
Estimation of rates of sedimentation and useful life of lakes / reservoirs by ^{137}Cs dating technique

1.0 Introduction

The atmospheric fallout Radioactive ^{137}Cs isotope has been unequivocally established as an useful tool in estimating the rate of sedimentation in surface water bodies. The technique is based on the following characteristics of ^{137}Cs .

- a) relatively short residence time in the atmosphere
- b) completely of anthropogenic origin with no natural production
- c) significant variation in production levels during different time periods
- d) moderate half-life of about 30 years
- e) reliable records of global distribution (fallout pattern)
- f) characteristic of Cs to adhere to organic and clayey material with negligible mobility in the environment

The fact that ^{137}Cs appeared in the environment only during the late 1950's make it useful for studying the recent rates of sedimentation in the lakes. Whilst this may be of disadvantage to certain extent, the knowledge of rates of sedimentation in water bodies are required only for the period of increased human activity. The development related anthropogenic activities increased manifold only during the post second world war period that coincides with the ^{137}Cs appearance in environment. Therefore, ^{137}Cs is an apt tool to study the changes brought about by development in the catchment area of lakes/ reservoirs.

In this manual the methodology adopted for using the ^{137}Cs dating technique as a tool to study the rates and pattern of sedimentation in lakes reservoirs has been discussed in detail.

2.0 Scope

All references made about the manufacturers of equipment, reagents and tools pertain to the current practices followed at the Environmental Isotope Section of the Nuclear Hydrology Laboratory at National Institute of Hydrology, Roorkee. The contents shall aid the users in employing the ^{137}Cs dating technique for estimating the rates of recent sedimentation (post-1963) and in estimating the useful life of lakes/reservoirs by this technique.

3.0 Sample Collection

3.1 Need for representative samples

Surface water storage structures such as a lake / reservoir is prone to sedimentation as a consequence of reduction in flow velocity. Within a lake environment, the rate of sedimentation need not necessarily be uniform. Combination of different factors such as velocity, turbulence, slope, depth of water column, size and nature of sediments etc bring about the variation in the rates of sedimentation in different parts of the lake. Therefore, for a more accurate estimation of rate of sedimentation in a lake, sediment cores must be collected from different parts of the lake based on distance from major inflow points, depth to lake bottom, bed slope, known information on thickness of sediment layer, area of lake etc.

3.2 Optimum number of samples

There is no definite law governing the optimum number of sediment cores to be collected from a lake, although guidelines have been proposed by some limnologists (Håkanson and Jansson, 1983). The number of samples to be collected for a study is usually decided based on budgetary constraints, logistics, time availability, and the project objectives. Preliminary estimates reveal that on an average it may cost about Rs. 350.00 per sample for measuring the ^{137}Cs

activity. This means that for sediment core of about 60cm length, it will cost around Rs. 10,000/-.

According to the guidelines proposed by Håkanson and Jansson (1983) the number of sediment cores collected from a lake should preferably be of even number. The following equation may be used to decide the optimum number of samples from a lake:

$$n = 2.5 + 0.5\sqrt{aF}$$

Where, n = number of samples,

a = area of the lake (km²)

F = lake bottom roughness factor

The lake bottom roughness factor, F, may be estimated using the following equation:

$$F = \frac{l_0}{2\sqrt{\pi A}}$$

Where, l_0 = shoreline length (km)

A = area of the lake including islands (km²)

For example if, F = 2 and A = 20, then n = 5.7 or say 6. Therefore, six cores have to be collected. In this case one has to collect six cores from the lake under consideration keeping in mind the need to make the cores representative of different sedimentary environment. This may be achieved by making grids and collecting the cores from the centre point of the grids. The grid size in the case of the above lake will be $20/6 = 3.3 \text{ km}^2$. Taking the square root of 3.3 is 1.8 km. The grid size will then be 1.8km X 1.8km.

3.3 Sampler description

Several sampling devices are commercially available. A description of different types of samplers may be had from literature. The sampler that is used presently by the isotope hydrology group of the NIH is a gravity corer, supplied by KC Maskinger og Laboratorieudstyr, Denmark (Kajak corer). A short description of the corer is given below:

The sample corer (Figure 1) consists of 22-kg counter weights, with either 50-cm long sampling tube or 100-cm sampling tube. The sampler has a steel threaded ring for attaching the sampling tubes. For smooth penetration of the lakebed, a bevelled steel ring is attached to the bottom of the core tube. For extraction of the samples, an adjustable piston rod with silicone packing is also available. The internal diameter of the sampling tube is approx. 5 cm.

3.4 Sample preservation, storage and numbering system

No special preservation procedures are required for ^{137}Cs dating technique. The collected cores are placed vertically and a light coloured opaque graduated adhesive tape may be fixed on the core tube, with the markings (in cm) running from top to bottom. The total length of the sediment core has to be recorded in the sampling information sheet (Appendix-A). Required number of sample storage device (polythene bags / plastic containers) may be numbered accordingly. The core is sliced into several sections of optimum thickness (usually 1 to 2-cm thick) with the aid of a piston (part of the sampling device) and a clean knife or any such sharp edged tool and stored in the sample storage devices (polythene bags / wide mouth plastic bottles) sequentially.

