## SEDIMENTATION STUDIES USING ISOTOPE TECHNIQUES

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# SEDIMENTATION STUDIES USING ISOTOPE TECHNIQUES

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### INTRODUCTION

Sedimentation is one of the serious problems of many lakes causing reduction in lake capacity and useful life of the lake. As such the knowledge of lake sedimentation process is very essential for control and management of the lake sedimentation. It is also essential for understanding the ecological behaviour of lakes. Although high concentrations of suspended matter may result in low primary production because of restricted light penetration, supply of excess nutrients to lake through sediments however, increases its productivity. The availability of dissolved oxygen in lakes may be limited due to high sediment oxygen demand. Benthos and fish are affected not only by the suspended sediments but also by the modification of habitat caused by depositional sediments Sedimentary processes may also cause changes in the lake basin forms. The sediment character controls the chemical composition of the lake water. So, the interaction of sediments with the lake water, both within the water column and at the bed of the lakes, controls the quality of the surface water. Sediment also contains biological life. It is a substrate for bacterial activity.

Nature of sediments coming to a water body depends upon the rock type in the drainage basin. Siltation rates are mainly dependent upon conditions in the drainage area. Other factors include morphological and hydrodynamic factors. Effects of drainage basin are mainly due to morphology, climate and land use. There are three main sources of sediments for lakes. Water and wind are the main natural agents which bring sediments into the lake. Organic matter produced in the lake is an internal source. Besides, there is also a problem of resuspended bottom material.

The bedload brought by the river are initially deposited preferably on shallow water near the shores. Fine particles have low settling velocities and are kept in suspension by the water motion. Fine material from the lake bottom is eroded and brought into suspension by influence of waves and currents. The shallow bottoms from which fine loose material is stirred up at every storm and where there are no extended periods of deposition are called erosion bottoms. Transportation bottoms prevail where fine material is deposited but where erosion takes place during severe storm. Areas where fine material is deposited and erosion never occurs are called accumulation bottom. In shallow lakes there are no accumulation

bottoms where material can deposit. The bottom sediments close to the shores as well as in the central parts of such lakes are a mixture of fine and coarse material.

Resuspension of the settled sediments is basically governed by the balance between hydrodynamic forces seeking to move the particle and the stabilizing forces due to gravity. Wave action is the dominating force for resuspension in lakes. Once the sediments have been resuspended, they will be redistributed in the lake according to the current pattern and diffusive character of the flow. When the energy level is reduced due to decrease in the wind speed, then the sediments will settle to the bottom. The result is that basically the near shore areas will be void of fine sediments. They will gather in the deeper areas. The shape and depth of the lake basins affect the circulation of water and thus the distribution and deposition of fine sediment particles.

The amount of sediment carried by the incoming river can be measured or estimated from models. However, the amount of locally produced material and resuspeded sediments can not easily be determined. Long term sedimentation in a lake is best determined by comparing bathymetric maps from different years. The sediment rate over the shorter periods can be determined using sediment traps. These can be bottom placed or floating. To get representative values for the lake, they should be placed on accumulation bottom and for the floating ones, 1-2 m above the bottom. Isotopes techniques are, however, more precise compared to the conventional methods and are hence, becoming popular recently for estimation of sedimentation rates in lakes.

## METHODOLOGIES FOR ESTIMATION OF SEDIMENTATION RATE OF LAKES

There are various techniques for the determination of the rate of sedimentation in lake/reservoirs, such as range line method, contour method, sediment balance method, remote sensing techniques and radiometric dating techniques. These are discussed in brief in following sections. The <sup>210</sup>Pb and <sup>137</sup>Cs dating techniques have been discussed in detail.

## Range Line Method

The range-line method is widely used for medium to large lakes and reservoirs requiring an underwater survey utilising hydrographic surveying methods. In range line method, a number of cross sections, called ranges, of the lakes are surveyed and then it is periodically repeated at the same cross sections. Specific details concerning the method can be found in many references (Vanoni, 1977; and Guy, 1978). Basically, the deposition of sediment during a period is estimated by measuring the depth to bottom (water column) at different locations, which can be compared to a previously constructed map to determine differences in the volume of sediment deposited.

#### **Contour Method**

The contour method uses essentially topographic mapping procedures (Wolf, 1974). To apply this method, it is important to have a good contour map of the lake when it is dry (Pemberton and Blanton, 1980).

The procedure for either of the methods involves the determination of bed elevation at many known locations in the lake/reservoir.

#### **Sediment Balance Method**

Sediment is carried into a lake by streams and rivers as well as by overland flow entering a lake. Sediment entering a lake may consist of a wide range of sizes, from gravel or boulders to silt and clay particles.

The surest way of obtaining an accurate determination of the amount of sediment being carried to a lake by streams is to measure the flow rate and sediment concentration of the inflowing waters just upstream of the lake. Ordinarily, records of sediment discharge (fine, coarse, suspended, or bedload) are determined on the basis of sample information obtained at non-uniform intervals of time. The information from the sediment samples can be used to estimate the total sediment discharge by developing a relation between the sediment discharge and water discharge, called a sediment transport curve. The sediment outflow from the lake, if any, is computed in the discharges from the lake. The difference of input and output sediment divided by lake area gives the sediment deposition rate.

