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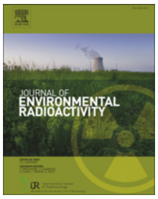

^{210}Pb as a tool for establishing sediment chronologies: examples of potentials and limitations of conventional dating models

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ABSTRACT

For aquatic sediments, the use of ^{210}Pb originating from the decay of atmospheric ^{222}Rn is a well-established methodology to estimate sediment ages and sedimentation rates. Traditionally, the measurement of ^{210}Pb in soils and sediments involved laborious and time-consuming radiochemical separation procedures. Due to the recent development of advanced planar (‘n-type’) semi-conductors with high efficiencies in the low-energy range which enable the gamma-spectrometric analysis of the 46.5 keV decay line of ^{210}Pb, sediment dating using this radionuclide has gained renewed interest.

In this contribution, potentials and limitations of the ^{210}Pb methodology and of the models used for estimating sediment ages and sedimentation rates are discussed and illustrated by examples of freshwater and marine sediments. Comparison with the use of ^{137}Cs shows that the information which may be gained by these two tracers is complementary. As a consequence, both radionuclides should be used in combination for dating of recent sediments. It is shown that for various sedimentation regimes additional information from other sources (e.g. sediment lithology) may be needed to establish a reliable chronology. A strategy for sediment dating using ^{210}Pb is recommended.

1. Introduction

Estimating the accumulation rates and the ages of marine and lacustrine sediments is of high interest. The results may give valuable information on the sediment dynamics of a watershed and of their changes induced anthropogenically, on historical trends of contaminant emissions at industrial sites, on the effects of single events, e.g. a storm surge or flood, and may assist in reconstructing local paleoclimatic conditions (e.g. Benninger and Krishnaswami, 1981; Stiller et al., 1985; French et al., 1994; Kirchner and Ehlers, 1998; Appleby, 2001; Barra et al., 2001; Oldfield et al., 2003; Lüder et al., 2006; Di Gregorio et al., 2007). Since its first applications about 40 years ago (Krishnaswamy et al., 1971; Koide et al., 1972), the use of the ^{210}Pb fraction which in sediments originates from the decay of atmospheric ^{222}Rn has become a recognized and widely used methodology to establish chronologies of recent aquatic sediments. Its potential misuse and the missing validation of ^{210}Pb derived geochronologies, however, are also a matter of concern (Smith, 2001).

Based on the experience of the author, potentials and limitations of the ^{210}Pb methodology and of the models commonly used for estimating sediment ages and sedimentation rates are discussed in the following and illustrated by examples of freshwater and marine sediments. Some general recommendations are derived for supporting an efficient evaluation of ^{210}Pb activity concentrations measured in sediments.

It should be emphasized that a number of non-radiological steps, that are not the subject of this paper, may also crucially influence the accuracy of the chronology established, including the selection of the sampling site(s), the coring itself, the transport, storage and handling of the cores, the sample preparation and the quality of the analyses of non-radiological sediment properties.

2. Materials and methods

2.1. Principle of the methodology

Lead-210 in recent sediments originates from two different sources. First, it is produced by decay of ^{226}Ra being part of the sediment minerals. Activity concentrations of this fraction (often called ‘supported’ ^{210}Pb) are time-independent, as they are in equilibrium with its long-lived precursor(s) within the ^{238}U decay chain. The second fraction originates from the decay of atmospheric ^{222}Rn into ^{210}Pb. After dry deposition or precipitation scavenging this radionuclide in the water column readily adsorbs to suspended matter which may be incorporated in sediments. This ‘unsupported’ or ‘excess’ ^{210}Pb decays with 22.3 y half-life and thus can be used to establish sediment geochronologies.

2.2. Estimation of sedimentation rates

There are two models which have been widely and successfully applied: the ‘constant initial concentration (c.i.c.)’ model (Krishnaswamy et al., 1971), also

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referred to as 'constant flux/constant sedimentation rate' model (Robbins, 1978), and the 'constant rate of supply (c.r.s.)' (Appleby and Oldfield, 1978) or 'constant flux' model (Robbins, 1978). Both approaches are based on observations that annual deposition rates of atmospheric ^{210}Pb at a given location are quite constant (e.g. Turekhian et al., 1977; Rangarajan et al., 1986) and assume that, as a consequence, the annual flux density of excess ^{210}Pb fixed to particulates across the water-sediment interface is time-invariant. It should be noted, however, that inter-annual variations of ^{210}Pb deposition rates may exceed a factor 2 (Winkler and Rosner, 2000).