The numbering system is very important to avoid misinterpretation of results. It is better to use a 4-digit alphanumeric system such as LI##, where L stands for the first letter of the name of the lake, I denotes the sediment core

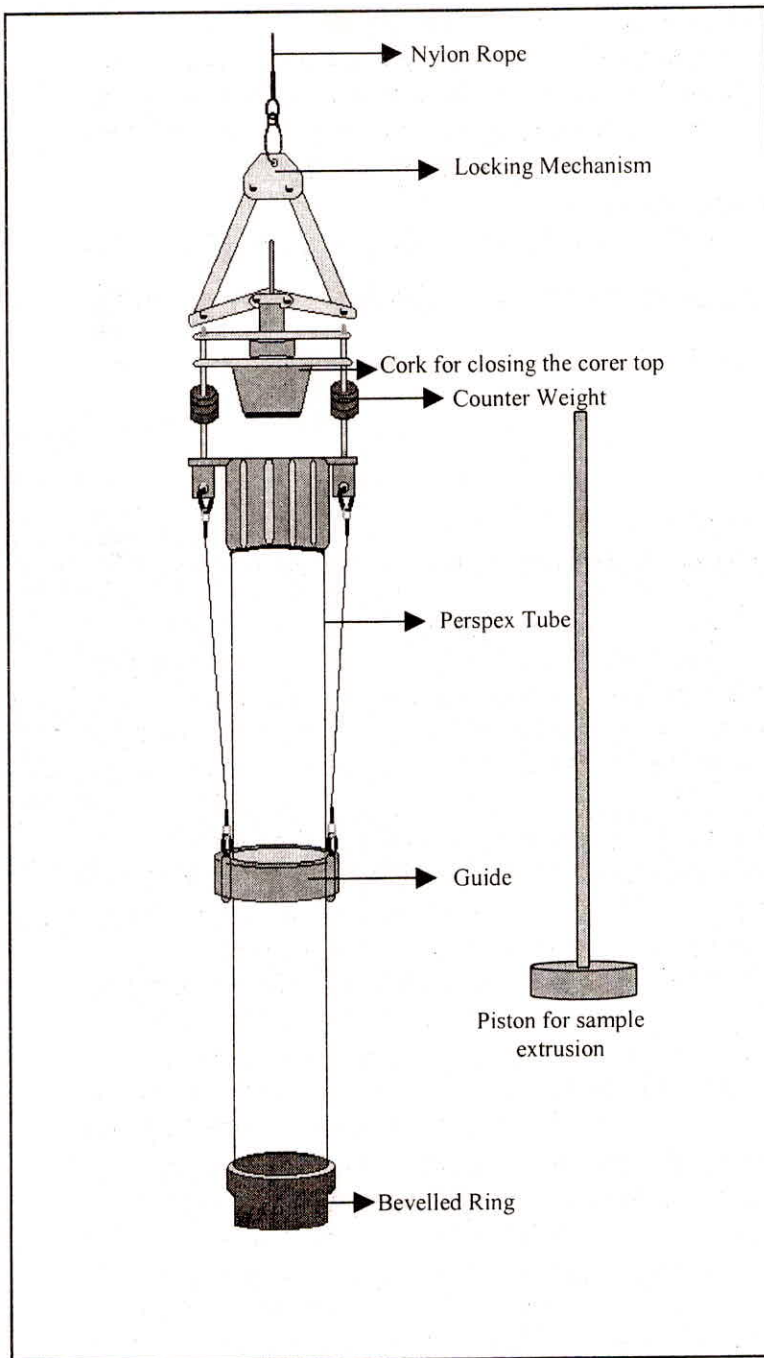


Figure. 1 Gravity type sediment corer for collecting sediment cores from water bodies

location within the lake, the last two numbers (##) denote the sample section from the core. For example in case of lake Nainital the following number was used: NS12 - N-Nainital, S-Core ID, 12 is the twelfth section from the bottom. Usually #1 corresponds to the bottom of the core and the highest number corresponds to the top of the core. Alternatively, the reverse may also be adopted. Nevertheless, it is essential that the system of numbering be kept uniform for a particular lake study.

3.5 Precautions

Cleaning the knife or sample cutting tool after cutting of each sample, with acetone and tissue papers is recommended. Care must be taken in numbering the sample bags / sample containers. It is essential that the piston removes exactly the selected thickness of samples. Due to the soft nature of the sediment samples, it has sometimes been observed that the thickness of all the samples cut from a core is not uniform. This is reflected by large variation in the bulk density of sediment samples.

3.6 Maps, sample records

The locations from where the cores have been collected are to be recorded in a map. The exactness of the markings is more relevant during interpretation. It is imperative that the detailed bathymetric map of the lake be prepared before initiating the sampling program.

4.0 Sample Processing

4.1 Determination of bulk density

As soon as the samples are transported to the lab, the samples have to be emptied into clean and dry stainless steel containers. The weight of the empty container and weight of the container plus the sample are to be recorded properly.

For weighing purpose an electronic weighing machine with a precision of 0.01g may be used. The bulk density (ρ_b) is calculated as:

$$\rho_b = W_w/V$$

where, W_w = wet weight of the sample (g)

V = volume of the sample (cc)

If a core is collected using the Kajak corer as described above, then the volume of a 2-cm section of sample is equal to 39.27 cm³.

4.2 Oven drying

The SS container is then placed in the temperature controlled oven at a pre-set temperature of 105°C for about 24 hours for removal of moisture.

