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**DETERMINATION OF AMERICIUM-241 IN SEDIMENTS
BY DIRECT COUNTING OF LOW-ENERGY PHOTONS**

S.R. Joshi

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Environmental Contaminants Division
National Water Research Institute
Canada Centre for Inland Waters
Burlington, Ontario, Canada L7R 4A6

• Environment Canada

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S.R.Joshi

MANAGEMENT PERSPECTIVE

Americium-241 is one of the most toxic radionuclides which first entered the environment as a result of nuclear weapons testing. Additional sources of this 432-year half-life radionuclide include nuclear fuel reprocessing activities and smoke detector manufacturing operations. Because of complicated chemical procedures involved in its determination, very little work has been done on the behaviour of this radionuclide in the aquatic systems. The present paper reports a simple method for its determination in sediment samples. The basis of the method is the low-energy gamma-emission from the radionuclide which has been successfully used to derive quantitative information as to its levels. A simple normalization technique corrects the self-absorption of this gamma-emission in the sample. The technique's utility is illustrated through results obtained on a Lake Ontario sediment core taken from an area known to be influenced by the U.S. nuclear fuel reprocessing activities.

DOSAGE DE L'AMERICIUM 241 DANS DES SÉDIMENTS PAR DÉNOMBREMENT DIRECT DES PHOTONS À FAIBLE ÉNERGIE

S.R. Joshi

PERSPECTIVE

L'americium 241 est l'un des radionucléides les plus toxiques, qui a d'abord pénétré dans l'environnement par suite des essais sur les armes nucléaires. Ce radionucléide dont la demi-vie est de 432 ans est issu également des activités liées au retraitement des combustibles nucléaires et à la fabrication des détecteurs de fumée. Peu d'études portant sur le comportement de l'americium 241 dans les systèmes aquatiques ont été réalisées vu la nature complexe des techniques chimiques à utiliser pour son dosage. Le présent article décrit une méthode simple de dosage dans des échantillons de sédiments. La technique s'intéresse à l'émission gamma à faible énergie du radionucléide et permet de connaître les teneurs. Une technique simple de normalisation corrige l'auto-absorption de cette émission gamma dans l'échantillon. L'utilité de la méthode est montrée par des résultats obtenus dans une carotte de sédiments prélevée dans le lac Ontario en un endroit que l'on sait touché par les activités américaines liées au retraitement des combustibles nucléaires.

A method is described for the non-destructive determination of ^{241}Am in sediments. The procedure involves direct counting of the 59.6-keV γ -emission of ^{241}Am . The dependence of self-absorption of this low-energy γ -ray on sample density is accounted for by a simple technique utilizing direct gamma transmission measurements on sample and efficiency calibration standard. Measurements on five sediment and soil reference materials show that the technique provides a detection limit of about 1×10^{-3} dps/g for a 2.5×10^5 s count. The utility of the technique in environmental studies is illustrated through results of ^{241}Am measurements on a ^{210}Pb -dated Lake Ontario sediment core.

Nous décrivons ici une méthode de dosage non destructif du ^{241}Am dans des sédiments. La technique comporte le dénombrement direct de l'émission gamma à 59,6 keV du ^{241}Am . Une technique simple comportant la mesure directe de la transmission gamma sur l'échantillon et une norme d'étalonnage de l'efficacité permet de tenir compte de la dépendance de l'auto-absorption de ce rayonnement gamma à faible énergie vis-à-vis de la densité de l'échantillon. Des mesures réalisées sur cinq matériaux contrôle de sol et de sédiment ont montré que la limite de détection est d'environ 1×10^{-3} dps/g pour un dénombrement de $2,5 \times 10^5$ s. L'utilité de cette technique dans le domaine environnemental est illustrée par des résultats du dosage du ^{241}Am dans une carotte de sédiments du lac Ontario datée au ^{210}Pb .

Very small amounts of ^{241}Am (half-life, 432.2 y) are present in the global environment as a result of atmospheric testing of nuclear weapons. Fallout ^{241}Am entering the water column of aquatic ecosystems is rapidly scavenged to the bottom sediments. Measurement of this ^{241}Am can serve as a tool for establishing the chronology of recently deposited sediments. Additional inputs of ^{241}Am to the local aquatic environment may result from other nuclear discharges, principally those associated with spent fuel reprocessing activities. This ^{241}Am is also rapidly removed from water column by sedimenting particulates (1). The aquatic transport of ^{241}Am is thus dominated by sedimentary processes and the analysis of this transuranic pollutant -- next in impact only to plutonium isotopes -- in sediments is of considerable importance.

