and ^{235}U are present at same levels, the increase in the ^{241}Am count rate will be less than 2% assuming a 0.56% emission probability for ^{231}Th. If the levels of ^{235}U are significantly higher and if the detector resolution does not discern the 58.6- and 59.6-keV photoemissions, the contribution due to ^{231}Th can be readily inferred from the relatively clean and simultaneously-measureable 143.7-keV emission from ^{235}U.

The 46.5-keV gamma-emission from ^{210}Pb is relatively free of interferences. The nearest gamma-emissions due to naturally-occurring ^{212}Bi (39.9 keV) and ^{214}Pb (53.2 keV) are easily resolved with the planar detector. Of the five gamma-emissions accompanying the decay of ^{226}Ra [16], only the one at 186.2 keV has measureable emission (3.28%). This emission is not resolvable, on the detector used, from the 185.7-keV gamma-emission from ^{235}U. In samples containing natural levels of ^{235}U and ^{238}U (in equilibrium with its daughters), this ^{235}U gamma-emission will account for nearly 42% of the total counts under this photopeak. As for ^{241}Am, this contribution can also be accounted for by using the 143.7-keV ^{235}U photoemission. In a relatively

simpler approach, ^{226}Ra may be measured via the 295.2 (19.2%) or 351.9 (37.1%) keV gamma emissions of ^{214}Pb which grows in with the 3.85-d half-life of ^{222}Rn. Of these two emissions, the one at 351.9 keV provides better sensitivity due to higher emission probability and the lower background in the spectral region. Also, the expected interference from the 351.1-keV ^{211}Bi gamma-emission, if unresolved, is only about 1.6%. On the other hand, the unresolved 293.7-keV ^{234}Pa gamma-ray can constitute about 7.9% of total counts in the 295.2 keV photopeak. Possible release of ^{222}Rn from the sample and its subsequent dispersal in or release from the counting vial constitutes a potential problem in this approach. Experiments using ^{226}Ra-spiked samples layered with epoxy glue did not reveal any discernible differences in concentrations of ^{226}Ra derived from non-layered samples. Similar results have been reported earlier for water [23] and marine phosphorite samples [24], and for shorter-lived ^{220}Rn in sediment samples [14]. All the current measurements in our laboratories, including the ones obtained for the present study are, therefore, performed on non-layered samples and employ the 351.9-keV photopeak for deriving the ^{226}Ra concentrations.

An important issue in low-level radioactivity measurements involves establishing the detection limit of the technique. The issue gains more significance in low-energy gamma-ray spectrometric measurements since the gamma spectral region of interest has much higher Compton background. Currie [25] has derived rigorous and

self-consistent expressions for evaluating the counting data. The central ideas in applying Currie's criteria to counting data involve determining the true mean background (μ_B) and in deciding whether the observations fall under the 'paired observations' (i.e. only one single background measurement is available) or the 'well-known blank' (i.e. background is precisely known from several previous measurements) category. In low-level gamma-ray spectrometric work, it is usually necessary to count the sample for relatively long periods of time and, therefore, one seldom makes more than one measurement on the same sample. Hence most such measurements fall under the more rigorous 'paired observations' category. Following Currie [25], the minimum detectable activity (MDA) values were derived using the expression

$$\text{MDA (mBq)} = \frac{L_D \times 10^3}{\text{count time in seconds} \times \text{fractional efficiency of the detector} \times \text{branching ratio for gamma-emission}},$$

where L_D, in counts, is an a priori detection limit given by the expression

$$L_D = 2.71 + 4.65 \sqrt{\mu_B}$$

For most practical situations, this expression leads to meaningful information since up to an evaluation based on L_D one has already eliminated the possibilities of committing errors of first (i.e.

reporting as detected a substance that in fact is not present) and second (i.e. failing to detect a substance that really is present) kinds.

The MDA values for the ^{241}Am photopeak region in various International Atomic Energy Agency (IAEA) and NBS reference materials are shown in fig. 3 as a function of sample size. The expression for MDA values, at first thought, seems to predict that lower MDAs will be obtained as the sample size is increased. However, as the sample size is increased there is a corresponding increase in the Compton background due to the relatively high-energy emissions in the sample itself. Consequently, lower MDAs with increasing sample size will be obtained only for samples relatively devoid of higher energy emissions. In the present investigation, this is best illustrated by the MDAs obtained for the IAEA 'Marine Sediment' SD-B-3 (fig. 3) which is by far the most high energy-active of all reference materials.