4.3 Determination of dry density

The dry density (ρ_d) may be determined as

$$\rho_d = W_d/V$$

where, W_d = dry weight of the sample (g)

V = volume of the sample (cc)

4.4 Moisture content

The moisture content (Q) is determined by

$$Q = (W_w - W_d)/W_w$$

where, W_w = wet weight of the sample (g)

W_d = dry weight of the sample (g)

4.5 Powdering and sample homogenisation

A sediment sample usually contains a considerable proportion of clay fraction. This leads to lumping of samples following the oven drying. For radioactivity measurements these lumps have to be powdered and homogenised. This may be carried out manually by using the Agate Pestle and Mortar that are cleaned with acetone and tissue papers regularly after every sample.

4.6 Determination of organic contents

The proportion of organic matter in a sediment sample may be determined by different methods. The most simplest and direct method is to burn the sample. This is done by taking a known weight of sample (usually one gram) in a pre-weighed dry and clean silica crucible. For weighing purpose, a high-precision (1-mg) balance (such as Mettler AT200) may be used. The crucible is then placed in a muffle-furnace. The sample is heated at a temperature of about 550°C for 30-minutes.

Usually it takes 3 - 4 hours to reach this high temperature, and cooling takes about another two hours. It is dangerous to open the front door of the muffle-furnace before it cools down to a temperature of about 50°C. The crucibles are then removed (with the help of steel tongs) from the furnace and allowed to further cool down to room temperature in a dessicator. The dessicator contains at the bottom some quantity of silica gel. Make sure that the colour of the silica gel is blue. If it is whitish then the silica gel has to be replaced. After cooling, remove the crucible containing the burnt sample from the dessicator and weigh. The loss on ignition (L_{OI}) is then calculated by:

$$L_{OI} (\%) = (IW-FW)/IW*100$$

where, W_1 = Initial weight (g)

= (Weight of crucible plus raw sample less the weight of empty crucible)

W_F = Final weight (g)

= (Weight of crucible plus burnt sample less the weight of empty crucible)

At NIH Roorkee, the sediment sample is burnt using a Muffle furnace, that has limited space. Therefore, one can burn only four samples a day.

5.0 Activity Measurement

5.1 Some basics of ^{137}Cs measurement by γ -spectrometry

The ^{137}Cs peak has energy of 661.66 KeV. In γ -spectrometry interference in ^{137}Cs peak is possible due only to ^{214}Bi peak at 666 KeV. This interference can easily be corrected by measuring the adjacent 609.3 KeV peak of ^{214}Bi peak and applying proportional correction.

5.2 HPGe detector - Multi-channel Gamma ray system

For measurement of ^{137}Cs activity the EG & G ORTEC multi-channel gamma ray spectrometer (MCA) is used (Figure 2). The connections between the HPGe detector and the MCA as well as that between the PC and the MCA are made as shown in the figure. The SD slot in the MCA panel is shortened with a 50Ω terminator. This is to bypass the requirement of a liquid nitrogen level controller that automatically shuts down the HV supply in case of very low level of liquid nitrogen (LN_2). However, in the present set-up at NIH Roorkee, two weighing balances are used, the combined reading of the two must be greater than 21kg. In the Figure 2, the double concentric circles show the positions of indicator lights such as that for indicating the a) activity of MCA and b) High Voltage operations. The potentiometer knob shown at the left bottom corner of the Figure