## **Remote Sensing Techniques**

Remote sensing has several main applications in the assessment of lake/ reservoir sedimentation. Contour maps prepared from aerial photographs can be used to determine sediment volumes provided the water level can be lowered greatly. Aerial photography can be used to trace turbidity plumes, which may help in defining the distribution of sedimentation. Digital image processing of high-resolution satellite data can also be used for lake sedimentation studies. The information of suspended sediments obtained at different times using this technique can be utilised to predict the deposition or settling rate of sediments in the lake.

A comparatively new technique, which is being developed by the U.S. National Ocean Survey, is to use laser hydrography (Enabnit et al., 1979). The airborne laser hydrographic technique uses an aircraft mounted pulsed laser system to collect a swath of discrete soundings along each flight line. It measures water depth exactly like a sonar using light instead of sound.

## Radiometric Dating Techniques

Several environmental isotopes including <sup>210</sup>Pb, <sup>137</sup>Cs and <sup>14</sup>C find applications in the estimation of rate of sedimentation in lakes. However, <sup>14</sup>C is more useful for paleohydrological studies (Kusumgar et al., 1992). Artificial radio-isotopes used for sediment accumulation studies include <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>241</sup>Am. However, for the dating of recent sediments, <sup>210</sup>Pb (100 to 150 years BP) and <sup>137</sup>Cs (post 1954) are widely used.

## RADIOMETRIC DATING TECHNIQUES

Lake sediments contain radioisotopes from both natural and artificial sources. Natural radioisotopes are generated by cosmic ray interactions of the earth's atmosphere while the artificial radioisotopes have been introduced into the environment as a result of the testing of atomic weapons and accidents involving nuclear power installations. Few radioisotopes of geogenic origin also escape into the atmosphere and later on naturally fallout with the precipitation and other decay processes. Out of all the radioisotopes that occur naturally and artificially in the environment, <sup>137</sup>Cs (Cesium-137) and <sup>210</sup>Pb (Lead-210) have been found very useful for the dating of the sediments that deposit in the lakes and reservoirs. The natural fall out of <sup>137</sup>Cs was found considerable during the years 1953-54, 1957-58, 1963-64, 1978-79. 1986-87 due to testing of various atomic devices and nuclear accidents. The <sup>137</sup>Cs activity that appeared during the 1986-87 was caused by the Chernobyl Nuclear Reactor accident while the peak corresponding to 1978-79 is due to atomic devices tested by China. These peaks are used as a marker/indicator for the estimation of rates of sedimentation in water bodies. Thus, keeping in view the potential for the estimation of recent to compararively old sedimentation rates, <sup>210</sup>Pb (100 to 150 years BP) and <sup>137</sup>Cs (post 1954) dating techniques are widely used.

The undisturbed sediment samples are collected from the bed of the lake/reservoir from the desired location with the help of a gravity type core sampler, up to the depth of 20 to 100 cm. Some practical difficulties associated with the collection of undisturbed bed sediment samples are as follows:

- i) The soft sediments in upper 20 cm. (may be more) depths may be disturbed during collection (penetration) and it may vary with different degree of compaction with increasing depth.
- ii) The sediment sample may be disturbed during removal with the help of core cutter (tube) due to suction effect.

The core tube (made of transparent type plastic material) ranges up to 1 metre in length and 5 cm in diameter, can be used for the collection of sediment samples from the bed using a hand corer, or a piston or box corer. However, gravity core sampling is convenient. Mackereth corer - a remotely operated pneumatic covers can also be used for this purpose.

Cores should be kept at or near in situ temperature to prevent expansion due to gas formation. Strong and Cordes (1976) have described a cloth sleeve filled with dry ice as a means of freezing cores. If this or a similar cooling procedure is not available in the field, the cores have to be sectioned as soon as possible.

The sediment samples (cores) are cut into sections of appropriate thickness (1-2 cm) with the help of an adjustable piston rod with silicone packing and packed in a polythene bag. An aliquot (say 2-5 grams) of the sliced core sections were used for measuring physical characteristics of the soil sample, including bulk density, water content and porosity and the remaining of the sliced core sections were freeze dried and stored in the laboratory for the radionuclide measurements (i.e. for <sup>137</sup>Cs and <sup>210</sup>Pb). Measurements are made for the bulk

density before drying samples in an oven at a temperature slightly above 100°C for about 7 to 8 hours prior to analysis. The dry unit weight, water content (determined as the difference in weight between the wet sediment sample and dry sediment sample, and expressed as percent of the weight of the wet sediment sample), and porosity (calculated as ratio of volume of water in the sediment pores to total volume of the sliced core sample, and expressed in percent) are also estimated after drying. The textural composition of the core samples is also determined.

The core sections are analyzed by gamma ray spectrometry using Ge(Li) or intrinsic Germanium detectors (HpGe type) directly for <sup>137</sup>Cs activity measurement while <sup>210</sup>Pb activity measurement requires a chemical treatment. The detailed procedures are discussed below under the respective sections.