The results of our measurements on five IAEA and NBS reference materials are given in table 1. These results show that direct gamma-ray spectrometry can yield values similar to those obtained by other methods. The value of 6.7 mBq/g for the IAEA 'Marine Sediment' SD-B-3 appears to be in line with the January 1977 IAEAvalue of 5.7 mBq/g and the values obtained by other researchers [13]. No dated $^{241}Pu/^{239,240}Pu$ activity ratios for this sample are available for estimating the growth of ^{241}Am since January 1977.

From these results it would appear that direct gamma-ray spectrometry is capable of providing a general sensitivity of about 1 mBq/g for ^{241}Am. On several occasions, however, we have obtained values of about 0.4 mBq/g for samples relatively free of high-energy emissions. It is this observation that prompted us to derive the concentrations of fallout ^{241}Am present in the recent sediments of Back Bay and Yellowknife Bay of the Great Slave Lake, NWT, Canada. The results of our measurements are shown in fig. 4. The dashed line in the excess ^{210}Pb profile in each case is the best fit to the data points obtained using eqns. (1) and (2).

The parameters listed on fig. 4 are also based on eqns. (1) and (2), while the calendar ages on the curves are deduced from eqn. (3). For comparison, the fallout ^{137}Cs profiles are also shown in fig. 4. In each profile the fallout ^{241}Am is detectable from about 1963 when the atmospheric testing of nuclear weapons peaked. The fallout ^{137}Cs profiles are very similar to the fallout ^{241}Am profiles.

These findings suggest that geochronologically-useful information can be derived by direct counting of the sample on a planar germanium detector, thus eliminating the need for separate measurements for ^{137}Cs on a coaxial germanium or a Ge(Li) detector. Furthermore, the measurement of ^{241}Am is preferable to ^{137}Cs since the latter has significant tendency to migrate deeper into the sediment core [26]. Another point in favour of ^{241}Am measurements is that its levels in sediments will increase in the future. This radioisotope is produced

by the beta-decay of ^{241}Pu (half-life, 14.4 y) and grows in following the expression

$$[^{241}Am]_t = [^{241}Pu]_o \cdot \frac{14.4}{432.2 - 14.4} \cdot \left[\exp\left(\frac{-t\ \ell n2}{432.2}\right) - \exp\left(\frac{-t\ \ell n2}{14.4}\right)\right] ,$$

where t is the time period involved in the growth of ^{241}Am from ^{241}Pu. The quantity of ^{241}Pu remaining following the decay of a given initial quantity of ^{241}Pu is, of course, given by

$$[^{241}Pu]_t = [^{241}Pu]_o \cdot \exp\left(\frac{-t\ \ell n2}{14.4}\right) .$$

The above two expressions were used to calculate the generation of ^{241}Am from fallout ^{241}Pu and the quantities of ^{241}Pu remaining, shown in fig. 5, for the 40-50°N latitudinal band using published [27-29] fallout data and assuming a fallout $^{241}Pu/^{239,240}Pu$ activity ratio of 14.0 [30]. It is quite clear from fig. 5 that the levels of fallout-derived ^{241}Am will continue to increase well into the 21st century. These levels should become readily measureable in most situations with continuing advances in measurement technology. It is also noteworthy that the measurements reported in the present study pertain to the 60-70°N latitudinal band which receives only about 72% of fallout plutonium when compared with the most commonly studied

40-50°N latitudinal band [31]. Thus, in many current situations, fallout ^{241}Am should be readily measureable using the described technique.

4. Conclusion

The results presented in this study clearly show that low-energy gamma-ray spectrometry can provide reliable analytical measurements for ^{210}Pb, ^{226}Ra and ^{241}Am in sediments. The non-destructive feature of the technique makes it especially suitable for environmental studies where the same sample can be subsequently analyzed for non-radioactive pollutants.

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Table 1

Results of gamma-ray spectrometric measurements on IAEA and NBS reference materials.

Reference material	Radionuclide	Certified activity (mBq/g)	Measured activity[a] (mBq/g)
IAEA 'Marine Sediment' (SD-N-1/1)	^{210}Pb	138 (range 101-155)	139±6
	^{226}Ra	109 (range 106-120)	106±4
	^{241}Am	0.56[b]	BDL[c]
IAEA 'Marine Sediment' (SD-B-3)	^{210}Pb	No value given	49.2±17.0
	^{226}Ra	No value given	12.9±6.3
	^{241}Am	5.7±1.0[d]	6.7±0.9
NBS 'Rocky Flats Soil' (SRM-4353)	^{210}Pb	No value given	62.0±6.4
	^{226}Ra	43.0±2.8[e]	46.8±3.2
	^{241}Am	1.37±0.10[b]	1.48±0.40
NBS 'River Sediment' (SRM-4350B)	^{210}Pb	No value	51.1±6.3
	^{226}Ra	35.8±3.6[e]	42.3±3.1
	^{241}Am	0.16±0.03[b]	BDL
NBS 'Peruvian Soil' (SRM-4355)	^{210}Pb	No value given	44.1±6.5
	^{226}Ra	39.0[f];42.0[f]	46.8±3.2
	^{241}Am	(4.2±1.5)10^{-3}[b,e]	BDL

[a]The values reported are averages of several measurements made on samples of different sizes. Each subsample was counted for 2.5×10^5 s and the errors are based on counting statistics of ±1σ.

