

Appendix 1: precision of lead dating

On a centennial time scale, dating of sedimentary layers was carried out based on ^{210}Pb and ^{137}Cs measurements. Concentrations of both nuclides, together with U, Th and ^{226}Ra , were determined by gamma spectrometry. The 2-cm-thick sediment layers were washed in de-ionised water and sieved. The sediment fraction <1 mm was first finely crushed after drying, and transferred into small gas-tight Polyethylene Terephthalate (PETP) tubes having an internal height and diameter of 38 and 14 mm, respectively, and then stored for more than 3 weeks to ensure equilibrium between ^{226}Ra and ^{222}Rn .

The activities of the nuclides of interest were determined using a CANBERRA Ge-well detector, by comparison with the known activities of an in-house standard. Activities of ^{210}Pb were determined by integrating the area of the 46.5 keV photo-peak. ^{226}Ra activities were determined from the average of values derived from the 186.2 keV peak of ^{226}Ra and the peaks of its progeny in secular equilibrium ^{214}Pb (295 keV and 352 keV) and ^{214}Bi (609 keV). In each sample, the ^{210}Pb (unsupported) excess activities were calculated by subtracting the ^{226}Ra (supported) activity from the total ^{210}Pb activity (Sabatier *et al.*, 2008).

The basic methodology of ^{210}Pb dating was established by (Goldberg, 1963). ^{210}Pb precipitates from the atmosphere through ^{222}Rn decay and accumulates in surface soils, glaciers or lakes, lagoons, where successive layers of material are buried by later deposits. Many ^{210}Pb models were proposed, allowing calculating sedimentation rates (Appleby and Oldfield, 1992). In the simplest model, the initial $(^{210}\text{Pb}_{\text{ex}})$, $[(^{210}\text{Pb}_{\text{ex}}) = (^{210}\text{Pb}) - (^{226}\text{Ra})]$ is assumed constant and thus $^{210}\text{Pb}_{\text{ex}}$ at any time is given by the decay law $[(^{210}\text{Pb}_{\text{ex}}) = (^{210}\text{Pb}^0_{\text{ex}}) \exp^{-\lambda_{210}t}]$. The CFCS model (Goldberg, 1963) supposes a constant ^{210}Pb flux and a constant sedimentation rate. In a logarithmic diagram, $^{210}\text{Pb}_{\text{ex}}$ should define a straight regression line whose slope allows calculating an average sedimentation rate. Using the average sedimentation rate W (mm.a^{-1}), the age T_m of the sediment layer can be calculated, for each depth Z_m (cm).

$$\ln((^{210}\text{Pb}_{\text{ex}}^m)) = \ln((^{210}\text{Pb}_{\text{ex}}^0)) - \left(\frac{\lambda_{210}}{W}\right) \times Z_m, \quad \text{with: } T_m = \frac{W}{Z_m} \quad \text{eqn. 1A}$$

This simple model gives an estimation of the average sedimentation rate.

The peak of ^{137}Cs is search in the top most sediment based on the fact that the depth of the maximum ^{137}Cs activity in the sediment corresponds to the 1963 maximum atmospheric production (Robbins and Edgington, 1975).

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