The c.i.c.-model additionally assumes that sedimentation rates and sediment densities do not vary with time. Then, concentrations of excess ^{210}Pb, $C_{ex}(z)$, in the sediment are constant initially and decline exponentially with depth z as

$$C_{ex}(z) = C_{ex}(z = 0)\cdot e^{-\frac{\lambda}{S}\cdot z} \tag{1}$$

where λ denotes the decay constant of ^{210}Pb and S the sedimentation rate.

The c.r.s.-model, on the other hand, focuses on the case of varying sedimentation rates. The age of a sediment at depth z, $t(z)$, then is given as

$$t(z) = \frac{1}{\lambda}\cdot \ln \frac{\int_0^\infty C_{ex}(z')\mathrm{d}z'}{\int_z^\infty C_{ex}(z')\mathrm{d}z'} \tag{2}$$

Modifications of these models are available to take into account a gradual post-depositional compaction of the sediment (e.g. Robbins and Edgington, 1975; Lu, 2007) and, by using a mass sedimentation rate (e.g. Oldfield et al., 2003), variations in sediment densities.

It should be emphasized that both models are complementary. Whereas any deviations from the 'ideal' exponential decrease with depth of the excess ^{210}Pb within the framework of the c.i.c.-model is assumed to be of statistical nature only (including inter-annual variability of atmospheric deposition rates), it is interpreted to reflect a true variation in the sedimentation regime with the c.r.s.-model.

More recently, the SIT ('Sediment Isotope Tomography') methodology has been developed and applied successfully to various sediments (Carroll and Lerche, 2003). Based on the signal theory of linear systems it estimates both the input function (here: ^{210}Pb supply during sediment deposition) and the system's properties (i.e. sedimentation regime) from fitting Fourier series functions to its response function (i.e. ^{210}Pb concentrations within the sediment sequence). Obviously, such an inverse modelling approach allows for time variations of both the flux density of excess ^{210}Pb across the water-sediment interface and of the sedimentation rates. Inverse modelling does not need any prior information on the system, but extracts this completely from the response function. On the one hand this may be regarded as a major advantage of the SIT methodology, as it does not rely on potentially unsubstantiated assumptions on the sedimentation regime. On the other hand, its results must be checked against independent time markers, since results of the numerical algorithms used do not necessarily represent physical reality.

2.3. *Analysis of ^{210}Pb*

Traditionally, the analytical procedure was based on either radiochemical separation of the ^{210}Pb and measurement of its short-lived beta-emitting daughter ^{210}Bi or on alpha-spectrometric counting of its decay product ^{210}Po separated from the sediment after it had attained secular equilibrium with ^{210}Pb – both laborious and time-consuming methods (Joshi and Mudroch, 1988). The commercial availability of advanced planar ('n-type') semi-conductor detectors which enable the gamma-spectrometric analysis of the 46.5 keV decay line of ^{210}Pb with high relative efficiency, however, has drastically reduced time and effort needed. Moreover, the gamma-spectrometry allows a simultaneous determination of the concentrations of supported ^{210}Pb by measuring its precursor ^{226}Ra – either directly via its 186 keV-line or via ^{214}Pb after establishment of decay equilibrium with ^{222}Rn (Kirchner and Ehlers, 1998). Based on our own experience, detection limits are ca. 5–10 mBq g^{-1} for well type detectors and may become somewhat lower for planar detectors if sufficient material (ca. 50 g) is available. Thus, detection limits may be considerably higher than those achievable with alpha-spectrometrical counting of radiochemically separated ^{210}Po. As a consequence, gamma-spectrometry may be of limited use in areas showing small atmospheric deposition rates of ^{210}Pb, e.g. small islands and locations close to oceans.