## <sup>210</sup>Pb Dating Technique

This method is very popular and has been applied in several lake studies, the world over. The basic principle is discussed below in brief. In uranium ( $^{238}$ U) decay series, radium ( $^{226}$ Ra) emits radon ( $^{222}$ Rn) that escapes into the atmosphere. Further decay of radon gives rise to  $^{210}$ Pb and other short-lived daughter products in the atmosphere, which are removed through aerosols by dry fallout and precipitation. The decay chain of  $^{238}$ U is given below with the half-lives of decay products.

$$^{238}U \longrightarrow ^{226}Ra \longrightarrow ^{222}Rn \longrightarrow ^{218}Po \longrightarrow ^{214}Pb \longrightarrow ^{214}Bi$$
4.5e9 a 1620 a 3.8d 3.05 m 26.8 m

(Stable) $^{206}Pb \longleftarrow ^{210}Po \longrightarrow ^{210}Pb \longrightarrow ^{214}Po$ 
138d 22.26 a 1.6e-4s

The residence time for <sup>210</sup>Pb in the lower atmosphere is less than a week (Poet et al. 1972). The <sup>210</sup>Pb flux from atmosphere to land and water surfaces is a continuous process. In other words, most of the <sup>210</sup>Pb entering the surface water body come from direct atmospheric input across the surface. They may also find their way through indirect atmospheric fall-out or by radon decay in water body (Eakins, 1983). The <sup>210</sup>Pb thus entering the lake is removed rapidly by suspended particulate matter and accumulate as lake sediments. This <sup>210</sup>Pb is termed as unsupported <sup>210</sup>Pb or <sup>210</sup>Pb <sub>excess</sub>. However, since <sup>210</sup>Pb is also produced in situ (supported <sup>210</sup>Pb) as a result of the decay of terrigenous <sup>226</sup>Ra present in the lake sediments, the activities of both supported as well as unsupported <sup>210</sup>Pb must be known for using the technique.

Over the past twenty years, <sup>210</sup>Pb dating has developed into a major tool for paleolimnologists, historians, geographers and environmental scientists. Many coring techniques are available, ranging from the simple to the quite complex, but all have been shown to give good results. The analytical methods for <sup>210</sup>Pb and <sup>226</sup>Ra are well established and can be carried out by a competent analyst in any reasonably equipped laboratory.

Two main approaches to the interpretation of <sup>210</sup>Pb sediment profiles are currently in use, the constant initial concentration (CIC) model and the constant rate of supply (CRS) model. These often yield similar chronologies for the more recent sediments, but there is sometimes

divergence in the time scales relating to deeper deposits. There are no definitive criteria for choosing one model in preference to another. Each sediment profile should be considered in relation to all available data, to other chronologies and, wherever possible, to other sediment cores taken from the same water body.

The applications of <sup>210</sup>Pb dating are many and varied. A sediment core records a detailed history of the environment in its vicinity and the <sup>210</sup>Pb dating technique provides a chronology covering a time scale of 100-150 years, uniquely suited to the period of man's greatest impact.

<sup>210</sup>Pb dating depends on the accurate determination of the level of unsupported <sup>210</sup>Pb in a series of sediment samples. In order to do this; the total <sup>210</sup>Pb content of each sample is determined initially. The supported <sup>210</sup>Pb is then measured by analysis for its precursor and <sup>226</sup>Ra and the unsupported <sup>210</sup>Pb derived by subtraction of this value from the total <sup>210</sup>Pb. Ideally, all sections of a sediment core should be analysed for <sup>210</sup>Pb and <sup>226</sup>Ra until a depth is reached at which the total <sup>210</sup>Pb is indistinguishable from the supported <sup>210</sup>Pb. However, for economic reasons this is not normally done and it is usual to determine <sup>210</sup>Pb in about 10 sections between the surface and the region where the unsupported <sup>210</sup>Pb is no longer detectable. If the <sup>226</sup>Ra content is relatively constant, then it is generally sufficient to analyse sections at the top, middle and bottom of the region of interest to establish the supported <sup>210</sup>Pb. If the <sup>226</sup>Ra is variable however, then all sections analysed for <sup>210</sup>Pb must also be analysed for <sup>226</sup>Ra.

## Determination of <sup>210</sup>Pb activity

The <sup>210</sup>Pb activity is measured either through direct beta counting or through the alpha counting of its granddaughter, namely <sup>210</sup>Po, which is assumed to be in secular equilibrium with its parent. The direct measurement of <sup>210</sup>Pb in sediment by detection of its 47 KeV photon emission has been reported by Gäggeler et al. (1976). This method has the advantage of being non-destructive and the sediment sample can be used for other measurements. However the 47 KeV emission from <sup>210</sup>Pb is only present in 4 per cent of its disintegration and the method is relatively insensitive. The limit of detection is about 2 pCi/g for a sample of about 100 g, which seriously limits its application to many cores where the concentration of <sup>210</sup>Pb, even in the surface sediments, may be less than 5 pCi/g. In some core sections, the weight of sediment available for analysis is only a few grams and this will further increase the limit of detection. However, the method may well have application in determining the total <sup>210</sup>Pb in a core where no profiling is required. In this case, the whole core should be homogenised to take a sufficiently large sample for measurement.