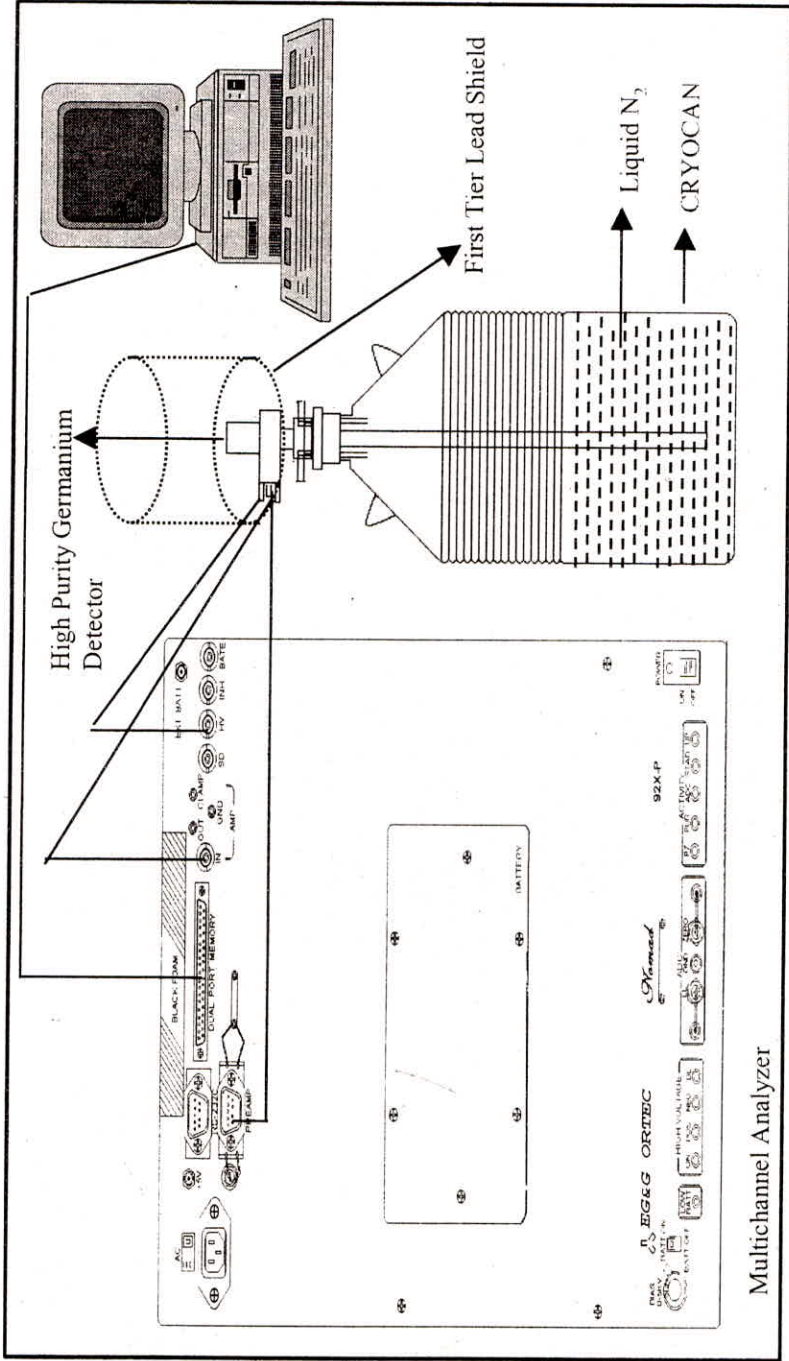


Figure 2. Set-up of Multichannel Gamma Ray Spectrometer with HPGe Detector

2, is the one which is used to set the High Voltage. Care must be exercised while operating the system. Proper procedure for using this is given in the subsequent sections.

The Priston Gamma Tech., USA (PGT) converts the radioactivity incident on the Germanium crystal into analogue signal that is received by the MCA system. The system processes the input signal from the detector and sends the processed digital signal to the computer attached to the system. The entire operation is controlled through the MAESTRO32 software supplied by EG & G. As the HPGe detector is very sensitive to all types of radioactivity a heavy and thick lead shield is provided around the detector to reduce the radioactivity emitted from the surroundings.

5.3 MAESTRO32 program

This is the software used for controlling the MCA operation and also for processing the digital signal received from the detector through the Analogue to Digital Converter (ADC) into a spectrum. This software in combination with Multi-channel buffer (MCB) emulates a MCA. The MCB performs the actual pulse height analyses while the computer and operating system make available the display facility and data-archiving hardware and drivers. The MAESTRO 32 software links these components to provide meaningful access to MCB. For detailed descriptions, refer the software manual.

The main menu comprises of six sets of sub-menus, viz. File, Acquire, Calculate, Services, ROI and Display. Further, the main menu screen also has several icons in the toolbar for file operations and display options. All the above commands are familiar to users of Windows Operating System and they principally address the management of spectrum files.

Before the actual commencement of using the software, it is imperative that the proper preset limits are selected and controls are adjusted.

5.3.1 Preset limits

The user shall set the preset limits such as required real time, live time, total counts, ROI counts, etc. The real time includes the period during which the detector did not send any signal to the MCA, whereas the live time refers to only the period in which the detector was active. The ROI indicates the *region of interest* such as a range of channels within which the peak of the selected radioisotope may fall. In the preset limits ROI counts are set during system calibration. The ROI can be marked by using the keys ALT+M, unmarked by ALT+U, and press ALT+O when finished.

5.3.2 Adjust controls

The software is also used for saving the spectrum files, ROI reports etc in the hard disk or any such data storage device. The most important utility is to control the MCA operation. The following operations are to be carried out only when the system was down or switched off. The following setting is currently followed:

Amplifier Gain	- Fine - 1.0, Coarse:100 [Effective Gain = 100.0]
Shaping time	- Long
Detector Bias	- 3500 Volts

The detector bias is set on by checking in the check box "Enable HV" and then the potentiometer knob located in the left bottom corner in the MCA is operated very carefully and increased at a rate of 10 - 20V per second (It should not be more than 100 V per second in any case, see precautions for more details).

Once the detector bias is raised up to 3500, the potentiometer knob is locked with the locking mechanism provided.

The radio-button in the bottom of the “Adjust Control window screen” is set to ‘Resistive Feedback/Auto PZ’. Place an activity source in the detector area, and then start acquiring the spectrum. Before spectrum is acquired is triggered by putting a radioactive source (may be of activity in mCi) Auto PZ.