[b]The certified activity on the reference date has been corrected for the ingrowth of ^{241}Am to the analysis date by using mass spectrometry data for Pu isotopes.

[c]BDL, below detection limit.

[d]Uncertified January 1977 value reported by IAEA Monaco Laboratory.

[e]Errors include those from counting statistics and other analytical protocols.

[f]Uncertified values for ^{214}Bi given by NBS.

Figure Captions

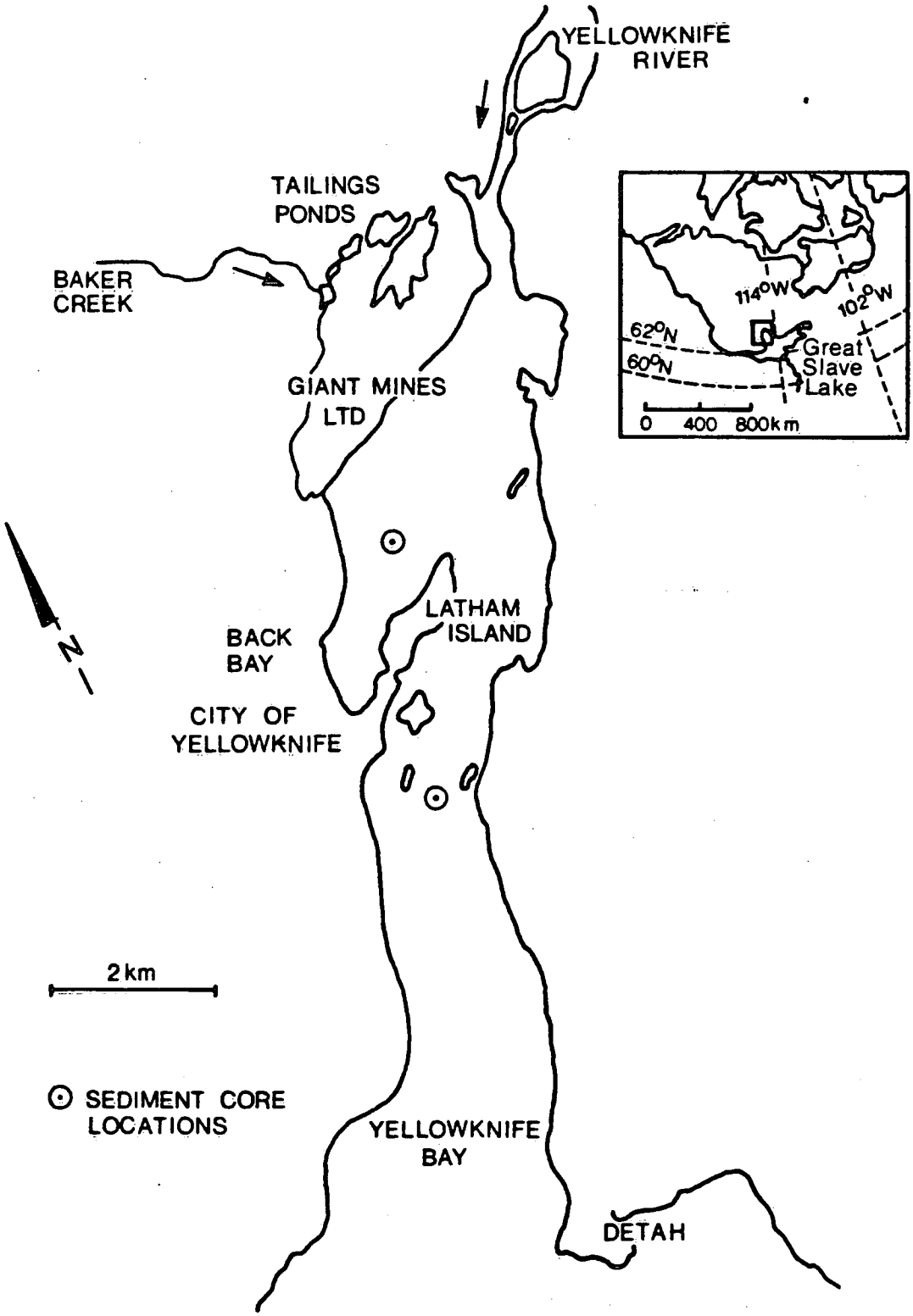
Fig. 1 Sampling locations in Back Bay and Yellowknife Bay, Great Slave Lake.