Because of the low energy of its beta emission <sup>210</sup>Pb is often determined by means of its <sup>210</sup>Bi daughter. This has a half-life of 5 days and emits a beta particle with E<sub>max</sub>1.16 MeV. A technique used by many workers (Krishnaswamy et al., 1971 and Koide et al., 1973) for determining <sup>210</sup>Pb in sediment is as follows. A sample of dried sediment is leached with hydrochloric acid and then the <sup>210</sup>Pb along with the added lead carrier is removed from the resultant solution by anion exchange. The lead is then precipitated, either as sulphate or chromate, and a suitable source prepared for beta counting. The chemical recovery is determined gravimetrically and the source is left for about 5 weeks before counting to allow

the <sup>210</sup>Bi to equilibrate with its parent <sup>210</sup>Pb.

An alternative procedure is to determine the <sup>210</sup>Po granddaughter of <sup>210</sup>Pb. This has a half-life of 158 days and decays to stable <sup>206</sup>Pb, emitting an alpha particle of 5.3 MeV. As deposition rates are generally less than 1 cm/y, the <sup>210</sup>Po will be in equilibrium with <sup>210</sup>Pb in all but the surface layer. A number of workers have adopted this approach, including Robbins and Edgington (1975), and Krishnaswamy et al. (1980). The basic radiochemical procedure is to add <sup>208</sup>Po as a yield tracer, wet oxidise or leach the sediment sample with strong mineral acids, filter off residual solids and convert the solution to one of dilute hydrochloric acid. Polonium nuclides are then spontaneously deposited on silver discs prior to counting by alpha spectrometry. Eakins and Morrison (1983) employed a different procedure, in which <sup>210</sup>Po is dry distilled from the sediment as the volatile tetrachloride prior to deposition on silver. There are a number of advantages in determining <sup>210</sup>Pb via <sup>210</sup>Po. Alpha counting is inherently more sensitive than beta counting and the <sup>210</sup>Po can be identified unequivocally. There is no need of any delay between preparation of the source and counting, and the separation of <sup>210</sup>Po from sediments is relatively simple.

Another technique for determining <sup>210</sup>Pb has been described by Jensen et al. (1977), in which <sup>210</sup>Pb is extracted from a sample of ashed sediment with nitric acid and electrodeposited on platinum. After allowing for ingrowth, <sup>210</sup>Po is detected by alpha track counting using a plastic detector. This method is very sensitive but lacks the specificity of alpha spectrometry.

## Determination of <sup>226</sup>Ra activity

There are a number of approaches to determine <sup>226</sup>Ra in sediments. If the concentration is fairly high and the sample is large, <sup>226</sup>Ra can be determined directly by gamma counting of its decay products. Radiochemical methods fall in two groups, those using <sup>222</sup>Rn emanation technique to detect <sup>226</sup>Ra daughters and those resulting in a solid source which is alpha counted.

In <sup>222</sup>Rn emanation technique, a solution of <sup>226</sup>Ra is first obtained from the sediment either by acid leaching or fusion and subsequent dissolution. The solution is stored in a closed system to allow ingrowth of <sup>222</sup>Rn, which is then flushed out with an inert gas into a scintillation cell. The cell is stored for a few hours before being coupled to a photomultiplier tube and then counted. There are different procedures for producing a solid source for alpha counting depending upon the method of obtaining a solution of <sup>226</sup>Ra from the sediment. <sup>226</sup>Ra is then separated from impurities by a variety of methods before a source is prepared for alpha counting. Koide and Bruland (1975) used <sup>226</sup>Ra as a yield tracer, coprecipitating the radium with lead nitrate. The lead is then removed by anion exchange and the radium is further purified by cation exchange. The purified radium is electrodeposited from a 2-propanol electrolyte onto a platinum disc prior to alpha spectrometry. The <sup>226</sup>Ra can be coprecipitated with lead sulphate and subsequently with barium sulphate in the presence of ethylene diamine tetra-acetic acid. The barium sulphate is purified by reprecipitation and its alpha activity determined after storing to allow ingrowth of the <sup>226</sup>Ra daughter activities.

The chemical recovery is determined using <sup>133</sup>Ba as a gamma yield tracer. Joshi and Durham (1978) coprecipitated <sup>226</sup>Ra with lead and barium carriers from strong nitric acid and then separated the barium and <sup>226</sup>Ra from lead by ion exchange. The <sup>226</sup>Ra is subsequently

coprecipitated with barium chromate which is stored prior to alpha counting.

