The Influence of Radon Diffusion on the ^{210}Pb Distribution in Sediments

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A mathematical model is presented which describes the distribution of radon 222 in sediments having a constant or variable depth distribution of radium 226. The model is extended to the distribution of lead 210, taking into account the mobility of radon (the precursor of ^{210}Pb) within the sediment column. The ^{210}Pb model is compared, at constant radium activity, with the conventional approach which disregards the radon diffusion when estimating sedimentation rates by the ^{210}Pb method. The ratio between apparent and real sedimentation rate, s'/s , expressed as a function of three dimensionless parameters, demonstrates the importance of the radon diffusion effect. This effect is particularly important for sediments with small initial excess ^{210}Pb activity, small sedimentation rate, large radon diffusivity, or a combination of these factors. Applied to Lake Geneva, the sedimentation is estimated to be larger by 30–50% than the original value by Krishnaswami et al. (1971). In sediments which are mixed at the surface (physical mixing or bioturbation), the ^{210}Pb activity in the mixed layer is diminished compared to that in the settling sediment material (Robbins et al., 1977), and radon diffusion makes the activity difference even larger, especially for low initial excess ^{210}Pb activity, small sedimentation rate, and large mixing intensity. This result may be of importance for the balance of ^{210}Pb in an aquatic system if the calculations are based on activities measured in the sediment.

INTRODUCTION

Since Krishnaswami et al. [1971] first used ^{210}Pb for dating recent lacustrine sediments, many contributions have appeared in the field of ^{210}Pb geochronology. A few examples follow: recent sedimentation rates were measured in Lakes Mendota, Trout, Tahoe, and Titicaca [Koide et al., 1973]; in Lake Washington [Schell, 1974]; in Lake Michigan [Robbins and Edgington, 1975]; in Lake Superior [Bruland et al., 1975]; in Greifensee (Switzerland) [Gäggeler et al., 1976]; in Lake Huron [Robbins et al., 1977]; in Lake Ontario [Farmer, 1978]; in Lake Erie [Nriagu et al., 1979]; in several lakes in Finland [Appleby et al., 1979]; and in Saguenay Fjord (Quebec) [Smith and Walton, 1980]. A review of lacustrine studies with ^{210}Pb is given in the work by Krishnaswami and Lal [1978].

Robbins [1978] has treated in detail the principles underlying the ^{210}Pb method and reviewed its applications. He considered models for which (1) both the sedimentation rate and the flux of ^{210}Pb are constant, (2) only the flux of ^{210}Pb is constant, and (3) the mixing of surface sediments caused by bottom currents and/or the activity of benthic organisms is taken into account.

A common feature of all the models is the assumption that the ^{226}Ra content of the sediment is in secular equilibrium with ^{210}Pb and represents the 'supported ^{210}Pb ' activity. This activity is to be subtracted from the analytically measured ^{210}Pb , to obtain the 'unsupported' or 'excess ^{210}Pb .' The exponential decay with depth of the 'unsupported ^{210}Pb ' is then used to derive the sedimentation rate. The ^{226}Ra content thus serves to distinguish between ^{210}Pb atoms which, otherwise, seem indistinguishable. However, an attempt to differentiate the ^{210}Pb atoms by selectively leaching the unsupported ^{210}Pb (at pH 5–6 with 2% disodium ethylene-diamine-tetraacetate (EDTA)) has been made by Krishnaswami et al. [1971]. By this method, 20%–40% of the supported ^{210}Pb seemed to be carried

away. Evidently, it is still not known whether the supported ^{210}Pb atoms differ in their chemical reactivity to an extent that selective separation from the unsupported ^{210}Pb atoms could be feasible.

Lead 210 levels measured in the uppermost sediment layers of many lakes are about 10–20 times larger than the ^{226}Ra content of these sediments; in these cases, more than 90% of the ^{210}Pb atoms are unsupported and the exact differentiation between supported and unsupported ^{210}Pb becomes of minor importance. However, in cores in which the measured ^{210}Pb activities are relatively low and only 2–3 times larger than their ^{226}Ra content, the assumption that the ^{226}Ra – ^{210}Pb atoms are at secular equilibrium should be carefully examined.

Several problems should be considered; the first lies in the essence of the ^{210}Pb method. Some of the ^{222}Rn , which is produced by decay of ^{226}Ra within soil particles, is permanently emanating from land surfaces (the oceanic flux of Rn is only about 1% of the continental flux) causing a depletion of radon and in turn a depletion of supported ^{210}Pb in these upper soil layers. The emanating portion of radon varies from 0.1 to 0.7 depending upon soil type (see references cited by Moore and Poet [1976]). Clearly in upper soil particles, the ^{226}Ra activity is not at radioactive equilibrium with ^{210}Pb . On the other hand, the uppermost layers of soil are also loaded with unsupported ^{210}Pb , since the radon emanated from the continents decays in the atmosphere and yields a flux of unsupported ^{210}Pb back to the earth's surface. Then, freshly sedimented detrital particles, enriched in ^{210}Pb , may be eroded from watersheds of lakes and transported to bottom deposits.

The second aspect which should be considered is the emanation of radon from bottom sediments: excess radon has been measured close to the floor of the oceans and of lakes. Profiles of excess radon have been used as a tracer for the vertical near-bottom eddy diffusion in the ocean [Broecker et al., 1967, 1968; Chung and Craig, 1972] and for the two-dimensional, vertical and horizontal, eddy diffusion in the hypolimnion of lakes [Imboden and Emerson, 1978]. In most cases,

particularly in deep lakes and oceans, the radon which diffuses from bottom sediments does not reach the atmosphere, but decays within the water body, producing a continuous flux of unsupported ^{210}Pb back to the sediment-water interface. Indeed, *Key et al.* [1979] have found a reasonable balance between integrated radon deficit in marine sediments and radon surplus in the overlying water. Their model predicts a radon leakage from deep-sea sediments of 30–40%.