Traditionally, the determination of ^{241}Am in sediments has relied on the wet chemical procedures involving sample dissolution, ^{241}Am isolation, and electrodeposition steps followed by alpha-particle spectrometry (2-4). These methods are time consuming, require expensive tracers, chemicals and considerably skilled personnel, and are destructive. Direct counting of 59.6-keV ^{241}Am photon (35.9% absolute) constitutes an attractive alternative to conventional methods although it is well recognized that this technique will not be quite as sensitive. Stanners and Aston (5) utilized this approach for measuring ^{241}Am in contaminated sediments using a NaI (Tl) crystal. A specially fabricated SiO_2 absorber was used to eliminate contributions (+3.52%) due to iodine K-X-ray escape peak which is one of the six

potential interferences (6,7). Even in cases when all other contributions can be accounted for, the inherently inferior resolution of NaI (Tl) crystal will permit satisfactory determination of ^{241}Am only in those cases where levels of this radionuclide are substantially higher than those of naturally-occurring radionuclides such as ^{210}Pb (46.5 keV) and ^{238}U - ^{234}Th (63.3 keV). This situation is hardly obtainable in environmental samples. For example, data collected in our laboratories indicates that the currently measured levels of ^{241}Am in Lake Ontario sediment core sections corresponding to the 1963 fallout maximum are about two orders of magnitude lower than the levels of ^{210}Pb and ^{238}U - ^{234}Th found in the same sections. Clearly, in such situations, γ -ray spectrometric technique using a NaI (Tl) crystal is incapable of yielding any meaningful measurements for ^{241}Am .

Brauer et al. (8) improved upon the gamma spectral measurements by using a Ge(Li) detector for the ^{241}Am γ -ray and a Si(Li) detector for Pu/ ^{241}Am L_{β} X-ray ratio. This was followed by high-purity germanium planar detectors which provide even better resolution in the low-energy range. The relative insensitivity to high-energy photons results in reduced Compton scattering in planar detectors. The ensuing corresponding reduction in the continuum background provides planar detectors with much improved sensitivity to low-energy photons when compared with Ge(Li) or germanium coaxial detectors. Cromwell (9) utilized two large-area planar detectors in opposition to measure

^{241}Am in soil samples. Standards fabricated in the same soil type were recommended to properly compensate for low-energy photon attenuation. Subsequently, Larsen and Lee (10) employed a single planar detector to assay ^{241}Am in soils and sediments. A previously described (11) technique was used to allow correction for the self-absorption of γ -rays. The method afforded satisfactory results over a range of approximate five orders of magnitude of ^{241}Am concentrations. The present article gives an account of our evaluation of the low-energy gamma-ray spectrometric technique using several certified reference materials from the International Atomic Energy Agency (IAEA) and the U.S. National Bureau of Standards (NBS). The utility of the technique in environmental studies is illustrated by assessing the ^{241}Am profile, measured by the described technique in 1983, in a ^{210}Pb -dated Lake Ontario sediment core.

EXPERIMENTAL SECTION

Instrumental Methods

The planar detector, shielding, calibration procedures and detector electronics are the same as describe earlier (12). Additional efficiency measurements were also made as follows.

Self-Absorption Correction

The principal difficulty in applying low-energy γ -ray analysis to ^{241}Am measurements in sediment samples 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. Cutshall *et al.* (11) have demonstrated this by measuring the transmission of low-energy γ -rays through ten major matrices comprising their standard for deriving the detector efficiency. Our own measurements on a Lake Ontario sediment sample and various IAEA and NBS reference materials, made by counting the same ^{241}Am source at four different thicknesses for each reference material and shown in Figure 1, clearly illustrate this point. These results lead to the realization that either the composition of the sample be accurately known or, preferably, that some method to correct for self-absorption be used. Wilson (13) has developed a method for measuring the absolute activity of solid samples independent of self-absorption effects. The method yields satisfactory results, but requires replicate counting of increasing volumes of the unknown sample. For most environmental measurements, this requirement is both impractical and time consuming. Cutshall *et al.* (11) have devised a relatively simple method for eliminating the self-absorption effects. The method, based on direct γ -ray transmission measurements on calibration standard and sample, has been applied to measurement of ^{210}Pb (11), ^{241}Am (10), and ^{238}U - ^{234}Th (14). We have also used a

normalization technique, based on the findings of Cutshall et al. (11), to relate 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