## Interpretation of <sup>210</sup>Pb profiles

The interpretation of a <sup>210</sup>Pb sediment profile and its conversion into one of life versus depth depends on assumptions concerning the supply of <sup>210</sup>Pb to the sediment column and its behaviour within it. It is generally assumed that the supply of atmospheric <sup>210</sup>Pb to the water surface and the catchment is constant on a time scale of 100-200 years. The flux may vary on time scales of the order of a year but, as sediment sections taken for analysis normally span several years of accumulation, these short-term variations will be smoothed out. It is also assumed that there is no migration or diffusion of <sup>210</sup>Pb within the sediment column through the pore water. There is indirect evidence that migration is minimal from the fact that in some profiles there are sharp peaks and inflexions which would be smoothed if migration was significant. Similarly, it is assumed in determining the supported <sup>210</sup>Pb that both <sup>210</sup>Pb and <sup>226</sup>Ra (which is actually measured) are in equilibrium. This has been confirmed from the determination of <sup>210</sup>Pb and <sup>226</sup>Ra in the lower sections of over a hundred cores where the unsupported <sup>210</sup>Pb is no longer detectable. However, Imboden and Stiller (1982) have discussed the influence of <sup>222</sup>Rn diffusion on the <sup>210</sup>Pb distribution in sediments and have produced a mathematical model to describe the distribution of <sup>222</sup>Rn within a core. They believe that in cores having a low unsupported <sup>210</sup>Pb content, the assumption that <sup>226</sup>Ra and <sup>210</sup>Pb are in secular equilibrium should be treated with caution. There may well be a small disequilibrium between <sup>210</sup>Pb and <sup>226</sup>Ra near the sediment- water interface but, if the unsupported <sup>210</sup>Pb is in considerable excess, the effect on dating will be negligible.

## Models used for <sup>210</sup>Pb profiles interpretation

Two main approaches to the interpretation of <sup>210</sup>Pb sediment profiles are currently in use, the constant initial concentration (CIC) model and the constant rate of supply (CRS) model. These often yield similar chronologies for the more recent sediments, but there is sometimes divergence in the time scales relating to deeper deposits. There are no definitive criteria for choosing one model in preference to another. Each sediment profile should be considered in relation to all available data, to other chronologies and, wherever possible, to other sediment cores taken from the same water body.

#### Constant initial concentration (CIC) model

The <sup>210</sup>Pb dating technique was first applied to lake sediments by Krishnaswamy et al. (1971) and to marine sediments by Koide et al. (1973). These and many other workers found that a semi-logarithmic plot of the total <sup>210</sup>Pb concentration per unit weight of sediment against depth showed a roughly monotonic decline until a region of unchanging concentration was reached. The region of unchanging concentration is the supported <sup>210</sup>Pb maintained by <sup>226</sup>Ra within the sediment. Subtraction of the supported <sup>210</sup>Pb component from the profile results in the lower curve, which is a plot of the unsupported <sup>210</sup>Pb against depth. The unsupported <sup>210</sup>Pb decreases exponentially and the slope of the line represents a mean sedimentation rate. By a comparison of the concentration at any depth with that at the surface, the life of the sediment at that depth can be calculated from the equation for radioactive decay, which in this case can be expressed as:-

$$C_{d} = C_{0} e^{-\lambda t} \qquad \dots \dots (1)$$

where,

 $C_d$  = concentration of  $^{210}$ Pb at depth d

 $C_0$  = concentration of <sup>210</sup>Pb at the surface

 $\lambda$  = decay constant for <sup>210</sup>Pb (0.031)

t = life of sediment at depth d.

This model assumes that, over the time scale being studied, the concentration of <sup>210</sup>Pb in sediment at the mud -water interface has been constant. Because of this, it is often referred to as the constant initial concentration or CIC model. It has been validated for marine sediments by the analysis of varied cores from the Santa Barbara Basin (Koide et al., 1972). In some profiles, a semi-logarithmic plot of the unsupported <sup>210</sup>Pb against depth is predominately linear, but the slope decreases near the surface. This is often simply the result of reduced compaction of near-surface sediments and the non-linearity is avoided by plotting the <sup>210</sup>Pb concentration against the cumulative dry mass of sediment. This technique has been adopted by many workers and has largely superseded the earlier procedure. Methods for assessing the effects of compaction on the calculation of sediment accumulation rates in the near-surface sediments of a core have been discussed by Robbins and Edgington (1975).

Non-linearity in a <sup>210</sup>Pb profile near the surface of a core will also occur if the surface sediments are disturbed. The most common cause of sediment mixing is bioturbation, which normally affects only the top few cm of a core. The redistribution of surface sediments by deposit-feeding organisms has been investigated and found that beneath the zone of mixing, the unsupported <sup>210</sup>Pb profile in a core of constant sediment accumulation rate was linear. Bioturbation is normally less in freshwater sediments than those of marine origin, partly because of the small size of the freshwater benthos. Skei (1979), in a study of sediments in the Norwegian fjords, found that anoxic sediments were in general suitable for <sup>210</sup>Pb dating but that toxic sediments were often too disturbed by bioturbation.

A further source of non-linearity in a <sup>210</sup>Pb profile is a change in the accumulation rate. During the last 50-100 years, the sediment accumulation rate in many water bodies has increased, often due to eutrophication. One of two assumptions is implicit in the use of the CIC model to interpret such a profile:-

- (1) There is a large excess of <sup>210</sup>Pb in solution in the overlying water and increased sedimentation merely entrains more of the excess. If the sediment has a limited capacity for <sup>210</sup>Pb this will result in its concentration remaining constant despite a change in accumulation rate.
- (2) The main source of <sup>210</sup>Pb in sediment is material uniformly labelled with <sup>210</sup>Pb on the catchment and more of this is being transferred to the lake because of environmental change.