On the one hand, radon emanation implies a deficiency of supported ^{210}Pb in the sediments, and on the other hand, radon decay in the water column produces a flux of unsupported ^{210}Pb to the sediment interface. If radon loss by outflow and decay within the lake is disregarded, the total deficiency of supported ^{210}Pb in the sediment column is balanced by the ^{210}Pb flux produced from excess radon decay in the bottom waters. This dislocation of supported ^{210}Pb thus becomes a source of unsupported ^{210}Pb at the sediment-water interface. It is possible that the 30%–40% of supported ^{210}Pb , found to be leachable with EDTA by *Krishnaswami et al.* [1971], represents an approximate measure of the dislocated ^{210}Pb .

A third problem in ^{210}Pb dating derives from the variable distribution of ^{226}Ra in the sediments. Where this occurs, the profiles of in situ produced (or supported) ^{210}Pb will not coincide exactly with those of ^{226}Ra , because of radon diffusion. This happens at all depths, not just in the zone where radon is lost from the sediments.

In this article, we present a model which describes the influence of radon mobility on the ^{210}Pb profile in sediments. Simple dimensionless relations are derived and discussed. These equations are used to identify those cases where ^{210}Pb dislocation is important for the determination of sedimentation rates. The model is further extended to assess the combined effect of sediment perturbation and radon diffusion; this analysis may be of relevance in order to balance the sources of ^{210}Pb flux to the sediment. Application of the model to Lake Geneva sediments is briefly discussed. A detailed treatment of the ^{226}Ra - ^{210}Pb balance and its application to Lake Kinneret will be presented elsewhere (*M. Stiller and D. M. Imboden*, manuscript in preparation, 1981).

RADON 222 IN SEDIMENTS

The sediment is treated as a one-dimensional column with time-independent porosity ϕ (no compaction). The depth coordinate z is zero at the sediment-water interface and positive downward into the sediment. The derivative $(\partial/\partial t)_z$ refers to constant z , where $(\partial/\partial t)_T$ means temporal change within a fixed layer described by its time of deposition T (see *Imboden* [1975] for further details). The variables are defined in the notation list. Note that all activity units are per unit bulk volume; this definition simplifies those equations which contain the activities of both dissolved and solid species.

The total radon 222 activity in sediments is given by the sum of two terms:

$$N = N_w + N_s \quad (1)$$

Radon in pore water (N_w) originates from the fraction f of radium 226 nuclei located close enough to the sediment particle surface that the decay product (radon 222) escapes into the pore water [*Imboden and Emerson*, 1976; *Key et al.*, 1979]. Radon contained in the solid phase (N_s) is given by

$$N_s = A(1 - f) \quad (2)$$

where A is the total radium 226 activity in the sediment.

N_w is determined by the diffusion/advection equation

$$\left(\frac{\partial N_w}{\partial t}\right)_z = \frac{\partial}{\partial z} \left(D \frac{\partial N_w}{\partial z} \right) - (s + V) \frac{\partial N_w}{\partial z} - \lambda_N N_w + \lambda_N f A \quad (3a)$$

where s is the sediment accumulation rate and V the pore water advection velocity originating from a possible water flux between the lake and an underlying aquifer.

Since the radon distribution is dominated by its rapid decay, quasi steady state can be assumed. The term describing sediment accumulation, $s(\partial N_w/\partial z)$, is very small compared to the diffusion and radioactive decay terms. In the layer where radon diffusion is important, the diffusion coefficient D is assumed to be constant, although its value may be larger than molecular diffusion [*Martens et al.*, 1980]. Thus, (3a) reduces to

$$0 = D \frac{\partial^2 N_w}{\partial z^2} - V \frac{\partial N_w}{\partial z} - \lambda_N (N_w - fA) \quad (3b)$$

First, (3b) is solved for constant radium activity A :

$$N_w(z) = N_w^0 e^{-\alpha z} + fA(1 - e^{-\alpha z})$$

$$\alpha = \frac{1}{2D} [-V + (V^2 + 4\lambda_N D)^{1/2}] \quad (4)$$

where α^{-1} is a measure of the scale length for radon loss from the sediment. To estimate its magnitude, we chose $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ which is the free solution diffusivity at 5°C [*Broecker and Peng*, 1974]. The influence of the sediment structure [*Manheim*, 1970] is ignored for simplicity and because porosity is usually very high near the sediment surface. With $V = 0$, we get $\alpha = (\lambda_N/D)^{1/2} = 0.51 \text{ cm}^{-1}$ or $\alpha^{-1} = 1.96 \text{ cm}$. In fact, α is independent of V as long as $|V| \ll (4\lambda_N D)^{1/2} \sim 260 \text{ cm yr}^{-1}$. Under the influence of a large seepage of water into the sediment ($V > 0$), however, the radon-depleted sediment layer (α^{-1}) may become larger.

N_w^0 , the radon activity at the sediment-water interface, expresses a dynamic equilibrium between radon flux in the pore water (D) and turbulent transport in the lake (D_E). The relative size of the two terms on the right-hand side of (4), i.e., the ratio (N_w^0/fA) , depends on $(D/D_E)^{1/2}$. Since D_E is usually much larger than diffusion in the sediments, the term containing N_w^0 can be disregarded. Indeed, in marine and freshwater systems, measured radon boundary values at the sediment surface are usually much smaller than the equilibrium activities in the sediment pore water.