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

where C' and S' are the photon emission rates of an ^{241}Am disc source through the unspiked calibrating material (Lake Ontario sediment) and sample, respectively. The 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 inferred from varying amounts (heights) of calibrating material spiked with ^{241}Am . The spiking procedure has been described earlier (12).

Reagents

Standardized ^{241}Am solution was obtained from Amersham Canada Limited.

Procedure

The dried, powdered and weighed (5-40 g) sample was placed in a 55-mm diameter polystyrene counting vial and γ -spectrum accumulated for up to 2.5×10^5 s by placing the vial directly on the planar detector. The events in the 59.6-keV ^{241}Am photopeak 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 sample count rate. The ^{241}Am concentration was then computed from the detector efficiency data and net sample count rate corrected for self-absorption effects as described earlier. The low energy of the γ -rays being counted dictates the use of a low Z thin-walled sample container. Most standard polypropylene/polystyrene vials are suitable containers.

RESULTS AND DISCUSSION

The partial γ -ray spectrum (at 0.21 keV/channel) of IAEA 'Marine Sediment' SD-B-3 is shown in Figure 2. This sample, collected from the vicinity of nuclear installations off the western coast of India, was available to us by virtue of our earlier (15) participation in an intercalibration exercise and is the most active natural matrix material available to us. An inspection of the spectrum clearly shows that the 59.6-keV ^{241}Am photopeak is well resolved from major

potential interfering contributions from naturally-occurring radio-nuclides. According to the decay scheme adopted in some compilations (16,17), a 59-keV γ -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 rich in ^{232}Th , in the γ -ray spectrometric determination of ^{241}Am . This γ -ray, however, has never been observed experimentally. The most recent investigation on the γ -rays from the ^{232}Th decay using a planar detector (18) also failed to detect the 59-keV γ -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 and soil samples are likely to contain ^{235}U , at least at natural levels, the 58.6-keV γ -emission from its 25.5-h 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 γ -ray spectra of normal uranyl acetate (20) and uranium ore (21). The γ -ray spectrum presented in Figure 2 shows that the planar detector is capable of resolving this γ -emission from the ^{241}Am γ -emission. (The fact that we observe a small peak at 58.6 keV in the γ -ray spectrum of SD-B-3, of course, does not necessarily imply that it is from ^{231}Th . A γ -ray of similar energy is also emitted during the decay of ^{60}Fe - $^{60\text{m}}\text{Co}$ with an absolute intensity of 2% (16). In most situations the presence of ^{60}Fe is unlikely, however, as this radionuclide is produced only by the charged-particle activation of ^{64}Cu . Thus, in

most instances, one may have to consider only ^{231}Th). Depending upon the detector resolution available to the observer, it is quite possible that the γ -emissions from ^{241}Am and ^{231}Th may not be adequately resolved. In such cases the contribution due to ^{231}Th γ -ray may be inferred from the simultaneously-measurable and relatively clean 143.7-keV emission from ^{235}U . The extent of interference-correction will, of course, depend on the relative levels of ^{241}Am and ^{235}U . In applying such a correction, one has to know precisely the intensity of the γ -ray emitted. Different values are reported in literature for the absolute intensity of the 58.6-keV γ -ray from ^{231}Th . These range from about .02 to 0.56% (16, 17, 19). A recent reevaluation of the γ -ray emission probabilities in the decay of ^{235}U , while confirming the presence of the 58.6-keV γ -emission, unfortunately, does not list the probability of its emission (22). In many cases, however, the contribution may be very small. For example, ^{235}U , present in the NBS 'Rocky Flats Soil' at 1.9×10^3 dps/g level, will increase the count rate due to the current ^{241}Am level of about 1.4×10^3 dps/g, by only about 2% assuming a 0.56% emission probability. Our own measurements on Lake Ontario sediment core sections indicate that, if unresolved, the 58.6-keV ^{231}Th γ -ray will increase the count rate due to ^{241}Am by between 1.2 and 3.7%.

