LEADAT: a MATLAB-based program for lead-210 data analysis of sediment cores

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Abstract
The program described herein, LEADAT, serves to calculate sediment date and sedimentation rate using the $^{210}$Pb method for the studies of environmental processes and pollution history on a time scale of 100 ~ 150 a. The program written in MATLAB (Version 7.0) permits the user to select the principal models of $^{210}$Pb method, i.e., the constant fluxes of sediment and lead-210 (CF-SL) model, the constant flux of lead-210 (CFL) model, the constant initial concentration of lead-210 (CICL) model and the two-layer mixing (TLM) model. Although appropriate model selection is essentially based on understanding of the sedimentary processes, the pattern of the excess $^{210}$Pb profile is also helpful for model selection. The excess $^{210}$Pb profiles for two sediment cores collected from a brackish lake and an embayment respectively are used to demonstrate the application of the program. With a graphical user interface, the program can be easily executed. Not only ASCII but also graphical output can be generated by means of the program. Meanwhile, the code can be modified easily for extension.

Key words: Lead-210 method, sediment dating, sedimentation rate, environmental change

1 Introduction

Tracking the history of man’s impact on the environment, it is clear that in many places the period of greatest impact and most dramatic transformation lies within the last 150 a (Oldfield and Appleby, 1984). Many aspects of this impact can be reconstructed by sediments which can be regarded as good archives of environmental processes and their impacts. Thus, the establishment of detailed and accurate geochronologies of sedimentation in other words, the estimation of sedimentation rate is of central importance in constructing a continuum of insight into environmental processes and pollution history (Oldfield and Appleby, 1984; Fox et al., 1999; Jha et al., 1999; Abril, 2004; Plater and Appleby, 2004; Meng et al., 2005; Xiong et al., 2005). The information of sedimentation is also valuable in modelling pollutants in waters (Matsumoto, 1988; Širca et al., 1999; James, 2002; Gu et al., 2003).

One of the most promising methods for estimating the sedimentation rate over a time span of 100 ~ 150 a is by means of $^{210}$Pb, a member of the naturally occurring $^{238}$U decay series with a half-life of 22.3 a. The $^{210}$Pb method was first outlined by Gold-
berg 1963 applied to laking sediments by Krishnaswamy et al. 1971 and introduced into marine sediments by Koide et al. 1972. The $^{210}$Pb method has proved its reliability in a large number of cases in environments presenting either constant or variable sedimentation rate. Sanchez – Cabeza et al. 1999. Four models, the CFSL model, the CFL model, the CICL model and the TLM model have been widely used by the researchers who are interested in temporal records of chemical and biological processes in environments presenting either constant or variable sedimentation rate. Sanchez – Cabeza et al. 1999. Four models, the CFSL model, the CFL model, the CICL model and the TLM model have been widely used by the researchers who are interested in temporal records of chemical and biological changes in ecosystems. For the CFSL, CFL, and CICL models, sediment mixing is generally assumed absent or insignificant. When sediment mixing is observed, the TLM model is often used. Sanchez – Cabeza et al. 1999. Masqué et al. 2002. In addition, it is a necessary and common practice in the published literature to use some anthropogenic nuclides e.g. $^{137}$Cs, $^{239}$Pu, $^{241}$Am or other time-markers as independent validation of the $^{210}$Pb-derived results. Robbins 1978. Matsumoto 1987. Ritche and McHenry 1990. Abril 2004. Plater and Appleby 2004.

The LEADAT serves to calculate sedimentation rate and sediment date based on the $^{210}$Pb profile data. Meanwhile, two sediment cores are selected to illustrate its application especially model selection and validation.

2 Calculation models

The concentration of excess $^{210}$Pb in sediment at the depth $x$ cm may be described by

$$Q(x) = \left( \frac{S}{F} \right) t e^{-\lambda t}$$

where $Q(x)$ dpm/g is the concentration of excess $^{210}$Pb at the depth $x$, $t$ dpm/cm$^2$ is the flux of excess $^{210}$Pb at the time $t$, $S$ is the sedimentation rate at the time $t$, and $\lambda$ is the decay constant of $^{210}$Pb by Krishnaswamy et al. 1971.

To remove the compaction effect, the cumulative mass of sediment hereinafter massdepth g/cm$^2$ is used instead of depth. Christensen 1982. Matsumoto 1987. 1988. Mass sedimentation rate $S$ g/cm$^2$ is correspondingly introduced to replace the linear sedimentation rate cm/a. Equation 1 can then be transformed into

$$Q(m) = \left( \frac{S}{F} \right) t e^{-\lambda t}$$

where $Q(m)$ dpm/g is the concentration of excess $^{210}$Pb at the massdepth $m$.