With $N_w^0 \sim 0$ the total radon activity in the sediment (equations (2) and (4)) becomes

$$N(z) = A(1 - f e^{-\alpha z}) \quad (5)$$

The corresponding radon loss from the sediment per unit time and sediment surface is

$$F_N = \left| D \frac{\partial N}{\partial z} \right|_{z=0} = D \alpha f A \quad (6a)$$

and for $V = 0$ [*Broecker*, 1965; *Imboden and Emerson*, 1976],

$$F_N = (\lambda_N D)^{1/2} f A \quad (6b)$$

Note that because of the particular units of A , these equations do not contain porosity ϕ . The ^{210}Pb generated by the decay of escaped radon partially returns to the sediment following

^{222}Rn decay and contributes to the excess ^{210}Pb activity of the fresh sediment material.

Second, for variable radium 226 activity, the steady state solution of (3b) is

$$N_w(z) = N_w^0 e^{-\alpha z} + \frac{\alpha}{2} f [b_1(z) + b_2(z)]$$

$$b_1 = \int_z^\infty A(\xi) e^{-\alpha(\xi-z)} d\xi$$

$$b_2 = \int_0^z A(\xi) e^{\alpha(\xi-z)} d\xi - \int_0^\infty A(\xi) e^{-\alpha(\xi+z)} d\xi \quad (7)$$

The upper boundary of the integrals, $\xi = \infty$, can be approximated by a finite value z' as long as $(z' - z) \gg \alpha^{-1}$. For constant A , (7) is identical with (4).

The influence of interstitial transport on the shape of the radon profile is exemplified by the application of (2) and (7) to a core from Lake Kinneret, Israel (Figure 1). In spite of the inhomogeneous radon source (radium 226), radon diffusion leads to a relatively smooth radon profile. In addition, at the surface the total radon activity drops to 40% of its equilibrium level with radium; this is the result of dissolved radon diffusion across the sediment-water interface into the lake.

LEAD 210 IN SEDIMENTS

The lead 210 distribution in the sediment is controlled by two processes: (1) the production of ^{210}Pb from ^{226}Ra via ^{222}Rn and (2) radioactive decay.

The ^{210}Pb activity (P) in a fixed sediment layer is given by

$$\left(\frac{\partial P}{\partial t} \right)_T = \lambda_P (N - P) \quad (8)$$

In the classic approach first introduced by Krishnaswami *et al.*

[1971], the radon activity N is assumed to be equal to the radium activity A . The latter is constant within a fixed layer provided that mobility and decay of radium can be neglected.

In the ^{210}Pb literature, the term 'excess ^{210}Pb ' is used in two different ways: (1) as the difference between the ^{210}Pb and ^{226}Ra activities, $Y = P - A$, at a particular depth and (2) as the difference between the ^{210}Pb activity at depth z and at that depth where no further change with z is observed, $X = P - P_\infty$. For reasons explained later, we use the first definition, Y , as excess ^{210}Pb . For the common approximation ($N = A = \text{const}$), X and Y are identical, and (8) leads to the familiar solution:

$$Y_T(t) = Y_T(0) e^{-\lambda_P(t-T)} \quad t \geq T \quad N \text{ approximated by } A \quad (9)$$

where T is time of deposition and $Y_T(0)$ the initial excess ^{210}Pb content for $t = T$. Any further transformation implies further assumptions on the sedimentation rate and the initial activity of ^{210}Pb , $Y_T(0)$. The reader is referred to Robbins [1978] for more details. For simplicity, we adopt the simplest case, the constant flux-constant sedimentation rate model. With $t - T = z/s$ and $Y_T(0) = Y_0 = \text{const}$, (9) can be transformed into a function of depth:

$$Y(z) = Y_0 e^{-\beta z} \quad \beta = \lambda_P / s \quad N \text{ approximated by } A \quad (10)$$

According to the applied definition of excess ^{210}Pb , this equation is also valid if the radium activity varies with depth, provided that A is actually measured and not just estimated from the ^{210}Pb activity at lower depths. So much for the classical approach.

Next, we discuss the question of how radon diffusion affects the ^{210}Pb profiles and the calculation of sedimentation rate from ^{210}Pb profiles. To give an answer, (8) is integrated for a time-variable radon source function $N_T(\tau)$ where $\tau = t - T$ is the age of the sediment layer T :

$$P(\tau) = P_0 e^{-\lambda_P \tau} + \lambda_P \int_0^\tau e^{-\lambda_P(\tau-\tau')} N(\tau') d\tau' \quad (11)$$

For constant s , the integration over layer age τ can be replaced by integration over depth z (cf. equation (10)). For constant radium distribution and $N_w^0 \sim 0$, $N(z)$ is given by (5), which inserted into (11) leads to the following expression:

$$[P(z) - A] = Y(z) = Y_0 e^{-\beta z} - \epsilon(z) \quad (12)$$

where $P(z)$ is the depth distribution of total ^{210}Pb for which the 'history of radon loss' of each sediment layer has been taken into account, and $\epsilon(z)$ represents the 'radon loss correction function.'

$$\epsilon(z) = \frac{Af}{1 - (\alpha/\beta)} (e^{-\alpha z} - e^{-\beta z}) \quad \text{for } \alpha \neq \beta$$

$$\epsilon(z) = Af\beta z e^{-\beta z} \quad \text{for } \alpha = \beta \quad (13)$$

The term $\epsilon(z)$ has been introduced because the intention of this article is to discuss the influence of radon mobility on the relative shape of the excess ^{210}Pb distribution in sediments, especially on the slope of $Y(z)$ from which the sediment accumulation rate is determined. Equation (12) resembles the classical solution (equation (10)), and the effect of radon diffusion appears only in the correction function $\epsilon(z)$. Note that $\epsilon(z)$ is proportional to the escapable radon activity Af so that for $f = 0$, (12) reduces to (10).