The crucial issue in many low-level radioactivity measurements involves establishing the sensitivity of the measurement technique. In high-resolution γ -ray spectrometry, where the superior energy resolution provides improved sensitivity, two other factors also

influence sensitivity. Firstly, in most low-level γ -ray spectrometric measurements where contributions due to external background have already been brought to a practical minimum, the sample-associated background is difficult to minimize. Secondly, if one attempts to improve the full energy peak efficiency by, for example, choosing a smaller sample size, there may be a corresponding increase in sample-associated background counting rate also. These two factors are sort of interrelated and may contribute singly or in unison with each other. Their possible interplay may be inferred from the results given in Figure 3 where each sample is found to have its own trend. Some of the variation may be related to sample homogeneity (or the presence of hot particles in cases such as the NBS 'Rocky Flats Soil' standard). The emergent idea from these observations is that sensitivity could be expressed in relation to sample-associated background. In this context, Currie's formulations (23) are reckoned to be the most rigorous, self-consistent and practical. Following Currie's nomenclature and from an operational point of view, an observed signal (net counts) must exceed (i) the critical level, L_c , to yield the decision 'detected', (ii) an a priori detection limit, L_d , at which the given procedure may be relied upon to lead to detection, and (iii) the determination limit, L_Q , at which analytical procedure will give sufficiently precise quantitative estimates. The important thing in applying Currie's criteria to actual measurements is to determine the true mean background, μ_B .

and in deciding whether observations fall under 'paired observations' (i.e. only one single background measurement is available) or 'well-known blank' (i.e. background may be quite precisely determined from several previous measurements). In low-level γ -ray spectrometric assays, 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 γ -ray observations fall under the more rigorous category 'paired observations'. The following working expressions for the 'paired observation' category, given by Currie (23), were used in our evaluation of five IAEA and NBS soils and sediments:

$$L_C = 2.33 \sqrt{\mu_B}$$

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

$$L_Q = 50 \left\{ 1 + \left(1 + \frac{\mu_B}{12.5} \right)^{1/2} \right\}$$

All three parameters are expressed in terms of net counts and are determined entirely by the error-structure of the measurement process, the defined risks, and the maximum acceptable relative standard deviation for quantitative analysis (10% in the case of above expression for L_Q).

The results of our measurements on IAEA and NBS sediment and soil reference materials are presented in Table I. The much-poorer sensitivity of the γ -ray spectrometric technique in relation to the α -particle spectrometric technique (used by the IAEA and NBS for certification) is quite obvious from the data (last two columns) for the NBS 'River Sediment' and 'Peruvian Soil' standards. In each case, the net counts, by γ -ray spectrometry, are also less than L_Q . The superior sensitivity of α -particle spectrometry is understandable in view of the much reduced background for this detection technique (which typically provides only 3-5 background counts per day in the ^{241}Am region of a 450 mm^2 surface-barrier detector in our laboratories, for example). It is to be noted that the net counts given in Table I have not been corrected for the self-absorption effect. If one chooses to apply correction for self-absorption to the net counts at this stage, one might get apparently better results. For example, if we consider unattenuated net counts for IAEA 'Marine Sediment' SD-B-3, we find that in two cases (1.0 and 2.0 cm samples) the unattenuated net counts do exceed L_Q . This is probably not a logical comparison since we are relating unattenuated net sample counts with attenuated background counts. Notwithstanding its limited sensitivity, the γ -ray spectrometric technique can provide useful results for samples containing elevated levels of ^{241}Am if one restricts the evaluation to L_D only. An inspection of the data in Table I reveals that in none of the measurements, does the net counts

exceed L_Q , though in some cases (such as SD-B-3, 1.0 cm) the requirement is nearly met. On the other hand, the dominance of ^{241}Am γ -emission over adjoining background areas is difficult to ignore. One possible way of effecting a compromise between the two observations is to evaluate measured activity of a sample in terms of minimum detectable activity (MDA) based on L_D and defined as

$$\text{MDA} = \frac{L_D \times 10^6}{(\text{dps/g}) \times \frac{\text{count time}}{\text{in seconds}} \times \frac{\text{fractional efficiency}}{\text{of the detector}} \times \frac{\text{branching ratio}}{\text{for } \gamma\text{-emission}} \times \frac{\text{sample weight}}{\text{in g}}}$$