Assuming that a constant flux of excess $^{210}$Pb from the waters to the sediment is coupled also with the constant mass sedimentation rate, namely $Q(m) = (S/F)t = Q_0 e^{-\lambda m}$ the excess $^{210}$Pb concentration in the sediments would vary exponentially in accordance with the formula

$$Q(m) = \frac{Q_0}{S} t e^{-\lambda m}$$

where $Q_0$ dpm/g is the concentration of excess $^{210}$Pb at the sediment surface. The sedimentation rate can be determined by using the least-squares fit. This model has been called the “simple” model by Krishnaswamy et al. 1971. Matsumoto 1975. Matsumoto and Wong 1977. Robbins 1978. or the constant flux constant sedimentation rate the ratio of cf to cs model Appleby and Oldfield 1983. Herein, this model is called the constant fluxes of sediment and lead-210 CFSL model.

For the changeable mass sedimentation with a constant supply of $^{210}$Pb i.e. $Q(m) = Q_0$ Eq. 4 can be transformed into

$$Q(m) = \frac{Q_0}{S} t e^{-\lambda m}$$

The age of sediments at the massdepth is then given by

$$t = \frac{1}{\lambda} \ln \left[ \frac{Q(m) dm}{Q(m) dm} \right]$$

The sedimentation rate at the massdepth is calculated.
from the formula
\[ \mathcal{S} t = \frac{\int e^{-At}}{m} = \frac{\lambda}{m} \int_0^t dm / \mathcal{Q} m. \]
This model was termed the constant flux model by Robbins 1978 or the constant rate of supply model by Appleby and Oldfield 1978 1983 Appleby et al. 1979. Herein it is called the constant flux of lead-210 CFL model. This model was initiated by Goldberg 1963 and was set out in more detail by Appleby and Oldfield 1978 and Robbins 1978.

Another alternative hypothesis used for excess 210 Pb profiles assumes that sediments have a constant initial excess 210 Pb concentration irrespective of changes in the mass sedimentation rate. The excess 210 Pb at the massdepth may therefore be expressed as
\[ \mathcal{Q} m = \mathcal{Q} 0 e^{-At}. \]
The age \( t \) at the massdepth is then calculated using the formula
\[ t = \frac{1}{\lambda} \ln \frac{\mathcal{Q} 0}{\mathcal{Q} m}. \]
Accordingly the mean sedimentation rate can be calculated using
\[ \mathcal{S} t = \frac{m}{t}. \]
This model has been termed the constant initial concentration cic mode by Appleby et al. 1979 or the constant specific activity model by Robbins 1978. Herein it is called the constant initial concentration of lead-210 CICL model.

As mentioned previously the sedimentation rates calculated from the above models are valid when the sediment column is not disturbed. Where mixing has occurred for most studies Appleby and Oldfield 1992 Sanchez - Cabeza et al. 1999 Masqué et al. 2002 a one-dimensional advection - diffusion model is used
\[ \frac{\partial \mathcal{Q} m}{\partial t} = \frac{\partial}{\partial m} \left( D \frac{\partial \mathcal{Q} m}{\partial m} \right) - S \frac{\partial \mathcal{Q} m}{\partial m} - \lambda \mathcal{Q} m \]
where \( L \) cm \( ^2 \) is a diffusive mixing coefficient \( \Sigma \) g cm \( ^2 \) is the mass sedimentation rate and \( L \) cm is the mixing depth. Herein the excess 210 Pb profile might be considered as a two-layer system with a surface mixed layer extending to the mixing depth and a layer below it where mixing effect is generally assumed negligible Masqué et al. 2002. Thus assuming the diffusive mixing coefficient to be steady state and constant in the mixed layer it can be induced by the following equation by Cochran 1985 Roberts et al. 1997
\[ \mathcal{Q} m = \mathcal{Q} 0 e^{D \frac{i}{L} m}. \]
The CFL CFL and CICL models are still applicable to calculating the sedimentation rate of the sediments below the surface mixed layer Appleby and Oldfield 1992. Herein this model is called the two-layer mixing TLM model.

3 Model selection and validation

Reliable interpretation of 210 Pb profiles is basically dependent on understanding the relevant processes controlling the delivery of sediments and 210 Pb on a time scale of 100 ~ 150 a. Meanwhile independent evidence is generally necessary to verify if appropriate models and reasonable assumptions have been selected Smith 2001.

The applicability of the above models might also be evaluated by a tentative procedure to inspect the profile of excess 210 Pb see Table 1 and Fig. 1. When a linear profile of excess 210 Pb is shown if the concentration is plotted on a logarithmic scale the sedimentation rate can be determined using the CFSL mode curve a in Fig. 1. In this case actually similar results can be derived from the CFSL CFL and CICL models. For nonlinear and non-monotonic profiles curve b in Fig. 1 application of the CFL model may be considered. If the CICL model is applicable the profile of excess 210 Pb must show a strictly
monotonic decline with massdepth curve c in Fig. 1. In addition, the applicability may be also examined based on the excess $^{210}$Pb flux. For the CFSL and CFL models, excess $^{210}$Pb flux is constant and comparable with the atmospheric flux or nearby cores. In the case of the CICL model, the excess $^{210}$Pb flux is in proportion to the sedimentation rate. This case may often occur in situations where sediment focusing takes place (Appleby and Oldfield, 1983).