Fig. 1. Profiles of ^{226}Ra and ^{210}Pb measured in a sediment core from Lake Kinneret taken in December 1971. The total ^{222}Rn activity was calculated from (7) using the radon escape probability $f = 0.6$.

Since $\epsilon(z)$ describes the deviation of $Y(z)$ from an exponential curve (equation (10)) with identical initial value Y_0 , the boundary value $\epsilon(0)$ must be zero. The $\epsilon(z)$ is always positive for $z > 0$, tends to zero for $z \rightarrow \infty$, and reaches one maximum for positive z . Thus, radon diffusion seems to diminish the total standing crop of excess ^{210}Pb per unit sediment surface (compare the integrals over z of (10) and (12), respectively). However, this is a fallacy, since we have arbitrarily assumed identical initial values Y_0 in both equations. As previously discussed, radon diffusion and subsequent decay of escaped ^{222}Rn in the water column may, in fact, add to the initial ^{210}Pb activity in the fresh sediment material. The nature of this 'external' or 'recycled' ^{210}Pb source is usually not considered in defining the initial excess ^{210}Pb , Y_0 —and it would not be easy to do so, since horizontal advection in the bottom water may invalidate the local balance between radon flux from the sediment and in situ production of ^{210}Pb by radon decay. Therefore, we do not further discuss the aspect of the absolute size of Y_0 .

In Figure 2, the ratio Y/Y_0 computed from (12) is compared (for different s and f) to this same ratio computed from the classical model (equation (10)) (where $f = 0$). The initial ratio A/Y_0 was assumed to be 1. Besides the linear dependence of the correction term $\epsilon(z)$ on Af , the effect of radon diffusion is determined by the ratio of the two scale lengths α^{-1} and β^{-1} :

$$R = \alpha/\beta = \frac{s}{2D\lambda_P} \{-V + (V^2 + 4\lambda_N D)^{1/2}\} \quad (14a)$$

For $V = 0$,

$$R = \left(\frac{\lambda_N}{D}\right)^{1/2} \frac{s}{\lambda_P} \quad (14b)$$

As R decreases below 1, the effect of radon diffusion on the ^{210}Pb profile becomes increasingly important; or in terms of the sedimentation rate:

$$R < 1 \leftrightarrow s < \lambda_P \left(\frac{D}{\lambda_N}\right)^{1/2} = s_{\text{crit}} \quad (15)$$

In Figure 2, we have chosen $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$. However, there is evidence that the migration of radon near the sediment surface may be faster than molecular diffusion [Martens *et al.*, 1980]. As pointed out earlier, seepage of lake water through the sediments into an aquifer or groundwater current ($V > 0$) may significantly increase α^{-1} and thus shift the 'critical sedimentation' s_{crit} to much higher values than shown in Figure 2. In Figure 3, the relation between s and R is plotted for different assumptions.

The interpretation of measured ^{210}Pb data from sediment cores in terms of the classical model (equation (10)) underestimates the sedimentation rate. In fact, for small R (i.e., for small s), the excess ^{210}Pb may become negative; this means that the ^{226}Ra activity is larger than the ^{210}Pb activity. One extreme example is given in Figure 2 ($s = 0.01 \text{ cm yr}^{-1}$). In this situation, it is important to note that the determination of radium activity by measuring the ^{210}Pb activity at a near-constant level can produce serious errors.

The case of very low sedimentation rate resembles the case in soil which can be interpreted as a sediment with $s = 0$ receiving excess ^{210}Pb from the atmosphere. In fact, Lewis [1977] and Moore and Poet [1976] have measured ^{226}Ra and ^{210}Pb profiles in soils which are similar to the case $s = 0.01 \text{ cm yr}^{-1}$ (Figure 2). As a result of radon diffusion to the atmosphere, ^{210}Pb in soil drops below ^{226}Ra at a depth of about 15 cm.

Since radon diffusion changes the exponential character of the ideal excess ^{210}Pb profile, the apparent sedimentation rate s' is no longer constant with depth. In order to analyze the relation between the true and the in situ apparent sedimentation rate, s and $s'(z)$, we adopt the approach of Robbins [1978] and write

$$s'(z) = -\lambda_P \left(Y \frac{dY}{dz} \right) \quad (16)$$

We use (12) and (13) and introduce a dimensionless depth coordinate $\zeta = \beta z$ in order to derive an expression which only depends on ζ , $R = \alpha/\beta$, and $Q = Af/Y_0$ and relates the true and apparent sedimentation rates, s and s' :

Fig. 2. Ratio of 'excess ^{210}Pb ' activities at depth z and $z = 0$, Y/Y_0 , calculated from (12) and (13) for different sedimentation rates s and radon escape probabilities f . The ratio of ^{226}Ra to excess ^{210}Pb activities in the fresh sediments, A/Y_0 , is 1, the radon diffusivity $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$. Note the changing scale for the depth coordinate, especially the nonlogarithmic scale for $s = 0.01 \text{ cm yr}^{-1}$ where Y reaches negative values, i.e., the ^{210}Pb activity drops below the ^{226}Ra activity.

$$(s'/s) = \frac{(1 - R + Q) - Qe^{(1-R)\xi}}{(1 - R + Q) - RQe^{(1-R)\xi}} \quad R \neq 1 \quad (17a)$$

$$(s'/s) = \frac{1 - Q\xi}{1 - Q(\xi - 1)} \quad R = 1 \quad (17b)$$

In Figure 4, the ratio (s'/s) is plotted as a function of ξ for different parameter sets R and Q . The graphical representation of (17) illustrates the following properties of the above relation:

1. The ratio (s'/s) is always smaller than 1; thus the apparent sedimentation rate s' tends to underestimate the real rate s .
2. At the sediment surface ($\xi = 0$), (s'/s) is independent of R and equals $(1 + Q)^{-1}$.
3. For every Q there exists one value $R^* = Q + 1$, which makes (s'/s) independent of ξ : $(s'/s) = R^{-1} = (Q + 1)^{-1}$. Then, for all $R > R^*$, (s'/s) increases with depth and eventually reaches its limit $(s'/s) = 1$. In this case, the slope of the excess ^{210}Pb profile approaches the constant value of the classic theory (equation (10)).