For most practical situations, this evaluation is likely to lead to meaningful information since L_D is based on rather strict formulations when compared with other criteria used in the literature for establishing the sensitivity of a given technique (23). Furthermore, up to this evaluation one has already eliminated the possibilities of 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. L_Q is then simply an extension of L_D under given minimum acceptable relative standard deviation. Thus, in measurements ignoring L_Q , as above, though the relative standard deviation will be higher than the 10% assumed by Currie (23), the more critical possibilities of committing errors of first and second kinds would have been avoided.

The MDA values for various samples are also given in Table I. The results show that in all four measurements on IAEA 'Marine Sediment' SD-B-3 and in three measurements on the NBS 'Rocky Flats Soil' standard, the net counts exceed L_D and the measured activities exceed the corresponding values for MDA. The average of the three measured values for the NBS 'Rocky Flats Soil' standard, 1.48×10^3 dps/g, is in agreement with the certified value of 1.37×10^3 dps/g. Similarly, the average of the four measured values for the IAEA 'Marine Sediment' SD-B-3, 6.7×10^3 dps/g, appears to be in line with the value of 5.7×10^3 dps/g measured by the IAEA Monaco Laboratory perhaps around January 1977 (15). Larsen and Lee (10) measured a value of about 7.1×10^3 dps/g for the IAEA 'Marine Sediment' SD-B-3 in 1982. We have made several measurements on this sample between 1983 and early 1986. All our measurements fall in the range 6.1 – 7.4×10^3 dps/g with no expected trend to increase due to the decay of ^{241}Pu to ^{241}Am . This could be due to sample inhomogeneity. No dated $^{241}\text{Pu}/^{239,240}\text{Pu}$ activity ratios for this sample are available to us for estimating the ingrowth of ^{241}Am . For the IAEA 'Marine Sediment' SD-N-1/1, the net counts for each of the four sample heights are lower than those predicted by L_D , and hence the measurements are deemed to be below detection limit.

From the results given above, it would appear that direct γ -ray spectrometry is capable of providing a sensitivity of about 1×10^3 dps/g for ^{241}Am . While such sensitivity may be of limited use in some studies measuring the absolute levels of fallout ^{241}Am in recently

deposited sediments, the technique can be of much value in cases where limited sample size necessitates the use of a non-destructive measurement methodology or where the ^{241}Am levels are high enough to permit a reliable direct γ -ray spectrometric assay. We first used the technique to measure the profile of ^{241}Am in 1-cm sections of a sediment core retrieved from Lake Ontario, at the mouth of the Niagara River, in September 1982. The study area sediments are known (24) to be influenced by the radioactive releases from a nuclear fuel reprocessing plant in West Valley, N.Y. that began operations in 1966. Though the reprocessing activities ceased in 1972, controlled amounts of radioactive wastes are still being discharged into the local aquatic system. These radioactive materials eventually reach Lake Ontario where some of the radionuclides are deposited in the bottom sediments. In our investigation, we wished to estimate the amount of West Valley-delivered ^{241}Am and establish the pattern of its deposition. For this purpose, we measured the ^{241}Am content of ^{210}Pb -dated sediment core sections. The simultaneous ^{210}Pb and ^{226}Ra measurements (to derive the concentration of atmospherically-delivered ^{210}Pb) were also made by low-energy γ -ray spectrometry. Using the flow characteristics of the study area, the ^{210}Pb -derived sedimentation rate, the monitoring data for the receiving waters (25), and the distribution coefficient for ^{241}Am , we were able to infer that nearly 79% of the total ^{241}Am in the sediment core originates from the West Valley site with fallout from nuclear weapons testing

accounting for the balance. Figure 4 shows the time patterns of fallout and West Valley-delivered ^{241}Am deposition. The ^{210}Pb -derived time scale applies to the measured values of ^{241}Am in the sediment core, while the arbitrary time scale corresponds to the fallout and West Valley-derived ^{241}Am . The ^{241}Am activity levels measured in the dried core segments varied between about 1×10^3 and 3×10^3 dps/g. The reliability of the measurements can be easily discerned by a qualitative interpretation of the ^{241}Am profile. Firstly, the detectable amounts of ^{241}Am appear first around the late 1950's when atmospheric testing of nuclear weapons started to increase. Secondly, the observed peak ^{241}Am activity corresponds to the peak discharges from the West Valley site (25). Thirdly, there is a good agreement between measured ^{241}Am values and the estimated (26, 27) fallout ^{241}Am input to the study area prior to the commencement of nuclear fuel reprocessing operations at West Valley.