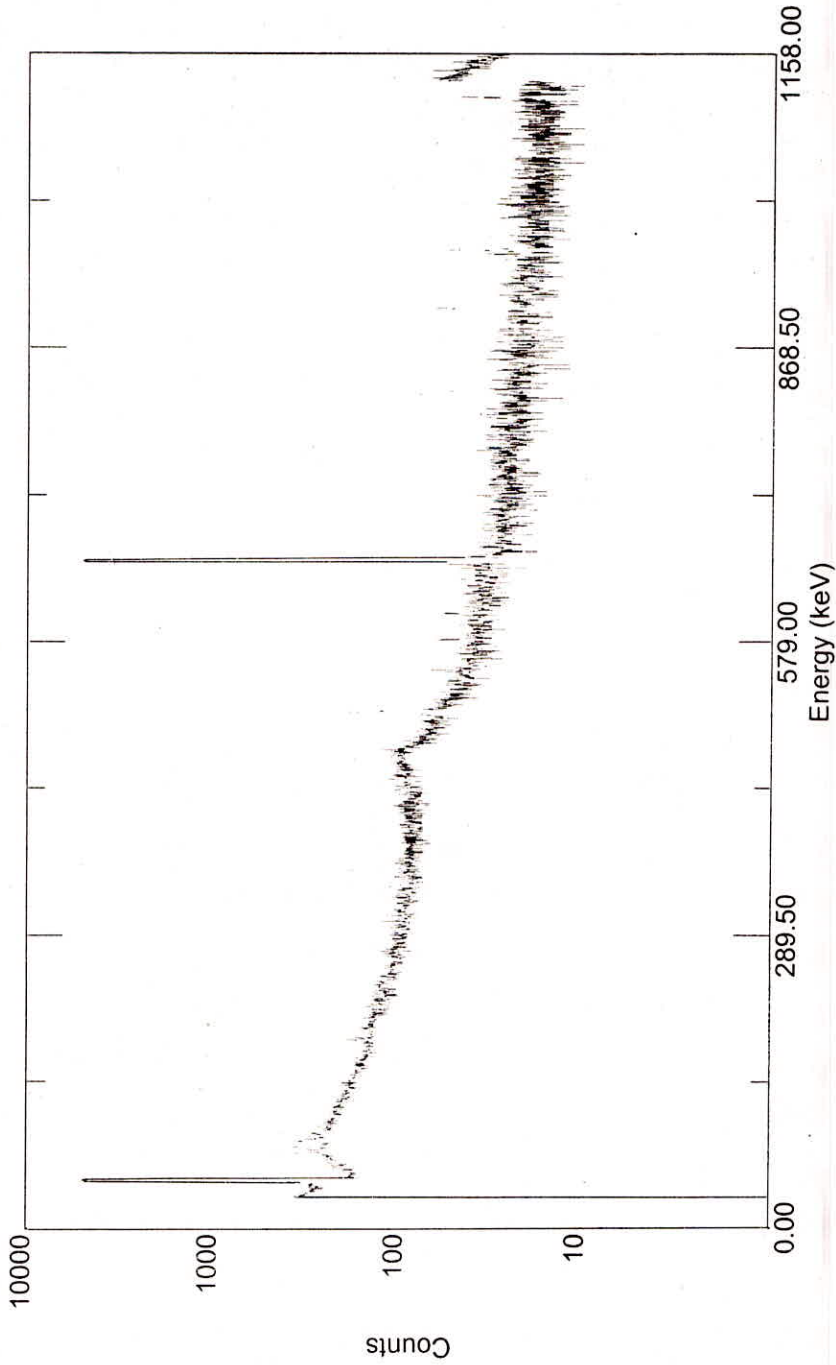
The regular operation involves the following four important steps:

1. Spectrum Stabilisation
2. Energy Calibration
3. Activity Calibration
4. Measuring activity of unknown samples

5.3.3 *Spectrum Stabilisation*

Following the above steps in the “Adjust Controls” Window, the Stabiliser operation is done. The stabiliser is operated only when spectrum is being acquired. The stabiliser operation is very sensitive and it is imperative that the proper protocol be observed for its invocation. At least two sources having an energy range appropriate to the radioisotope of interest is selected. For example, in case of ^{137}Cs , two sources are selected viz., Pb-210 and ^{137}Cs . The gamma energy of Pb-210 is about 45keV and that of ^{137}Cs is 661.62keV. Place the sources in the detector area and start acquiring the spectrum. Mark the ROI for the two selected peaks corresponding to Pb-210 and ^{137}Cs . A typical spectrum for ^{137}Cs and Pb-210 are shown in Figure 3. First the cursor is placed at the left end of the peak of Pb-210. Open the Adjust Controls Window and check the “Stabiliser” button. There are two options in the “Spectrum Stabiliser” window viz., “Zero” and “Gain”. Check the “Initialise” button in the “Zero” option. Return to the spectrum. Place the cursor at the centre of the Pb-210 peak and then check the

Spectrum Calibration



Acquired: 10/7/02 4:48:35 PM
File: C:\LABMAN~1\CS-137\Spectrum Calibration.Chn
Detector: #1 MCB 1
Real Time: 48458.96 s. Live Time: 46825.36 s.
Channels: 8192

“Peak set” button in the “Zero” option of the spectrum stabiliser. Repeat the same operations for “Gain” option in the Spectrum stabiliser window with the ^{137}Cs peak. After, this the stabiliser indicator {STAB} in the MCA panel will glow. It is essential that the Spectrum be acquired continuously during the spectrum stabiliser operation.

5.3.4 Energy Calibration

The system is calibrated with known radioactive sources. The calibration is mainly carried out to improve the spectrum resolution such as matching the channel number with a particular energy peak. As multi point calibration is always preferred over single point calibrations it is recommended that more than two sources be placed in the detector area. Presently ^{137}Cs and ^{210}Pb are being used for the purpose of calibration. Before calibration option is checked, the sources are counted for sufficiently long time so that a minimum of 5000 counts is registered in the respective ROI. Following this, the calibration operation is performed.