4. If $R < R^*$, (s'/s) decreases with depth, the ^{210}Pb profile becomes less steep with increasing depth, and the discrepancy between real and effective sedimentation rate becomes larger.

Since in most sediments the escapable radon activity (A_f) is much smaller than the initial excess ^{210}Pb activity (Y_0), Q is usually not larger than 0.1. In these cases, the influence of radon diffusion on the slope of the ^{210}Pb profile is only significant if $R < 1$. Such values for R occur for a very small sedimentation rate, a large radon diffusion coefficient (D), a large pore water infiltration flux ($V > 0$), or a combination of these factors (Figure 3). However, large deviations of (s'/s) from unity at greater depths (say $\xi > 1.5$), as they occur for $Q =$

Fig. 3. Dimensionless parameters R as a function of sedimentation rates for various conditions. R represents the ratio of ^{210}Pb decay scale length to radon diffusion scale length. $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ and $V = 0$ (curve A); $D = 2.4 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ and $V = 0$ (curve B); $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ and $V = +250 \text{ cm yr}^{-1}$ (advection into sediments) (curve C); and $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ and $V = -250 \text{ cm yr}^{-1}$ (advection from sediments) (curve D). Influence of radon migration on ^{210}Pb increases for decreasing R values.

Fig. 4. Ratio between apparent and real sedimentation rate, s'/s , as a function of normalized depth $\xi = \beta z$ for different parameters $R = \alpha/\beta$ and $Q = A_f/Y_0$ (see (17)).

0.05 and $R < 0.5$, for instance, 'distort' the excess ^{210}Pb profile in a portion of the sediment column where the initial excess activity has already dropped to a small fraction of its original value and where other factors (counting error, computation of Y as a difference of two activities) may dominate the uncertainties in the slope of $Y(z)$.

So far, we have only discussed the case of constant radium activity A . For variable A , the radon distribution in the sediment (equation (7) for N_w and (2) for N_s) can serve as a base for the integration of (11). In fact, $N_T(\tau')$ had to be known for $0 \leq \tau' \leq (t - T)$, i.e., from age 0 to the present age of the layer, $t - T$. One way to calculate $N_T(\tau')$ would consist in evaluating (7) for earlier times, that is, by moving the sediment-water interface downward until it finally reaches the layer under consideration, T . However, in the appendix, it is shown how this rather complicated procedure can be simplified.

Similar to the surface sediment layers, zones of negative excess ^{210}Pb can also occur in the deeper part of the sediment if the radium distribution shows strong vertical gradients which, for ^{210}Pb , have been partially smoothed by radon diffusion.

Among the larger lakes, Lake Geneva (also known as Lac Léman) and Lake Kinneret have the largest initial ratio of ^{226}Ra to excess ^{210}Pb activity. A detailed analysis of the complete ^{226}Ra - ^{210}Pb system in Lake Kinneret demonstrates that a relatively small atmospheric input is responsible for the unique conditions (see Figure 1). However, the sediments are

also influenced by other factors. A discussion of these data lies beyond the framework of this article and will be presented elsewhere (M. Stiller and D. M. Imboden, manuscript in preparation, 1981).

Lake Geneva belongs to the first series of lakes to which the ^{210}Pb dating method has been applied [Krishnaswami *et al.*, 1971]. At this time, the ^{210}Pb measurements were still suffering from relatively large errors, and the lateral spacing of the data points was sparse due to the large quantities of sediment material needed for the analysis. Following the analysis by Krishnaswami *et al.*, the activities of initial excess ^{210}Pb (Y_0) and ^{226}Ra (A) are 5.8 and 3.8 dpm/g, respectively, and the apparent (uncorrected) sedimentation rate s' is 0.12 cm yr $^{-1}$. There are no measurements of ^{222}Rn for Lake Geneva sediments, so we guess f to be about 0.5, a slightly higher value than the mean for deep-sea sediments found by Key *et al.* [1979] but still within the range of 0.3 to 0.7 calculated by these authors. With $D = 8 \times 10^{-6}$ cm s $^{-1}$ we get $Q = 0.33$ and $R = 2.0$ (using s' as a first approximation for the calculation of R (see Figure 3)). Equation (17) yields $s = 0.16$ cm yr $^{-1}$ for the sediment surface, $s = 0.14$ cm yr $^{-1}$ at 5 cm depth, and $s = 0.12$ cm yr $^{-1}$ below 10 cm.

Now, we can recalculate the profile starting with a constant real sedimentation rate $s = 0.15$ cm yr $^{-1}$ (thus producing an excess ^{210}Pb profile with a depth-dependent apparent sedimentation rate s' which is still in accordance with the measured data points). If a larger radon diffusion coefficient were adopted (a possibility which at least cannot simply be excluded since the hypolimnetic layers of Lake Geneva are low in oxygen and bubbling of methane could occur [Martens *et al.*, 1980]), sedimentation rates of 0.15–0.20 cm yr $^{-1}$ would still be compatible with the data.