In conclusion, the results presented in this article clearly demonstrate that direct γ -ray spectrometry can provide meaningful measurements at about 1×10^3 dps/g level. The most attractive feature of the technique is that simultaneous, non-destructive measurements on the same sample can be made for other radionuclides (such as ^{210}Pb , ^{226}Ra , ^{238}U , etc.) as is often required in environmental studies. An evaluation of low-energy γ -ray spectrometric technique for ^{238}U (14) has recently been reported (28).

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Table 1. Results of Measurements on NBS and IAEA Sediment and Soil Reference Materials

reference material	certified activity, dps/g($\times 10^3$) ^{c,d}	height, cm	M_B , counts	L_C , counts	L_D , counts	L_Q , counts	net counts ^a	MDA, dps/g($\times 10^3$)	measured activity, dps/g($\times 10^3$) ^b
NBS 'Rocky Flats Soil' (SRM-4353)	1.37 ± 0.10	0.5 1.0 1.5 2.0	494 936 560 1001	52 71 55 74	106 145 113 150	368 486 388 500	81 150 133 290	2.1 1.3 0.8 1.0	BDL ^e 1.51 \pm 0.45 1.03 \pm 0.27 1.90 \pm 0.31
NBS 'River Sediment' (SRM-4350B)	0.16 ± 0.03	0.5 1.0 1.5 2.0	365 302 697 372	45 40 62 50	91 84 125 92	325 301 427 327	12 0 0 25	1.8 0.9 0.9 0.6	BDL BDL BDL BDL
NBS 'Peruvian Soil' (SRM-4355)	$(4.2 \pm 1.5) \times 10^{-3}$	0.5 1.0 1.5 2.0	447 383 686 787	49 46 61 65	101 94 125 133	353 331 424 450	0 20 0 4	2.0 0.9 0.9 0.9	BDL BDL BDL BDL
IAEA 'Marine Sediment' SD-B-3	5.7 ± 1.0	0.5 1.0 1.5 2.0	2073 4281 7401 6992	106 152 200 195	214 307 403 392	696 977 1268 1234	558 967 1004 1095	2.4 1.9 2.2 2.0	6.86 \pm 0.83 7.10 \pm 0.72 6.20 \pm 0.78 6.57 \pm 0.75
IAEA 'Marine Sediment' SD-N-1/1	0.56 [confidence limit (0.05): 0.55-0.58]	0.5 1.0 1.5 2.0	273 1106 1047 1326	38 77 75 85	80 157 153 172	289 523 510 567	21 118 135 161	1.0 1.1 0.7 0.7	BDL BDL BDL BDL