Place the cursor in the centre of the ^{137}Cs peak. Select the Calibration command under the “Calculate” menu. In the pop-up window enter the energy of the peak. The corresponding channel number will also be displayed. Place the cursor in the Pb-210 ROI. Again select the Calibrate command and enter the energy of the peak. The energy values are usually entered in keV units. If any error has occurred, use the Destroy Calibration command and do the entire calibration procedure again.

5.3.5 Activity Calibration

The efficiency of low level gamma counting is influenced to a great extent by the sample geometry. Therefore, it is imperative that an exercise be performed to evaluate the efficiency under different sample geometry. One of the chief

causes of concern in the sediment samples is the low sample quantity. Therefore, the activity calibration curve be developed for regular use.

Background

It is essential in radioactive counting that the background counts be known. The background could be due to noise generated by the electronic system and/or due to the radiation emitted by surrounding materials. The background counting is usually achieved by counting a blank sample of same geometry as standards or samples. The gross counts registered in the selected ROI are then considered as the Background. In order to reduce the background counts due to radiation emitted by surrounding materials etc, it is essential to use a thick lead shielding of good quality that should not have background activity in the region of interest. In the set-up at NIH, Roorkee, a two tier lead shielding have been used. Firstly, the detector is kept in the circular lead chamber of 2.5 cm thick wall and secondly, it is surrounded by a thick wall (5cm) of lead bricks.

Gross and Net Counts

Gross counts can be had by right clicking the mouse in the ROI. The gross count data is also stored in the corresponding data file.

Efficiency

The efficiency may be calculated as follows:

$$E = [(G_c - B_g) * 1000] / [72 * A * W_s]$$

E is the efficiency (%); G_c is the gross counts in the ROI; B_g is the Background; A is known activity corrected for radioactivity decay (mBq/g); W_s is weight of the standard used for efficiency estimation (g). The above equation is valid for a counting time (live time) of 7200 seconds.

Presently IAEA-300 Baltic Sea sediment is used as standard at NIH, Roorkee. 10g of the standard is taken in a Tarson petri dish. As the standard is very costly and difficult to obtain it is to be used very carefully and sparingly.

It is essential that the sample for which the activity is unknown as well as the standard used for efficiency calculation are placed in sampler holders of similar dimensions. Further, the geometry of standard as well as the sample be identical. Presently good quality Tarson make TPX petri dishes having a diameter of 75mm (same as the counting area of the HPGe detector) are used. The samples are weighed in a high precision weighing balance (e.g. Mettler AT200)

As noted earlier, the efficiency of counting varies with the amount of sample placed in the counting chamber. A typical calibration curve to account for this variation is shown in the plot of Efficiency versus weight of sample (Figure 4).

5.3.6 Developing protocol

To facilitate better statistical accuracy the counting may be performed by iteration [similar to do-loops in normal computer programming]. For this a macro may be written which is called as *Job Control*. A typical job is as follows:

CLEAR

SET_PRESET_CLEAR

SET_PRESET_LIVE 7200

LOOP 1

BEEP "C:\WINDOWS\MEDIA\THE MICROSOFT SOUND.WAV"

CLEAR

RECALL_ROI "FILE NAME.ROI"