Fig. 2 The low-energy gamma-ray spectrum of the NBS reference material 'Rocky Flats Soil' (SRM-4353). Energies in KeVs.

Fig. 3 Variation in MDA in the ^{241}Am photopeak region with different amounts of reference materials. Each sample was counted for 2.5×10^5 s. Numbers in brackets refer to sample height in cm.

Fig. 4 Profiles of excess ^{210}Pb, fallout ^{137}Cs, and fallout ^{241}Am in sediment cores from Back Bay and Yellowknife Bay, Great Slave Lake. BDL, below detection limit.

Fig. 5 Estimated growth of ^{241}Am from accumulated fallout ^{241}Pu.


YELLOWKNIFE
RIVER
TAILINGS
PONDS
BAKER
CREEK
GIANT MINES
LTD
LATHAM
ISLAND
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N
2 km
SEDIMENT CORE
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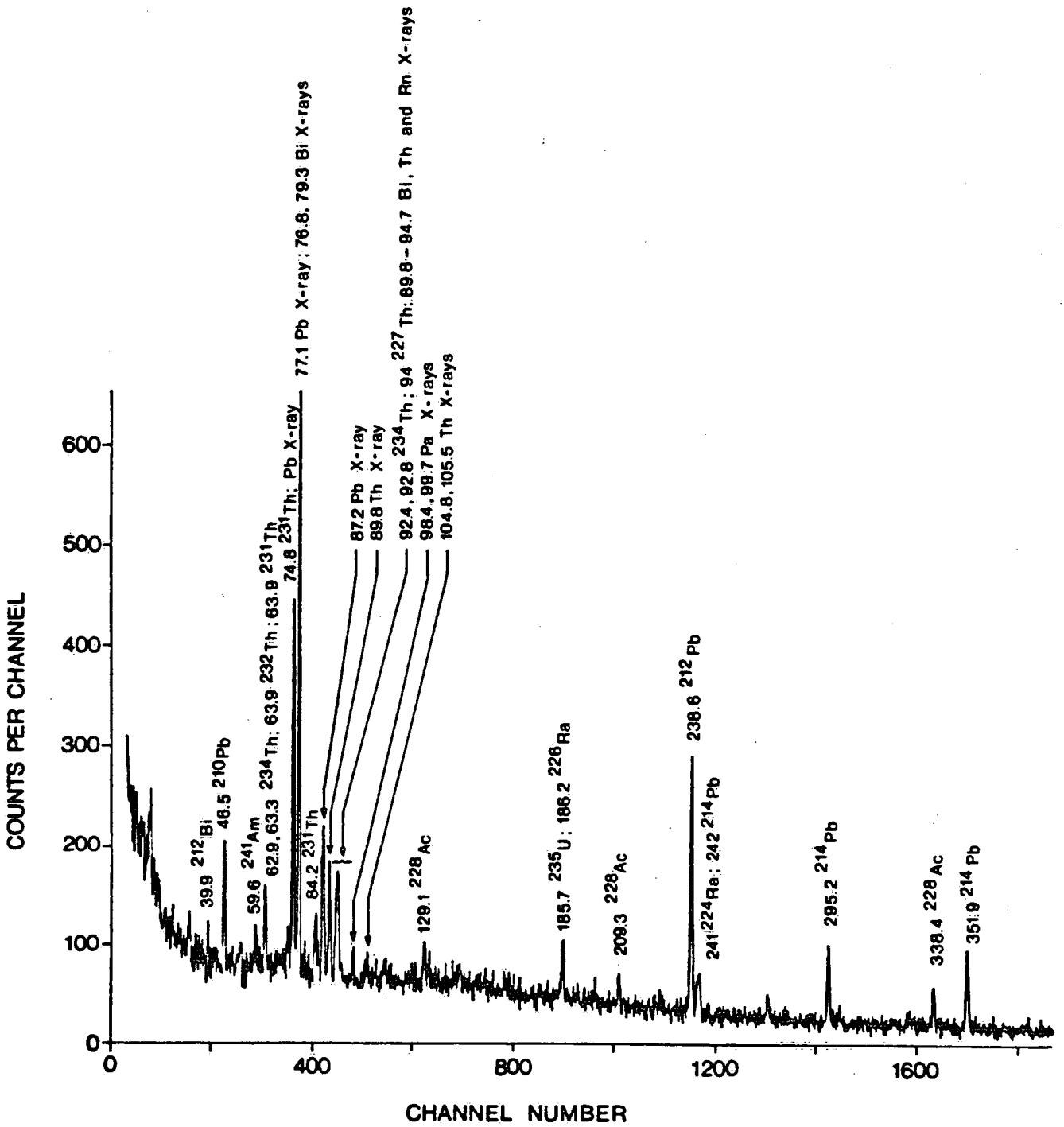
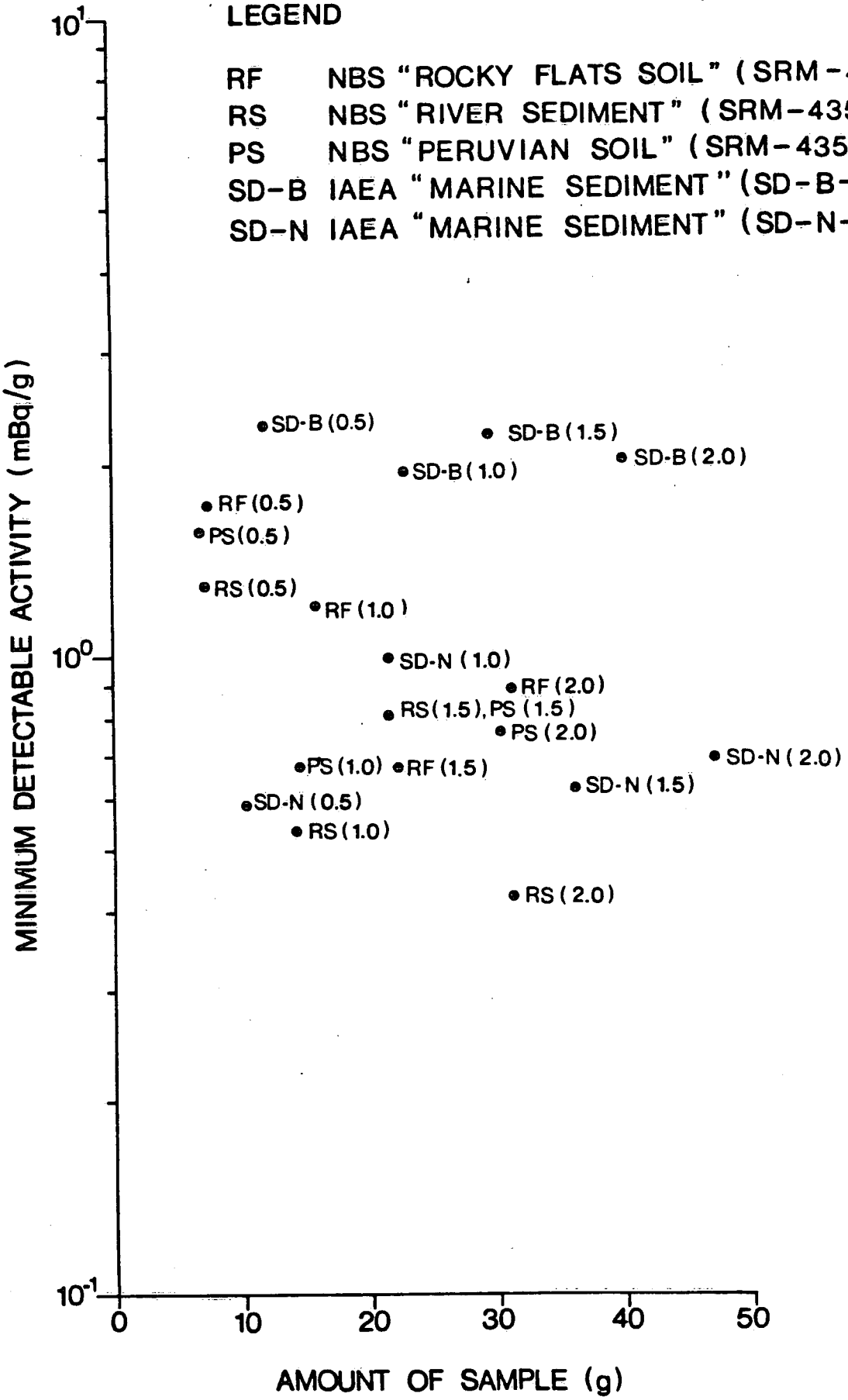
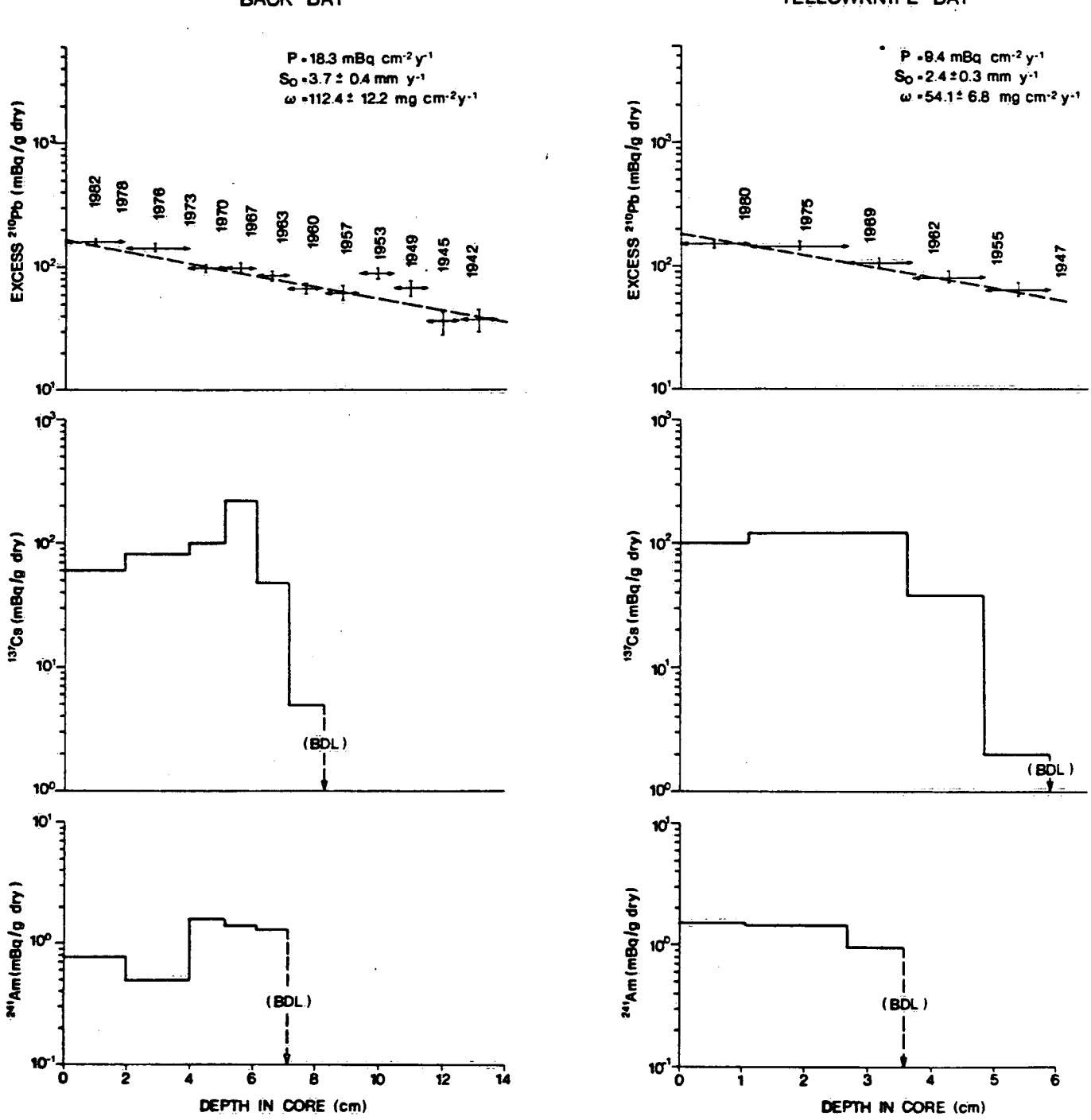
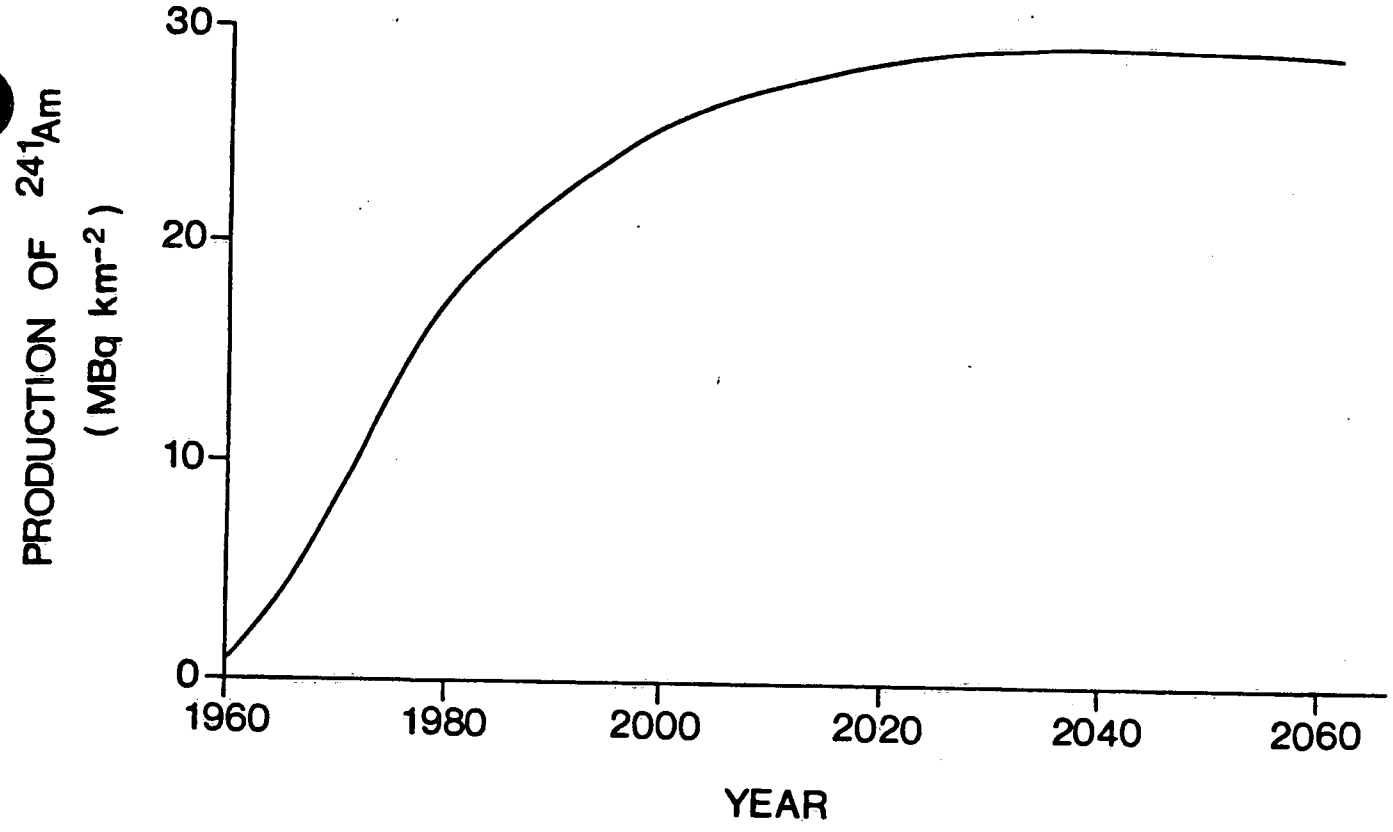
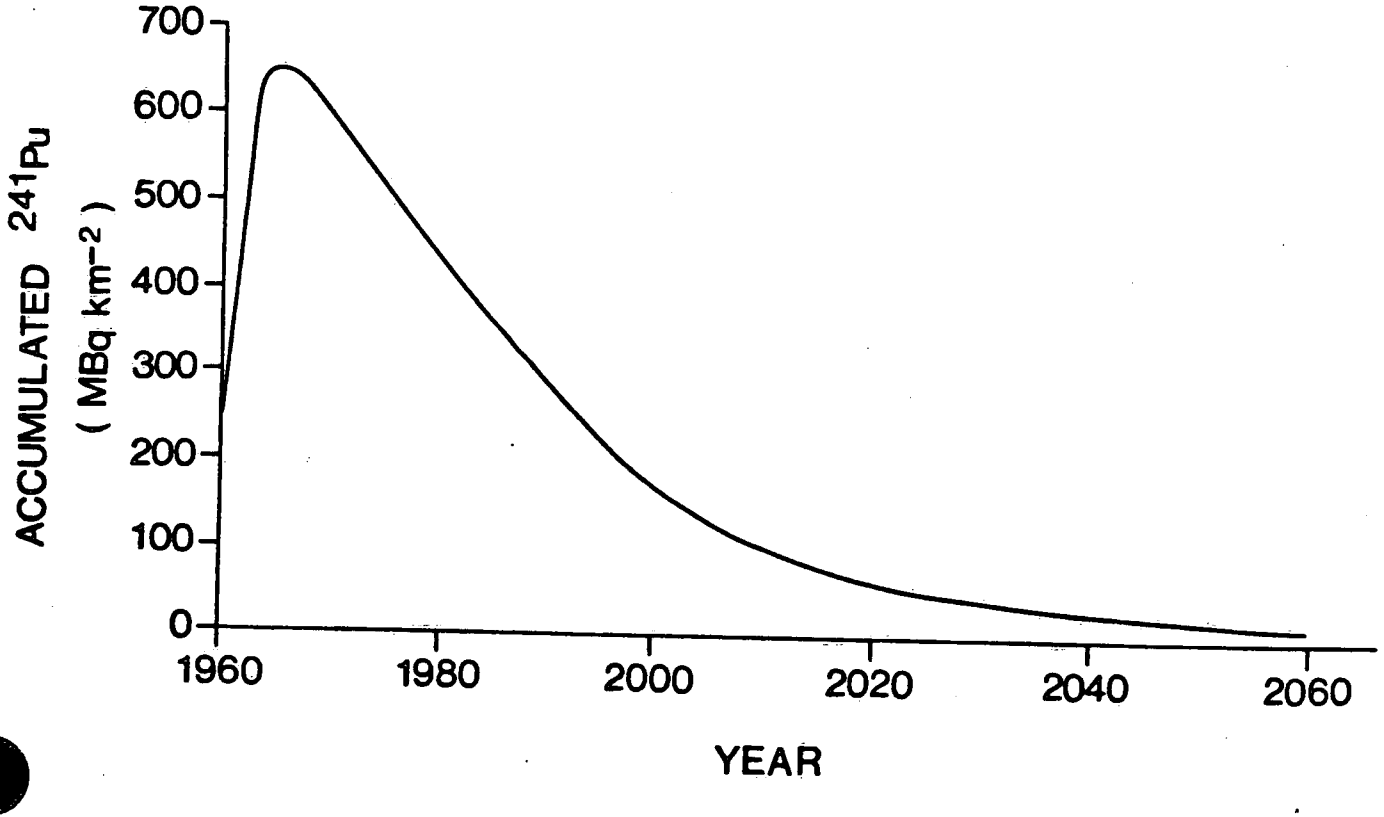