From this short analysis we can conclude that due to the effect of radon diffusion, the sedimentation rate is probably about 30% larger than the values calculated by Krishnaswami *et al.* [1971], and that values up to 0.20 cm yr $^{-1}$ are possible. Any further conclusion would certainly overstate the available data, especially since the influence of sediment compaction, i.e., the transformation to a normalized depth scale, would have also to be taken into account. New measurements in Lake Geneva including radon measurements to determine the escape rate f could lead to additional insight into this problem.

PERTURBATION OF THE SEDIMENT COLUMN

Robbins and Edgington [1975] and Robbins *et al.* [1977] have discussed the influence of sediment mixing at the surface on the vertical distribution of ^{210}Pb and ^{137}Cs . In their model [Robbins *et al.*, 1977], mixing is described as a quasi-diffusional process [Guinasso and Schink, 1975] using a constant diffusion coefficient D_B within a mixed layer of thickness z_m and $D_B = 0$ below the mixed layer. In this section, the effect of radon diffusion on the bioturbated model is examined (Figure 5).

In order to simplify the discussion, radium is assumed to have constant activity with depth. Regarding the mixing effect, this is not an unpalatable assumption. Then, radon in the solid phase, N_s , is not affected by sediment mixing (see (2)). However, the diffusion equation for dissolved radon (equation (3b)) has to be replaced by two expressions, one for each zone, which differ from each other only by their diffusion coefficients, $(D + D_B)$ for the mixing zone ($0 < z < z_m$) and D for $z > z_m$. Using appropriate continuity conditions at the bound-

ary $z = z_m$ for the simpler case $V = 0$ and $N_w^0 = 0$, the total radon activity is given by

$$\begin{aligned} N(z) &= A(1 - n_1 e^{-\alpha_m z} - n_2 e^{\alpha_m z}) & 0 \leq z \leq z_m \\ N(z) &= A(1 - n_3 e^{-\alpha z}) & z > z_m \end{aligned} \quad (18)$$

where

$$\begin{aligned} \alpha &= \left(\frac{\lambda_N}{D} \right)^{1/2} & \alpha_m &= \left(\frac{\lambda_N}{D + D_B} \right)^{1/2} \\ n_1 &= (f/\psi) e^{\alpha_m z_m} (1 + \gamma) \\ n_2 &= (f/\psi) e^{-\alpha_m z_m} (1 - \gamma) \\ n_3 &= (2f/\psi) e^{\alpha z_m} \\ \psi &= e^{\alpha_m z_m} (1 + \gamma) + e^{-\alpha_m z_m} (1 - \gamma) \\ \gamma &= \left(\frac{D}{D + D_B} \right)^{1/2} = \frac{\alpha_m}{\alpha} \end{aligned}$$

This result could be used to integrate the equation for ^{210}Pb (equation (11)). However, the effect of mixing can be significantly simplified by the assumption that within the mixed layer, ^{210}Pb should be homogeneously distributed. This approximation is reasonable as long as D_B is at least a few times larger than $z_m^2 \lambda_p$, for instance, $D_B \geq 5 z_m^2 \lambda_p$. For $z_m = 5$ cm, mixing intensities of 10^{-7} cm 2 s $^{-1}$ or larger would be in accordance with this limit. Krishnaswami *et al.* [1980], using ^7Be measurements, have estimated D_B to be in the range of 10^{-7} cm 2 s $^{-1}$ for Long Island Sound sediments. In Lake Whitney (Connecticut) this is an upper limit for D_B , and the ^{210}Pb activity is not constant near the sediment surface, though changing slopes in the profile could (among other possibilities) originate from particle mixing. Lead 210 has not been measured in the cores from the sound.

Goldberg and Koide [1962] and later Berger and Heath [1968] have formulated the homogeneous mixing model, and Robbins *et al.* [1977] applied it to excess ^{210}Pb in sediments. Taking into account the influence of radon diffusion, we re-

Fig. 5. Model for sediment mixing (bioturbation): The activity of ^{210}Pb in the settling material (P_0) is homogeneously mixed to the depth z_m leading to the activity P_m ($< P_0$) in the mixed layer. Due to its much larger decay constant, ^{222}Rn is not necessarily constant within the mixing layer.

write the model for the total activity of ^{210}Pb . In the mixing zone the balance for ^{210}Pb is given by

$$z_m \frac{dP_m}{dt} = s(P_0 - P_m) + \lambda_P \int_0^{z_m} N(z) dz - \lambda_P z_m P_m \quad (19)$$

where P_m is the ^{210}Pb activity in the mixed layer and P_0 the ^{210}Pb activity in the settling material (Figure 5). Since ^{222}Rn has a much larger decay rate than ^{210}Pb , it could hardly ever become homogeneously mixed, so that (18) is used to evaluate (19). The steady state solution is

$$(P_m - A) = \frac{s}{s + \lambda_P z_m} \left\{ (P_0 - A) - A f \frac{\lambda_P}{s \alpha_m} \cdot \frac{\sinh(\alpha_m z_m) + \gamma [\cosh(\alpha_m z_m) - 1]}{\cosh(\alpha_m z_m) + \gamma \sinh(\alpha_m z_m)} \right\} \quad (20)$$

which for $f = 0$ (no radon escape from sediment) is identical to equation (5) of Robbins *et al.* [1977].