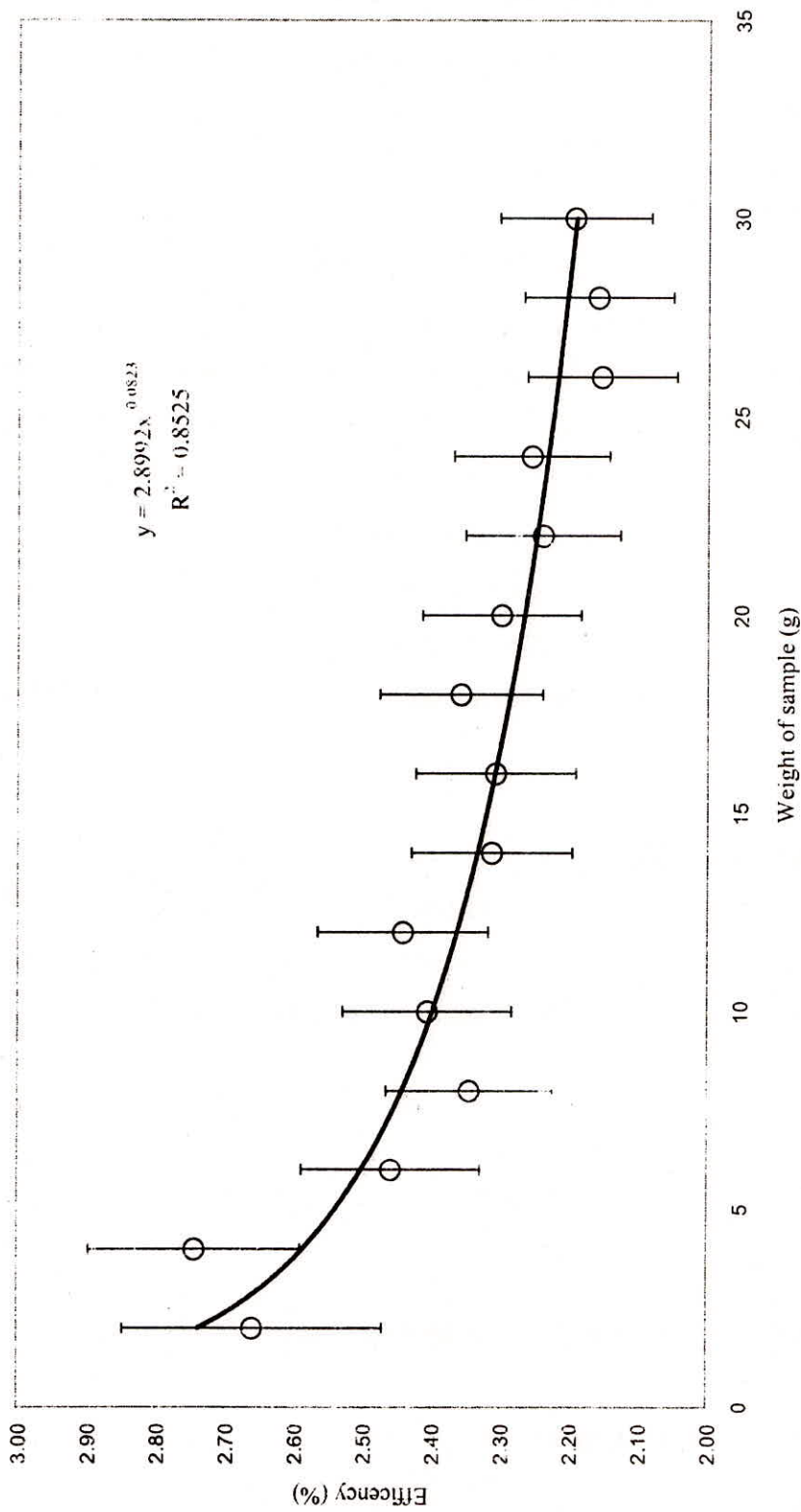


Figure 4. Influence of weight of sample on the efficiency of the HPGe-MCA for measuring the activity of ^{137}Cs (661.62 keV)

START
WAIT
FILL_BUFFER
DESCRIBE_SAMPLE "SAMPLE DISCRIPTION"
SAVE "/PATH/FILENAME.CHN"
REPORT "/PATH/FILENAME.RPT"
END_LOOP
WAIT 1
STOP

5.4 Precautions

Care has to be taken while placing the sample in the detector chamber, through the service window so as not spill the sample in the ultra-clean counting chamber. Non-compliance may contaminate the detector area leading to increased Background.

Saving files or describing the samples in the MAESTRO32 software have to be done carefully.

WARNING: Liquid Nitrogen level in the cryocan has to be monitored regularly. If the combined reading of the two weighing balances at the foot of the cryocan is less than 21 kg, high voltage operation may damage the costly HPGe crystal. The system has to be immediately shut down.

5.5 Estimation of Measurement Errors

The standard error in radioactive measurements may be estimated by:

$$\sigma = \text{SQRT}(G_c/T)$$

where, G_c is the gross counts, and T is the time in seconds. When Net counts are considered the following equation may be used:

$$\sigma = \text{SQRT}(G_c/T + B_g/T)$$

where, B_g is the background counts

6.0 Data Processing

6.1 ^{137}Cs activity

The ^{137}Cs activity in the sediment sample may be estimated by using the following equation:

$$A = [(G_c - B_g) * 1000] / [72 * E * W_s]$$

A is activity of sample (mBq/g);

G_c is the gross counts in the ROI;

B_g is the Background counts in the same ROI;

E is the efficiency (%);

W_s is weight of the sample used for activity measurement (g).

The above equation is valid for a counting time (live time) of 7200 seconds.

Once the ^{137}Cs activity is known for all the samples of a sediment core, then plot the activity against the position of sample in the relevant sediment core. The position may be represented as the depth from mud-water interface in cm. A typical ^{137}Cs activity of a core matches with that of the global fallout pattern shown in figure 5.