Pennington et al. (1976) used the CIC model to interpret non-linear <sup>210</sup>Pb profiles of a series of cores from Blelham Tarn. The <sup>210</sup>Pb chronology was consistent with both <sup>137</sup>Cs and paleomagnetic dating and gave similar ages for visible stratigraphic changes in sediments of

different cores. However the use of the CIC model to interpret <sup>210</sup>Pb profiles where the sediment accumulation rate is changing does not always yield a chronology consistent with independent time scales and an alternative interpretation has been sought.

## Constant rate of supply (CRS) or constant flux (CF) model

In equation (1) discussed under section 3.1.6,  $C_0$ =F/R where F is the flux of unsupported  $^{210}$ Pb to the sediment/water interface and R is the sediment accumulation rate. A change in the accumulation rate implies a change in either  $C_0$  or F (or possibly both). The CIC model assumes that  $C_0$  is constant but an alternative approach is to consider that F, the flux of unsupported  $^{210}$ Pb to the sediment, is constant. This is termed the constant rate of supply or CRS model. It was first outlined by Goldberg (1963) and has since been developed by Appleby and Oldfield (1983). The constant flux assumption implies a constant residual of unsupported  $^{210}$ Pb within the sediment column and the life t of sediments at depth d may be calculated from the radioactive decay law expressed as

$$\mathbf{A}_{\mathsf{d}} = \mathbf{A}_0 \; \mathsf{e}^{-\lambda \mathsf{t}}$$

where  $A_d$  is the unsupported  $^{210}\text{Pb}$  in the core below depth 'd' and  $A_0$  is the entire unsupported  $^{210}\text{Pb}$  below the mud/water interface. The varying sediment accumulation rate r can be calculated from

$$r = A_d / C_d$$

where C<sub>d</sub> is the unsupported <sup>210</sup>Pb concentration. Oldfield et al. (1978) have used this model to reassess <sup>210</sup>Pb data for several lakes, and have obtained chronologies more consistent with independent dating methods than those obtained using the CIC model. Appleby et al. (1983) have validated the method in freshwater sediments by dating varved cores from Finland and Batterbee et al. (1980) have reassessed the <sup>210</sup>Pb chronology of Lake Växjösjön on the basis of a CRS model. The general effect of applying the CRS model is to assign greater ages to sediments at lower depths in a core, whereas there is often reasonable agreement between the chronologies for near-surface sediments. It should be noted that both the CIC and CRS models will give the same chronology if a semi-logarithmic plot of unsupported <sup>210</sup>Pb against the cumulative dry mass of sediment is linear. The other model can be a combination of both, i.e., constant initial concentration and constant flux or constant flux and constant rate of sediment (CFCS).

### Selection of a model

Although the CRS model has led to a reassessment of many <sup>210</sup>Pb sediment profiles, it does not have universal application and it is not always clear which model should be used. Oldfield and Appleby (1983) have found empirically that, if the mean flux of unsupported <sup>210</sup>Pb to the sediment falls within the range expected from measured atmospheric fluxes (0.2-1.0 pCi/cm²/y) the CRS model will give the more valid chronology but, where fluxes are much less than the atmospheric range, results have been poor. Oldfield and Appleby recommend analysis of more than one sediment core wherever possible, to indicate whether sediment resuspension and focusing are occurring. This will lead to reduced unsupported <sup>210</sup>Pb at sites where sediment erosion has taken place and enhanced deposits where focusing

has occurred. In such cases, although the CRS model will not be valid for a single core, it may be valid for the lake bed as a whole.

The CRS model is used for the near-surface sediments where the accumulation rate may be changing and the CIC model for the deeper sediments where it is constant. If a horizon in a sediment core can be dated independently, for example by <sup>137</sup>Cs, pollen or magnetic measurements, or by an input from a known historical event such as ash from a forest fire or a volcano, then the model which gives the best fit to the independent chronology should be used.

#### Limitations and Uncertainties

In some circumstances it is impossible to date a sediment core by the <sup>210</sup>Pb technique, whichever model is used. A very high supported <sup>210</sup>Pb content can completely mask the unsupported <sup>210</sup>Pb derived from the atmosphere. Occasionally a sediment will have so little unsupported <sup>210</sup>Pb that it is difficult to distinguish it from a 'normal' supported <sup>210</sup>Pb content.

The reason for a very low unsupported <sup>210</sup>Pb content in sediment from a water body where there is no obvious sediment loss is not known, but it may be significant that it has only been observed in lakes with small catchment areas. This suggests that unsupported <sup>210</sup>Pb in a lake sediment may be derived predominately from the catchment. Perhaps the biggest uncertainty in <sup>210</sup>Pb dating is the extent to which the catchment contributes to the unsupported <sup>210</sup>Pb in sediment and this is an area, which warrants further study. A further area of uncertainty lies in the distribution of <sup>210</sup>Pb within the various components of sediment. Cooper et al. (1981) in a study of the speciation of radionuclides in sediments and soils found that over 30 per cent of the <sup>210</sup>Pb activity was bound to organic matter. There is therefore mounting evidence that <sup>210</sup>Pb is associated with organic material in sediments, but further work is required to confirm this.