Equation (20) can be transformed into dimensionless form which relates the ratio of excess ^{210}Pb in the mixed layer and the settling material (Y_m/Y_0) to the dimensionless mixing depth $\xi_m = \beta z_m$, the modified scale length ratio $R_m = \alpha_m/\beta$, $\gamma = [D/(D + D_B)]^{1/2}$, and $Q = A f/Y_0$:

$$\frac{Y_m}{Y_0} = \frac{1}{1 + \xi_m} \left[1 - \frac{Q}{R_m} \frac{\sinh(R_m \xi_m) + \gamma [\cosh(R_m \xi_m) - 1]}{\cosh(R_m \xi_m) + \gamma \sinh(R_m \xi_m)} \right] \quad (21)$$

The second term on the right-hand side describes the influence of radon diffusion on (Y_m/Y_0); it is linearly related to Q and always positive.

Since the sedimentation rate s influences two parameters, ξ_m and R_m , we have preferred to use (20) to demonstrate the rise of the correction term as a function of s . In Figure 6, the expression by Robbins *et al.* [1977] ($f = 0$) is compared to the function which includes radon diffusion. The pore water diffusion coefficient of radon ($D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$) and $Q = 0.5$ are held constant in all the calculations; the influence of the latter results in a linear relation between Q and the deviation of the curve corrected for radon diffusion from the curve where $f = 0$. Three different mixing depths ($z_m = 5, 10, 20 \text{ cm}$) are used in combination with mixing intensities between 10^{-7} and $10^{-2} \text{ cm}^2 \text{ s}^{-1}$ (the upper limit is too high to be realistic and will only demonstrate the ultimate influence of D_B). Note that for $z_m > 5 \text{ cm}$, a mixing intensity of $10^{-7} \text{ cm}^2 \text{ s}^{-1}$ would no longer be compatible with the homogeneous ^{210}Pb mixing model.

The discussion of Figure 6 can be summarized by pointing out that the influence of radon diffusion on (Y_m/Y_0) is enhanced by a large value of Q , a small sedimentation rate s , a large mixing intensity D_m , and a small mixing depth z_m . An accurate knowledge of the relation between Y_0 and Y_m may be of importance insofar as the ^{210}Pb balance for a lake requires the knowledge of the true ^{210}Pb content of the settling sediment material. Of course, as mentioned in the previous section, most of the ^{210}Pb parent lost by radon diffusion is not really lost from the lake, but simply returns as an increase in the ^{210}Pb activity of fresh sediment. However, the ^{210}Pb originating from ^{222}Rn decay in the water overlying the sediments may be transported by horizontal advection and increase P_0 elsewhere. Although negative excess ^{210}Pb activities P_m are not likely to occur, the relation expressed in (20) and in Figure 6 is still essential to the ^{210}Pb balance as long as it depends on the

Fig. 6. Ratio between 'excess ^{210}Pb ' activity in mixed layer and in settling material, Y_m/Y_0 , as a function of sedimentation rate s for different mixing depths z_m and mixing intensities D_B . The curve by Robbins *et al.* [1977] is marked by $f = 0$ for comparison. $Q = 0.5$ and $D = 8 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ are held constant in all calculations.

measured ^{210}Pb content in the sediment column and not on that in the settling material.

Below the mixing zone, the modified ^{210}Pb profile could be calculated from (18) and (11) using P_m as boundary value for the integral which now extends only over that time period during which the layer under consideration is located below the mixing zone. Since, however, in those depths, the influence of the radon diffusion on the ^{210}Pb distribution is generally of minor importance, we do not further discuss this point.

SEDIMENT COMPACTION

Sediment compaction may affect the depth distribution of an element in two ways: (1) by the induced advective flow in the pore water and (2) by the change of the in situ distance between the annual layers.

Completely immobile components are not influenced by either effect provided that the accumulated dry weight of the sediment column is used to renormalize the depth scale [Robbins and Edgington, 1975]. This is true for radium and lead 210, which are treated here as occurring only in the solid phase. But, since part of the radon occurs in the pore water, compaction may influence the final ^{210}Pb distribution via the radon migration.

Imboden [1975] has shown that for stationary sediment accumulation the advective pore water velocity induced by compaction is smaller than the sedimentation rate s . Since in (3a) the term containing s could be neglected, the same will also be true for a term expressing the flow induced by compaction. Therefore, it seems that radon profiles in pore waters are not affected by advective flow induced by compaction.

The change of the layer distance s by compaction changes the space scale within which diffusion takes place, and since we have found diffusion to be a major mechanism determining the radon distribution in the sediment, the changing space scale could be of importance. One possible way to overcome this difficulty is by choosing a normalized depth coordinate in such a way that it corresponds to the real space scale in the uppermost zone of the sediment where radon diffusion is of importance (the penetration depth of radon deficiency is usually about 4 cm). As already shown, radon diffusion below 4 cm is of significance only for a depth-variable radium distribution. The depth scale appropriate for the surface may then be too large in the deeper layers. Perhaps this could be compensated for by using a larger diffusion coefficient. In conclusion, compaction is of no major importance for the ^{210}Pb profile. At the most, it would slightly affect the radon profile which itself is only a correction to the simplified model for excess radon (equation (10)).

APPENDIX: THE TIME-VARIABLE PROFILE OF DISSOLVED RADON ^{222}Rn IN SEDIMENTS WITH A NONCONSTANT RADIUM DISTRIBUTION

In a sediment column with nonconstant radium activity A , the ^{222}Rn and ^{210}Pb activities are in nonsteady state condition; i.e., they depend on both time of deposition T and layer age τ : $N_T(\tau)$, $P_T(\tau)$.

In fact, $N_T(\tau)$ becomes independent of τ when the layer T is far away from the sediment surface, or more precisely, if the layer depth $z_T(\tau) = s\tau \gg \alpha^{-1}$. In this case, $N_T(\tau)$ is completely determined by the local distribution of ^{226}Ra .