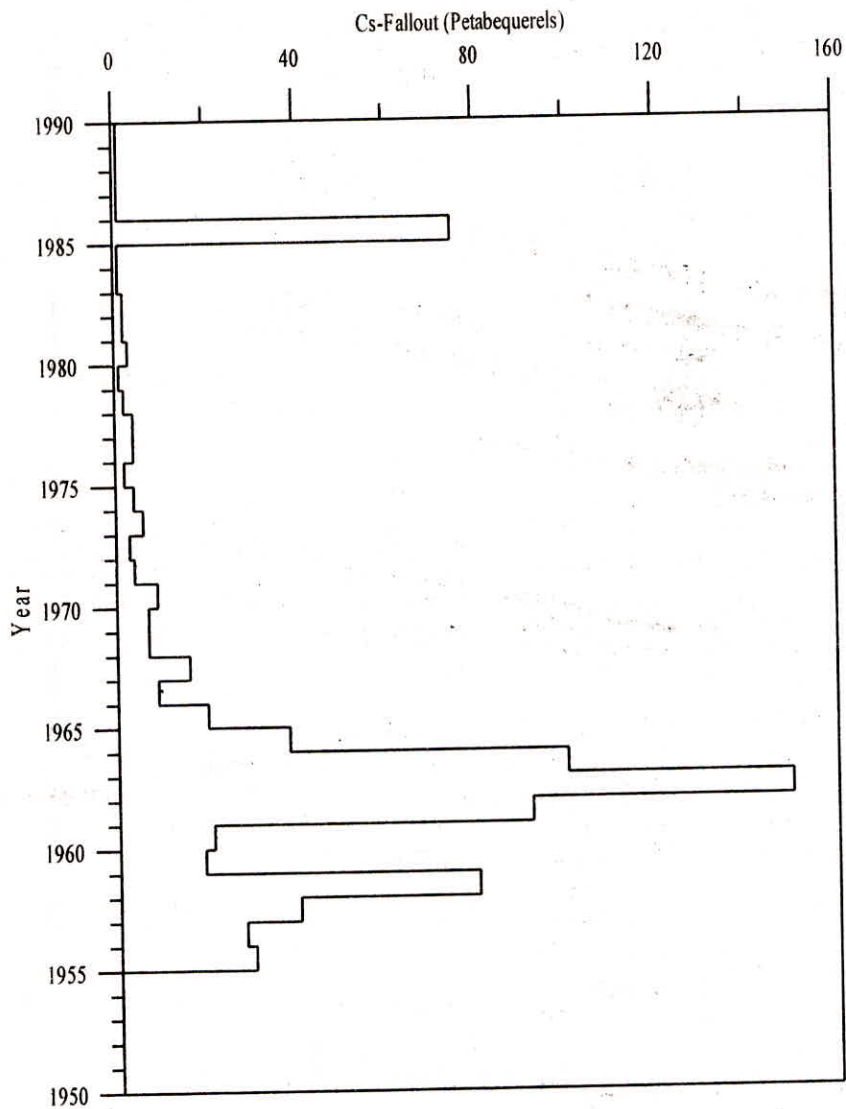


Figure 5. Fallout pattern of ^{137}Cs in the Northern Hemisphere
 [Data Source: Zapata and Agudo, 1996]

6.2 Activity matching

Global studies on ^{137}Cs fall out reveal:

1. initial appearance during 1954/55
2. marked increase during 1958-62
3. maximum fallout during 1963/64
4. perturbations during 1978 (Lop Nar testing)
5. a stand alone peak during 1987/88 (Chernobyl accident)

The last two are not seen in global scale, although widely reported. These activities were not observed in several lakes in India (Lakes Nainital, Mansar, Sagar etc.) However, they were noted in the in the Higher Himalayan region (Lakes Dal, Nagin, and Manasbal located in the Kashmir valley, glaciers located in Sikkim and also in Barapani lake in Megalaya).

The ^{137}Cs activity in a sediment core matches the typical ^{137}Cs fall out pattern as given in Figure 5.

6.3 Estimation of Rate of sedimentation

Once the peaks are identified and matched with fall out pattern as discussed above, the rate of sedimentation may be estimated by using the depth as a marker.

$$R_S = D_P/T_E$$

where, R_S is the rate of sedimentation (cm/y)

D_P is the depth of peak occurrence (cm)

T_E is the time elapsed since the corresponding fallout event (y)

6.3.1 Uncertainty in estimated rates

The uncertainty the estimated rate of sedimentation may be computed by the following relation:

$$\sigma_{R_s}^2 = R_s^2 * \left(\frac{(\sigma_{D_p})^2}{(D_p)^2} + \frac{(\sigma_{T_E})^2}{(T_E)^2} \right)$$

6.4 Estimating the Useful Life of a Lake

The useful life of a lake may be estimated by using the mean sedimentation rate and the mean depth of the lake. Mean sedimentation rate may be estimated by using appropriate weighting factors. The weighting factors may be calculated by using any one of the following methods.

1. By delineating the area represented by a particular sediment core based on the bathymetry such as, deeper zone with comparatively low slope, zone near the lake shore with steeper slope and the intermediate zone. This is particularly applicable for small deeper lakes with small catchment area.
2. By delineating the area represented by a particular sediment core based on the flow direction. The rate of sedimentation decreases with increasing distance from the major inflow point. This is particularly applicable for large shallow lakes with much larger catchment area.

The weighting factors (f) are then the fraction of the area of the delineated portion represented by a particular sediment core to the total surface area of the lake. The mean rate of sedimentation is calculated by the following expression:

$$R_s = \sum_{i=1}^n f_i R_{S_i}$$

where, R_s is the rate of sedimentation (cm/y), f is the weighting factor, subscripts m and n represent the mean rate and the total number of sediment cores considered for the purpose respectively.

6.4.1 Estimation of mean depth of a lake

The mean depth of the lake is calculated by the following expression:

$$D_m = V/A_s$$

where, D_m is the mean depth (m), V is the lake volume (m^3) and A_s the total surface area of the lake (m^2)

6.4.2 Uncertainty in the estimated mean depth of a lake

The uncertainty the estimated mean depth of the lake may be computed by the following relation:

$$\sigma_{D_m}^2 = D_m^2 * \left(\frac{\sigma_V^2}{V^2} + \frac{\sigma_{SA}^2}{SA^2} \right)$$

The useful life, L_U , of the lake is then estimated by:

$$L_U = (D_m * 100) / R_s$$

Where, L_U is the estimated useful life of the lake (in years). Note that in the above expression the unit of the mean depth of the lake is converted from metres to centimetres.

6.4.3 Uncertainty in the estimated useful life of a lake

The uncertainty in the estimated useful life of the lake may be computed by the following relation:

$$\sigma_{L_U}^2 = L_U^2 * 100^2 * \left(\frac{\sigma_{D_m}^2}{D_m^2} + \frac{\sigma_{R_s}^2}{R_s^2} \right)$$

References

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