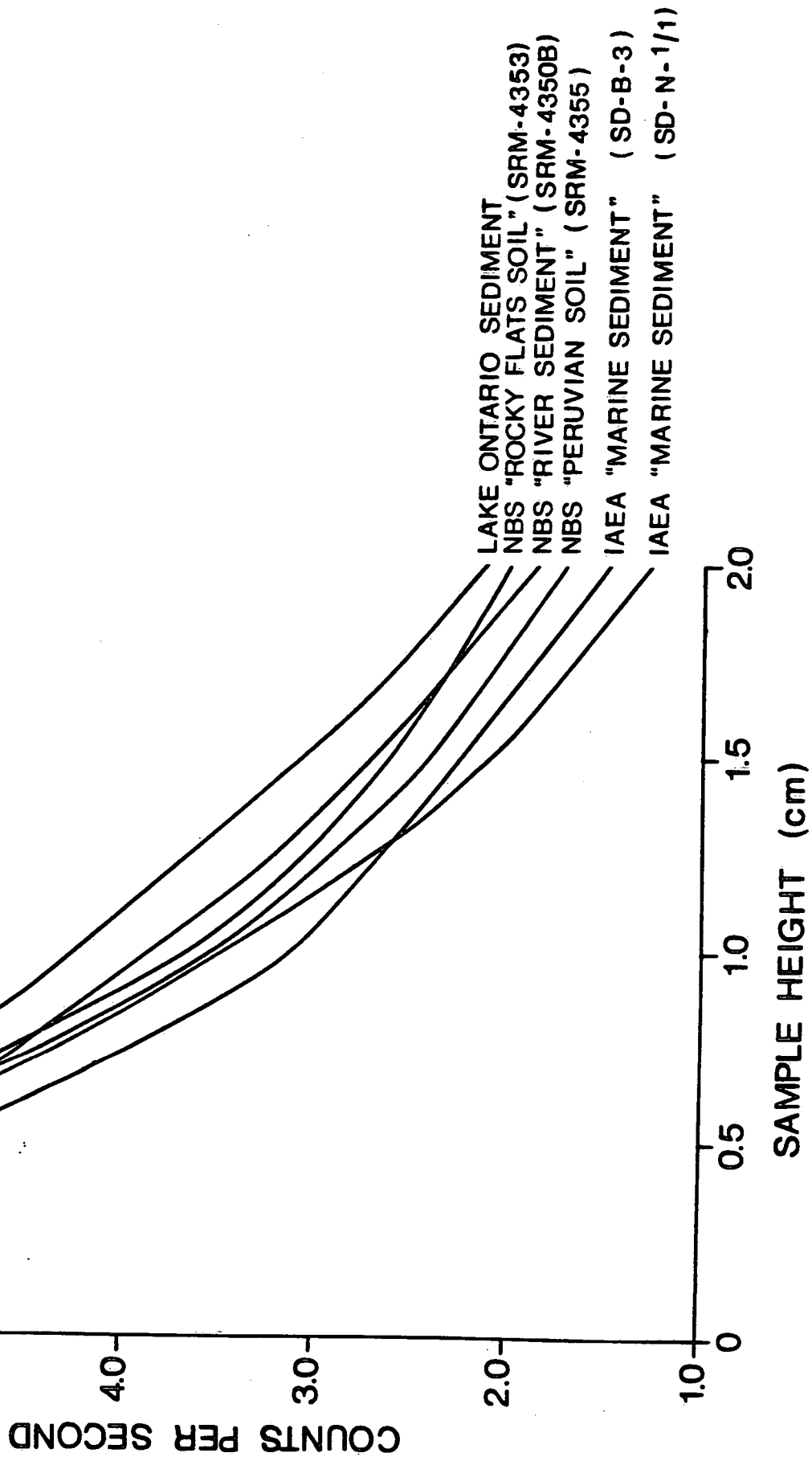
^aEach subsample was counted for 2.5×10^5 s.^bErrors are based only on counting statistics of $\pm 1\sigma$.^cErrors include those from counting statistics and other analytical protocols.^dThe certified activity on the reference date has been corrected for the ingrowth of ^{241}Am to the analysis date by using mass spectrometry data for plutonium isotopes and the relation