COUNTS PER CHANNEL
CHANNEL NUMBER
0
100
200
300
400
500
600
0
400
800
1200
1600
39.9 212Bi
46.5 210Pb
59.6 241Am
62.9, 63.3 234Th; 63.9 232Th; 63.9 231Th
74.8 231Th; Pb X-ray
77.1 Pb X-ray; 76.8, 79.3 Bi X-rays
84.2 231Th
87.2 Pb X-ray
89.8 Th X-ray
92.4, 92.8 234Th; 94 227Th; 89.8 - 94.7 Bi, Th and Rn X-rays
98.4, 99.7 Pa X-rays
104.8, 105.5 Th X-rays
129.1 228Ac
185.7 235U; 186.2 226Ra
209.3 228Ac
238.6 212Pb
241 224Ra; 242 214Pb
295.2 214Pb
338.4 228Ac
351.9 214Pb


LEGEND
RF NBS "ROCKY FLATS SOIL" (SRM-4353)
RS NBS "RIVER SEDIMENT" (SRM-4350B)
PS NBS "PERUVIAN SOIL" (SRM-4355)
SD-B IAEA "MARINE SEDIMENT" (SD-B-3)
SD-N IAEA "MARINE SEDIMENT" (SD-N-1/1)
MINIMUM DETECTABLE ACTIVITY (mBq/g)
AMOUNT OF SAMPLE (g)
10^1
10^0
10^{-1}
0
10
20
30
40
50
SD-B (0.5)
SD-B (1.5)
SD-B (2.0)
SD-B (1.0)
RF (0.5)
PS (0.5)
RS (0.5)
RF (1.0)
SD-N (1.0)
RF (2.0)
RS (1.5), PS (1.5)
PS (2.0)
SD-N (2.0)
PS (1.0)
RF (1.5)
SD-N (1.5)
SD-N (0.5)
RS (1.0)
RS (2.0)