## 137Cs Method for Sediment Dating

<sup>137</sup>Cs is produced in the atmosphere due to cosmic ray interactions. However, its concentration increased many folds in the atmosphere due to the test of nuclear weapons and since 1954, it has been globally detectable. <sup>137</sup>Cs is strongly absorbed on tiny particles like clay materials, silts and humic materials. Surface soils with an adsorptive capacity will have a <sup>137</sup>Cs content and therefore be able to act as a self tracer. In a catchment, accumulation of a sediment layer in a lake is a measure of its trap efficiency. A comparison of <sup>137</sup>Cs of catchment soils with that of associated lake sediment shows a pronounced build up of the latter. The rates of sedimentation can be calculated from the depths of two principal time horizons i.e. 1954 and 1964, in the <sup>137</sup>Cs concentration profile. Presently, this has been considered as more reliable technique for the dating of sedimentation rate in past 40 years.

## Temporal variations in <sup>137</sup>Cs fallout

The principal sources of information on <sup>137</sup>Cs levels in fallout are the various reports of measurements from a global network of monitoring stations (Cambray et al., 1980; US Health and Safety Laboratory, 1977). Supplementary sources include reports on individual

national measurements (Bonnyman et al., 1972; Baltakmens and Gregory, 1977). The pattern of annual deposition at Australian stations has found the following principal features:

- (i) First appearance of <sup>137</sup>Cs in 1953/54 and second of significant amount in 1957/58;
- (ii) maximum fallout in 1963/64;
- (iii) marked decrease in rate of deposition from 1959 until 1962, which appears as a minor maximum; and
- (iv) considerable fallout in 1978-79 and 1986-86 due to Lopnor atomic test of atomic and Chernobyl accident.

It is known that uptake of fallout by soils and sediments is rapid (Eyman and Kevern, 1975), and it follows that surface soil minerals have been labelled continuously at levels which depend on the prevailing concentration of <sup>137</sup>Cs in the total fallout. It must be remembered, however, that after the peak in fallout the integrated source function of <sup>137</sup>Cs does not follow the pattern of atmospheric fallout which has decreased dramatically. Processes resulting from the overlaying of an original 1954 interface with sediment material lead to the formation of a <sup>137</sup>Cs concentration profile that relates to the annual variations in atmospheric fallout. The preservation of these structured concentration changes provides at least two time markers (dates of first appearance and maximum fallout) that are the basis of an absolute geochronology of these sediments.

#### Measurement of sediment redistribution with <sup>137</sup>Cs

The transport and spatial distribution of sediments entering an impoundment are functions of the balance between the flow velocities, gravitational forces and the secondary forces of flow turbulence. In the case of reservoirs, drawdown procedures can be a very powerful initiator of sediment flows and redistribution. Three generalised zones of sedimentation are given for reservoirs (Wiebe and Drennan, 1973; for sedimentary process in lakes, see Sly, 1978):

- (i) The upper zone in which complex deltas form as a result of flows entering the reservoir retaining their identity for some distance into the reservoir pool. The deltas grow outward by the formation of forest (longitudinal flow) beds and upward through topsets beds.
- (ii) The intermediate zone in which the residual river velocity, waves and wave-induced currents transport and deposit most of the river's wash load, and some of the fine sediments eroded from the banks of the reservoir, to form bottom set beds of fine clays, silts and colloids.
- (iii) The lower zone containing sediments eroded from the reservoir banks and transported by waves and wave-induced currents.

The relevance of these sediment classifications can be seen in the work of Simpson et al. (1976) in the Hudson River estuary, New York. They were able to classify three main types of distribution of <sup>137</sup>Cs in estuary sediment cores:

(a) Relatively low activities (19 mBq/g) generally confined to the upper 5-10 cm of sediment and associated with subtidal banks.

- (b) Higher activities than in (a) of about 90 mBq/g in the top 10 cm decreasing rapidly to about 7 mBq/g in the 10-15 cm layer and sometimes distributed down to 40 cm with an activity of about 1 mBq/g. This type of profile was interpreted as being indicative of a high sedimentation rate in a shallow protected environment.
- (c) Profiles with variable but high activities of about 70 mBq/g down to 40 cm and, in one case, down to 250 cm. <sup>137</sup>Cs activity below 40 cm was stated by Simpson et al. (1976) to be a clear indicator of rapid sediment accumulation. It is speculated that temporal changes in <sup>137</sup>Cs profiles along a transact at each of the three zones could be interpreted as changes due to redistribution or accretion in exactly the same way that McHenry and Bubenzer (1982) interpreted changes in field distribution of <sup>137</sup>Cs.

Perhaps the best example of the potential of the <sup>137</sup>Cs technique for this type of application is the Lake Michigan work of Plato and co-workers (Plato and Goldman, 1972; Plato, 1974; Plato and Jacobson, 1976). The ability to interpret the structured concentration of <sup>137</sup>Cs in sediment is a very powerful aid in studying environmental influences.