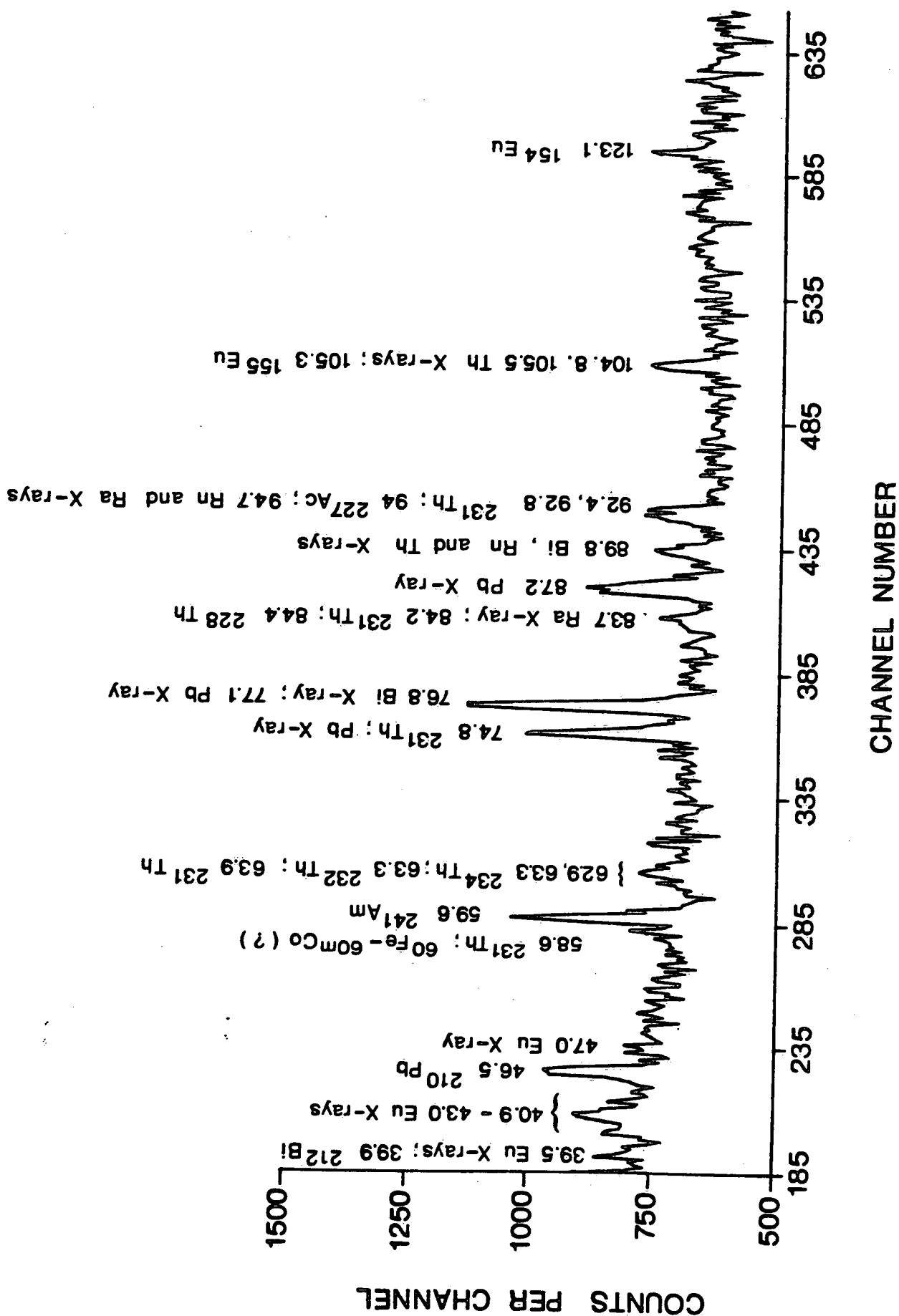
$$[\text{Am}]_t = [\text{Pu}]_0 \cdot \frac{T_{\text{Pu}}}{T_{\text{Am}} - T_{\text{Pu}}} \cdot \left\{ \exp\left(\frac{-t \cdot \ln 2}{T_{\text{Am}}}\right) - \exp\left(\frac{-t \cdot \ln 2}{T_{\text{Pu}}}\right) \right\},$$

where t is the time period involved in the growth of ^{241}Am from ^{241}Pu .^eBDL, below detection limit defined by MDA.^fUncertified January 1977 value reported by IAEA Monaco Laboratory; see text also.

FIGURE CAPTIONS

- Figure 1 Gamma-emission rates from a ^{241}Am source at various thicknesses of different sediment and soil samples.
- Figure 2 Relevant portion of the γ -ray spectrum of IAEA 'Marine Sediment' SD-B-3 obtained with the planar detector. Energies are in kiloelectronvolts.
- Figure 3 Variation in background counts in the ^{241}Am peak region with different amounts of sediment and soil samples. Each sample was counted for 2.5×10^5 s. Numbers in brackets refer to sample heights in cm.
- Figure 4 Time pattern of ^{241}Am deposition in a Lake Ontario sediment core. The shaded areas, corresponding to an arbitrary time scale, depict estimated inputs of fallout and West Valley-derived ^{241}Am . Inputs from Lake Erie (fallout) ^{241}Am , though extremely small, are included.





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)
 LO LAKE ONTARIO SEDIMENT