Knowledge of the detection limit of the analytical equipment which is available for analyzing a specific core is essential for the decision on how to section a core for the ^{210}Pb measurements. Obviously, smaller layers provide a better time resolution of sedimentation rates, but in aged sediments sample sizes may become too small to allow detection of the small fraction of excess ^{210}Pb not already decayed. For a given flux density of excess ^{210}Pb into the sediment, its concentrations are inversely related to the sedimentation rate. As a consequence, some prior guess of the sedimentation rate is needed to section the core such that the slices become not too small for ^{210}Pb analysis, but also provide high resolution data on the sedimentation regime. Such an independent estimate may be available from lithological analyses of the core (e.g. varves). Otherwise, the core should be sectioned into small slices and starting with the youngest sediment measured layer by layer. If ^{210}Pb activities approach the detection limit, layers can be combined prior to analysis.

3. Results and discussion

There are a number of biotic and abiotic effects which may influence the distribution of excess ^{210}Pb in the sediment column including post-depositional diffusion of ^{210}Pb, bioturbation or compaction of the sediment, grain size effects, remobilisation of sediments, and loss of material during sampling. Sedimentation regimes may also be affected by human activities, such as ship traffic, (episodical) discharge of waste water which may even provide an additional source of ^{210}Pb and human recreational activities.

In the following, examples are given to illustrate some of these complications and to discuss strategies to cope with such effects. Because of the advantages discussed previously, all radionuclide concentrations presented in the following have been gained by gamma-spectrometry.

3.1. *Example 1: Lake Kinneret*

The core presented in the following has been taken from the deep central part of the lake in December 1994 (Kirchner et al., 1997). Taking into account the lower density of the top core layer, the ^{210}Pb derived sedimentation rates are almost constant over the major part of the core as documented by the good fit of the c.i.c.-model (Fig. 1, left). It must be emphasized, however, that the ^{210}Pb methodology does not allow establishing an absolute sediment chronology, since the well-known problem of a potential loss of material at core top can not be identified from the depth distribution of this radionuclide. This problem can be solved by combining the ^{210}Pb derived information on sedimentation rates with the information gained from concentrations of ^{137}Cs in the core. This anthropogenic radionuclide shows marked peak concentrations (Fig. 1, right) corresponding to the maximum of atomic weapons testing fallout in 1963 and to the Chernobyl accident in 1986. From the depths of these peaks, mean sedimentation rates of 0.5 cm y^{-1} (1994–1986) and 0.52 cm y^{-1} (1986–1963), respectively, are derived which also indicate a time-invariant sedimentation regime between 1963 and the time of sampling. Least-squares fitting of the c.i.c.-model to the depth profile of ^{210}Pb$_{ex}$ (1–19 cm) results in a mean sedimentation rate of 0.525 ± 0.068 (s.d.) cm y^{-1}. Such a close agreement of a ^{210}Pb based

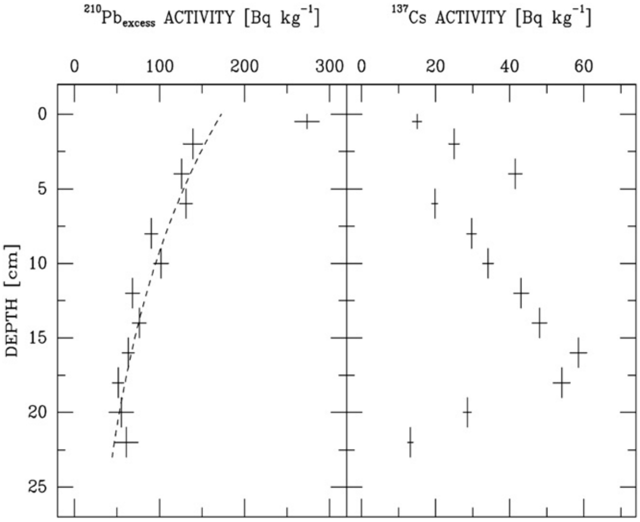

Fig. 1. Concentrations of excess ^{210}Pb (left) and ^{137}Cs (right) in a sediment core taken from Lake Kinneret, Israel (Kirchner et al., 1997); the dashed line (left figure) indicates a least-squares fit (0–19 cm) of the c.i.c.-model.