### Table 1. Characteristics of the $^{210}$Pb models

<table>
<thead>
<tr>
<th>Model</th>
<th>Initial activity of excess $^{210}$Pb</th>
<th>Sedimentation rate</th>
<th>Flux of excess $^{210}$Pb</th>
<th>Rate of surface mixing</th>
<th>Profile of excess $^{210}$Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>CFSL</td>
<td>constant</td>
<td>constant</td>
<td>constant</td>
<td>0</td>
<td>linear [curve a in Fig. 1]</td>
</tr>
<tr>
<td>CFL</td>
<td>variable</td>
<td>variable</td>
<td>constant</td>
<td>0</td>
<td>nonlinear and non-monotonic [curve b in Fig. 1]</td>
</tr>
<tr>
<td>CICL</td>
<td>constant</td>
<td>variable</td>
<td>variable</td>
<td>0</td>
<td>nonlinear but monotonic [curve c in Fig. 1]</td>
</tr>
<tr>
<td>TLM</td>
<td>constant/variable</td>
<td>constant/variable</td>
<td>constant/variable</td>
<td>constant/variable = $O$ homogenized in the surface layer and linear/non-linear below if curve d in Fig. 1.</td>
<td></td>
</tr>
</tbody>
</table>

![Fig. 1. Pattern of excess $^{210}$Pb profiles.](image)

In practice, due to its relative simplicity and robustness, the scatter within certain extent of $^{210}$Pb data has little influence on the CFSL model is widely used to obtain a mean sedimentation rate although the concentration of excess $^{210}$Pb does not show a perfect linear profile. The accuracy of dating certainly declines with the increase of non-linearity of excess $^{210}$Pb profile. The CFL model is a relatively precise model to obtain changing sedimentation rates. A whole profile of $^{210}$Pb measurement is however needed. The CICL model has the lowest applicability for the strictly monotonic profile of excess $^{210}$Pb is rare in the real sedimentary environment.

Homogenized concentrations of excess $^{210}$Pb in the surface layer curve d in Fig. 1 are often attributed to sediment mixing. If so, the TLM model would be applicable to taking account of the effect of sediment mixing. In the TLM model, the CFSL, CFL and CICL models can be combined to calculate the sedimentation rate below the surface mixed layer according to the pattern of excess $^{210}$Pb profile. For example, for the profile as shown by d curve of Fig. 1, the CFSL model may be combined with the TLM model. It should be noted that the estimate of mixing should not be only based on the shape of the excess $^{210}$Pb profile because the homogenized concentrations of excess $^{210}$Pb in the surface layer may be also associated with other processes such as accelerated sedimentation rates (Appleby and Oldfield, 1992; Carroll and Lerche, 2003). Therefore, when mixing is suspected, attention should be paid to check if mixing has really occurred using other evidences such as biological assay, sedimentological observation and nuclide tracers e.g. $^{239+240}$Pu, $^{137}$Cs and $^{234}$Th (Roberts et al., 1997; Lewis et al., 2001).
4 The LEADAT program and its application

4.1 Program description

The LEADAT is programmed with the latest version of MATLAB® Version 7.0 using the SSADM structured systems analysis and design method approach. In the programming each model algorithm is performed module-based.

The LEADAT runs under the MATLAB with a simple graphical user interface Fig. 2. The input and output files are in ASCII format. Meanwhile the user can select the figures to plot in the figure frame which are figures of excess $^{210}\text{Pb}$ depth/mass depth profile sediment date vs. depth/mass depth and sediment date vs. linear/mass sedimentation rate. The graphical outputs are displayed in the individual figure windows on the screen where the user can also customize the appearance of the graphs easily. Calculation models the CFSL, CFL, CICL and TLM models can be selected individually or in combination in the model frame.

It should be noted that the linear sedimentation rate is also calculated by the program according to the data of mass sedimentation rate and water content of the sediment. An first-order analysis also called general error propagation is used in the program to estimate error in the above calculations Binford 1990.

In addition as sediment mixing might homogenize the pollutants in the sediment the significantly mixed sediment would often become unsuitable to constructing the temporal trend of pollution. Therefore two unmixed cores see Table 2 are selected to illustrate the use of the LEADAT program.