BACK BAY
P = 18.3 mBq cm⁻² y⁻¹
S₀ = 3.7 ± 0.4 mm y⁻¹
ω = 112.4 ± 12.2 mg cm⁻² y⁻¹
EXCESS ²¹⁰Pb (mBq/g dry)
1982
1978
1976
1973
1970
1967
1963
1960
1957
1953
1949
1945
1942
¹³⁷Cs (mBq/g dry)
(BDL)
²⁴¹Am (mBq/g dry)
(BDL)
DEPTH IN CORE (cm)
YELLOWKNIFE BAY
P = 9.4 mBq cm⁻² y⁻¹
S₀ = 2.4 ± 0.3 mm y⁻¹
ω = 54.1 ± 6.8 mg cm⁻² y⁻¹
EXCESS ²¹⁰Pb (mBq/g dry)
1980
1975
1969
1962
1955
1947
¹³⁷Cs (mBq/g dry)
(BDL)
²⁴¹Am (mBq/g dry)
(BDL)
DEPTH IN CORE (cm)


PRODUCTION OF ^{241}Am
(MBq km^{-2})
30
20
10
0
1960
1980
2000
2020
2040
2060
YEAR


ACCUMULATED ^{241}Pu
(MBq km^{-2})
700
600
500
400
300
200
100
0
1960
1980
2000
2020
2040
2060
YEAR