#### Practicalities of the 137Cs method

There are few details in the literature of the methods and rationale used by individual workers to obtain their samples. In taking a core sample there are three main difficulties:

- (i) to avoid disturbing the very soft sediments in the upper 20 cm or so of the sedimentary sequence, yet be able to cope with a varying degree of compaction of the sediments with increasing depth (age). (The upper 20 cm region would include much of the <sup>137</sup>Cs data of interest).;
- (ii) to overcome suction effects during removal of the core tube from the sediment bed, or from the outer casing of the coring device; and
- (iii) to avoid compression or other disturbances of the core during penetration of the core tube into the sediment bed or extrusion of the core section, which will give, rise to serious errors in an accurate determination of the sediment-water interface or the true length of the core.

A Mackereth corer (Mackereth, 1958; Mackereth, 1969) or an adaptation of its design features, would provide a good working answer to these problems. However, it requires careful handling, since its fast return to the surface can be hazardous. The core tubes range up to 1 m in length, and generally have a diameter of 5 cm. The restricted diameter means that numerous cores have to be taken at each site to accumulate sufficient material for gamma spectrometry. Ritchie and McHenry (1978) collected eight cores per site and composited them by 10 cm increments. Where it is available, clear plastic rubbing, such as polycarbonate, is to be preferred as the integrity of the core can be appraised at the surface and the coring repeated if necessary. Cores should be kept at or near in situ temperature to prevent expansion due to gas formation. Some organisations have available a cooled room for this purpose. Strong and Cordes (1976) have described a cloth sleeve filled with dry ice as a means of freezing cores. They reported that stratigraphic disturbance due to ice crystal

formation did not appear to be a problem. If this or a similar cooling procedure is unavailable in the field, the cores have to be sectioned as soon as possible. An alternative to hydraulic extrusion is to section the core tube carefully along the midline to enable one half to be removed entirely. Obviously a well consolidated sediment is necessary for this technique.

#### Gamma spectrometry

The analysis of <sup>137</sup>Cs by gamma spectrometry, using Ge(Li) or HyperPure Ge detectors, is relatively simple. The <sup>137</sup>Cs peak has an energy of 662 keV and the only interference is from a peak at 666 keV due to <sup>214</sup>Bi. This interference can be corrected by measuring the adjacent 609.3 keV peak, which is also due to <sup>214</sup>Bi, and applying a proportional correction to the sum of the 602 and 606 peaks (McCallan et al., 1980). The net peak area is proportional to the concentration of <sup>137</sup>Cs. The US National Bureau of Standards, Research Material b, homogeneous river sediment for radioactivity measurements, is available as a primary standard. When multi-element analysis is required, <sup>137</sup>Cs, Th, U and K standards can be prepared, using trisodium phosphate (12 H<sub>2</sub>O) as the matrix (McHenry et al., 1973).

#### COMPUTATION OF LAKE LIFE

The computation of lake life provides an idea about the time period after which the lake will not be useful for water related activities. This may happen in two ways, i.e., when the water of the lake is polluted upto an extent that it can not be used for all practical purposes or the capacity of the lake is reduced due to sedimentation upto an extent that it is not possible to meet out the water demand. This exercise should be done periodically to understand the lake environment and to plan the suitable measures timely for lake restoration, if the reduction of lake capacity is found at a higher rates or higher order of lake eutrophication is observed.

The lake sounding data of different time periods are used to determine the volume and area of the lake. The average volume of the lake is divided by the average area to get the mean depth of lake. The weighted average rate of sedimentation is determined using the different rates of sedimentation obtained in different parts of a lake. The expected useful life of the lake is determined by dividing mean depth of the lake by weighted average rate of sedimentation. However, the validity of predicted lake life depends on the accuracy of mean depth of the lake. Although, the effect of compression of sediments by its weight is automatically included partly while estimating the rate of sedimentation, but the estimated useful life of a lake may be more accurate if the effect of compression is considered fully.

Both <sup>210</sup>Pb and <sup>137</sup>Cs geochronological dating are powerful techniques for determining recent sediment accumulation rates. Therefore, the dating lake life can be determined using the sediment accumulation rates estimated using radiometric techniques.

Ritchie and McHenry (1985), while comparing <sup>137</sup>Cs dating method with bottom contour method for measuring rates of sediment accumulation, has recommended the <sup>137</sup>Cs method for a quick and accurate estimates of sediment accumulation since 1954. Ritchie and McHenry, further pointed out that it is crucial to know the control points for the survey lines for the success of the bottom contour method. However, the following points should be given due care while determining life of a lake.

- a) For the estimation of lake life, the sediment accumulation rates should be known accurately.
- b) Sedimentation rates obtained through lake sounding data collected without mechanical or electronic positioning systems may often be misleading. Care should also be taken to use the same sounding weight or rod every time, to minimize the errors.
- c) Sedimentation rates determined through radiometric dating techniques provide precise data and it is also cost effective, as it needs one time sampling only.

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