On the other hand, the variation of the dissolved fraction $N_{w,T}(\tau)$ near the surface in analogy to the solution for constant A (equation (4)) should approximately be proportional to $(1 - e^{-\alpha z_T(\tau)})$. In fact, if we split the depth distribution of A into its mean value within the surface layer, $\bar{A}$, at $0 < z < 2\alpha^{-1}$, and the depth-depending variance, a ,

$$A = \bar{A} + a \quad (\text{A1})$$

$N_{w,T}(\tau)$ can be written in the form (see (7) with $N_w^0 = 0$)

$$N_{w,T}(\tau) = \bar{A}f(1 - e^{-\alpha z}) + \frac{\alpha}{2}f(b_1' + b_2') \quad (\text{A2})$$

$$b_1' = \int_z^\infty a(\xi)e^{-\alpha(\xi-z)} d\xi$$

$$b_2' = \int_0^z a(\xi)e^{\alpha(\xi-z)} d\xi - \int_0^\infty a(\xi)e^{-\alpha(\xi+z)} dz \quad (\text{A3})$$

with the short notation z for $z_T(\tau)$. Due to the definition of $\bar{A}$, the variance (a) is small in the surface layer provided that the depth variation of A is not too large over vertical distances of the order of α^{-1} . For $\xi \gg 2\alpha^{-1}$, because of the exponents the integrands do not contribute very much if $0 < z < 2\alpha^{-1}$. Thus, near the surface, for $z < 2\alpha^{-1}$, $b_1' \sim b_2' \sim 0$ and

$$N_{w,T}(\tau) = N_{w,T}(z) \sim \bar{A}f(1 - e^{-\alpha z}) \quad (\text{A4})$$

as stated before. Thus, the radon activity in layer T which at present has the age τ_0 , can be calculated for earlier ages $\tau < \tau_0$ by the relation

$$N_{w,T}(\tau) = N_{w,T}(\tau_0) \frac{1 - e^{-\alpha z_T(\tau)}}{1 - e^{-\alpha z_T(\tau_0)}} \quad (\text{A5})$$

where $z_T(\tau_0)$ is the present depth of the layer T , and $z_T(\tau) = s\tau$ is its depth at age τ . The advantage of (A5) lies in the fact that the evaluation of (7) has to be carried out only once, that is, for the present situation $N_{w,T}(\tau_0)$. Of course, if the radium gradients are too large to be in accordance with the mentioned conditions, $N_T(\tau)$ can still be calculated by the integration of (7) for different sediment ages, i.e., for different positions of the sediment surface.

NOTATION LIST

The basic units are centimeters (bulk), years, and disintegrations per minute. Note that in order to facilitate comparison with other information, specific values for diffusion coefficients are given in ($\text{cm}^2 \text{s}^{-1}$) but are transformed into ($\text{cm}^2 \text{yr}^{-1}$) when used in the equations.

Isotope Activities (dpm per Bulk Sediment Volume)

- A ^{226}Ra in sediment;
- $N = N_w + N_s$ total ^{222}Rn in sediment, the sum of ^{222}Rn dissolved in pore water, N_w , and enclosed in the solid phase N_s ;
- N_w^0 dissolved ^{222}Rn at sediment-water interface;
- P, P_0, P_m total ^{210}Pb in the sediment column, in the fresh sediment, and in the mixing sediment layer, respectively;
- $Y = P - A$ excess ^{210}Pb (Y_0, Y_m as for P);
- $X = P - P_\infty$ excess ^{210}Pb as often defined when ^{226}Ra is not determined.

Properties of Sediment Column, Rate Constants, and Transport Coefficients

- D, D_E diffusion coefficient of dissolved ^{222}Rn in pore water and in the lake water (eddy diffusion), $\text{cm}^2 \text{yr}^{-1}$;
- D_B mixing intensity in surface layer of sediments expressed as quasi-diffusional process, $\text{cm}^2 \text{yr}^{-1}$;
- f probability for ^{222}Rn nucleus produced by ^{226}Ra decay to escape into the pore water (radon escape probability);
- s, s' sedimentation rate, real, and apparent in situ, respectively, cm yr^{-1} ;

- $t, T, \tau = t - T$ absolute time, time of deposition of sediment layer, and age (time since deposition) of this layer, years;
- V advection velocity in pore water ($V > 0$ for flow from lake into sediment), cm yr^{-1} ;
- z depth coordinate, positive downward and $z = 0$ at sediment-water interface, cm;
- z_m depth of mixing layer, cm;
- λ_N, λ_P decay constant for ^{222}Rn (66 year^{-1}) and for ^{210}Pb (0.031 year^{-1}), year^{-1} ;
- ϕ sediment porosity.

Characteristic Parameters for Theory of Radon Diffusion

- $\epsilon(z)$ radon loss correction function, dpm cm^{-3} ;
- Inverse 'scale lengths' (cm^{-1})
- α radon in sediments, equal to $(1/2D)[-V + (V^2 + 4\lambda_N D)^{1/2}]$;
- α_m radon in mixing layer of sediments, equal to $[\lambda_N/(D + D_B)]^{1/2}$;
- β ^{210}Pb decay in accumulating sediments, equal to λ_P/s .
- Dimensionless parameters
- R ratio of scale lengths in undisturbed sediments, equal to α/β ;
- R_m ratio of scale lengths in mixing sediments, equal to α_m/β ;
- Q initial activity ratio, equal to a/Y_0 ;
- ζ normalized depths in undisturbed sediments, equal to βz ;
- ζ_m normalized depths in mixing sediments, equal to βz_m ;
- $\gamma = [D/(D + D_B)]^{1/2}$.

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