4.2 Example 1

The first sediment core Core SJ85 – 9 was collected in a brackish lake Lake Shinji Japan in 1985. The core appears to be undisturbed by physical or biological mixing detailed description see Matsu-moto 1987. As shown Fig. 3 the $^{210}\text{Pb}$ profile is linear and monotonic decreasing with mass depth. In this case the CFSL, CFL and CICL models are all applicable. Indeed the mean sedimentation rates derived from the CFSL, CFL and CICL models are in good agreement which are $0.12 \pm 0.01$, $0.10 \pm 0.02$, and $0.11 \pm 0.02$ g/(cm$^2$·a) respectively. In this core a distinct peak of $^{137}\text{Cs}$ indicating the year 1963 is observed at the 10 ~ 15 cm section. A mean sedimentation rate $0.09 \pm 0.03$ g/(cm$^2$·a) can be inferred from the $^{137}\text{Cs}$ peak. Obviously it is consistent with the values derived from the $^{210}\text{Pb}$ method within calculation error. Meanwhile the consistency can also be seen from the age/mass depth curve shown in Fig. 4 although the CFL-derived dates show relatively large errors for the deep layer.

4.3 Example 2

The other sediment core Core M1 was collected in the Ise Bay Japan in 2004 Lu and Matsu-moto 2005. As a semi-closed embayment the water circulation and exchange near the sea bottom is limited and hypoxia occurs seasonally Fujiwara et
Table 2. Two examples of input file for Cores SJ85 - 9 and M1:

<table>
<thead>
<tr>
<th>Line</th>
<th>File content</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Core SJ85 - 9</td>
<td>core name</td>
</tr>
<tr>
<td>2</td>
<td>1985 - 09 - 15</td>
<td>sampling time: yyyy - mm - dd</td>
</tr>
<tr>
<td>3</td>
<td>cm g dpm</td>
<td>units of length, mass and radioactivity</td>
</tr>
<tr>
<td>4</td>
<td>depth, depth_err, Pbex, Pbex_err</td>
<td>names of data columns</td>
</tr>
<tr>
<td>5</td>
<td>2.5, 2.5, 35.96, 1.30, 90.40, 1.0, 0.47, 83.97</td>
<td>data field</td>
</tr>
</tbody>
</table>

Notes: Depth refers to the depth at section center. Error of depth is given by the section thickness. WC denotes water content (\%).

Fig. 3. Profiles of excess $^{210}$Pb for Cores SJ85 - 9 and M1.

The core seems to be undisturbed by physical or biological mixing. As shown in Fig. 3, the $^{210}$Pb profile is nonlinear and non-monotonic decreasing with massdepth. In this case, both the CF-SL and CICL models are not applicable. As shown in Fig. 4, the CFL-derived date/massdepth curve shows the consistency between the $^{137}$Cs and $^{210}$Pb methods. In addition, the temporal change of the CFL-derived sedimentation rates sees Fig. 5 has showed an increase especially since the 1950s, which is in good agreement with the intensification of reclamation in the coastal area of Ise Bay during that period.

5 Concluding remarks and future work

The LEADAT is written in MATLAB Version 7.0. It serves to calculate sedimentation rate and sediment date using the $^{210}$Pb method. Four principal models of the $^{210}$Pb method i.e., the constant fluxes of sediment and lead-210 CFSL model, the constant flux of lead-210 CFL model, the constant initial concentration of lead-210 CICL model and
Fig. 4. Date vs. mass-depth curves for Cores SJ85-9 and M1. The triangle, cross and circle marks refer to the results derived from the CFSL, CFL and CICL models respectively.

The two-layer mixing TLM model are included in the program. With a graphical user interface the program can be easily executed for the user can select options primarily with mouse-clicks or through interactive dialogue boxes. Not only ASCII but also graphical output can be generated by means of the program. In addition the code can be modified easily for extension.

The excess $^{210}\text{Pb}$ profile for two sediment cores are used to illustrate the application of the program. Some tentative methods are proposed for model selection. However appropriate model selection is substantially based on enough understanding of the relevant processes.

Interpretation of the $^{210}\text{Pb}$ profiles would be more complicated when sediment mixing is involved. The present program generally focuses on dating and calculation of sedimentation rate for the unmixed or partially-mixed sediments. In the future an extended version of the program will include more mixing models such as the depth-dependent models (Christenson 1982; Smith et al. 1995) the age-dependent models (Lauerman et al. 1997) now in progress as well as the inductive model (Carroll and Lerche 2003).

Fig. 5. Temporal trend of sedimentation rates for the core M1.

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Appendix

Software availability
Name LEADAT
Developer Lu Xueqiang and Matsumoto Eiji
Year first available 2003

Hardware required PC compatible
      Pentium III with RAM
      256MB or above
Software required Windows 2000/NT4.0/XP
      MATLAB
      Version 7.0
Program language MATLAB
      Version 7.0
Availability on request via email
Cost freeware