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Estimation of sedimentation rates based on the excess of radium 228 in granitic reservoir sediments

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ABSTRACT

Knowledge of sedimentation rates in lakes is required to understand and quantify the geochemical processes involved in scavenging and remobilization of contaminants at the Sediment-Water Interface (SWI). The well-known ^{210}Pb excess (^{210}Pb$_{ex}$) method cannot be used for quantifying sedimentation rates in uranium-enriched catchments, as large amounts of ^{210}Pb produced by weathering and human activities may dilute the atmospheric ^{210}Pb. As an alternative dating method in these cases, we propose an original method based on ^{232}Th decay series nuclides.

This study focuses on an artificial lake located in a granitic catchment downstream from a former uranium mine site. The exponential decay of ^{228}Ra excess (^{228}Ra$_{ex}$) with depth in two long cores yields sedimentation rates of 2.4 and 5.2 cm yr^{-1} respectively. These sedimentation rates lead to the attribution of the ^{137}Cs activity peak observed at depth to the Chernobyl fallout event of 1986. The ^{228}Ra$_{ex}$ method was also applied to two short cores which did not display the ^{137}Cs peak, and mean sedimentation rates of 2.1 and 4.0 cm y^{-1} were deduced. The proposed method may replace the classical radiochronological methods (^{210}Pb$_{ex}$, ^{137}Cs) to determine sedimentation rates in granitic catchments.

1. Introduction

The present study focuses on the mobility of natural radionuclides downstream from a former uranium mine closed at the beginning of the 1980's in the Massif Central (France). The waters from the underground mine works and the former open-pit mine are discharged after physicochemical treatments into the river Besbre, which supplies the artificial lake of Saint-Clément. The treated mine waters contain several radionuclides of the ^{238}U decay series, including ^{238}U, ^{226}Ra and ^{210}Pb that are deposited in bottom lacustrine sediments. Assessing sedimentation rates in the lake is required to understand the geochemical mechanisms involved in natural radionuclides deposition and quantify their fluxes at the SWI. Moreover, sedimentation rates are essential for the development of solute transport models, especially for predicting radionuclides and metals behavior in sediment pore waters.

In sedimentary deposits less than 150 years old, ^{210}Pb is a powerful radioactive tracer for environmental investigations including the dating of lacustrine sediment (Appleby and Oldfield, 1978; Etienne et al., 2013; He and Walling, 1996; Lavrieux et al., 2013; Perga et al., 2010). ^{210}Pb is generally not in secular equilibrium with its parent radionuclide ^{226}Ra. The daughter isotope of ^{226}Ra, ^{222}Rn, is exhaled from soils to the atmosphere and decays to ^{210}Pb, which attaches to aerosols and deposits on the earth's surface via wet and dry deposition leading to excess ^{210}Pb (^{210}Pb$_{ex}$) concentrations (Turekian et al., 1977). In the site studied here, however, the addition of large and variable inputs of ^{210}Pb from the mine, diluting the atmospheric ^{210}Pb, renders the ^{210}Pb$_{ex}$ method unusable.

The artificial radionuclide ^{137}Cs is frequently used to confirm sediment radiometric dating (Appleby et al., 1991; Lavrieux et al., 2013). In western Europe, two temporal markers of ^{137}Cs input can be distinguished: i) the maximum of fallout atmospheric nuclear bomb tests in 1963–1964, and ii) the Chernobyl accident in 1986 (Sabatier et al., 2014; Schuller et al., 1993, 2002).

In this context, the need of a complementary dating method is fundamental for: i) dating recent sediment less than 30 years old

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without significant ^{137}Cs activity maxima, and ii) allocating ^{137}Cs peaks in sediments when they are detected.

The proposed technique is based on the disequilibrium between ^{228}Ra and its ^{232}Th parent ($^{228}Ra_{xs}$) (*i. e.* in addition to supported ^{232}Th) observed in recent sedimentary deposits. A simplified version of the ^{232}Th decay series is given in Fig. 1.

A few applications of the ^{232}Th decay series nuclides for dating lake sediments are reported in the literature. Disequilibrium within ^{232}Th series for environmental studies was proposed by Koide et al. (1973) but only for the couple ^{228}Th/^{232}Th, with ^{228}Ra assumed to be in secular equilibrium with ^{232}Th and (ii) the couple ^{228}Th/^{228}Ra in marine sediments. Rabouille et al. (2009) uses ^{228}Th excess in surface for dating marine sediments. More recently, the excess of ^{228}Ra over ^{232}Th was reported for fluvial sediments and probably due to desorption of radium from soil profiles (Olley et al., 1997). The same disequilibria were used in podzolic soils to probe the kinetics of colloid migration (Rihs et al., 2011).

The aim of the present work is to develop and validate a new method for the determination of sediment accumulation rates based on the decay of $^{228}Ra_{ex}$ in a granitic, uranium-rich environment. Further, we apply the method on two short cores where no other dating method could be used.

2. Materials and methods

2.1. Site description

The studied area is located in the Massif of Bois Noirs, a part of the Massif Central in France (Fig. 2) characterized by a Hercynian granitic horst. The "Bois Noirs Limouzat" uranium ore body is a hosted-vein deposit associated with important quartz veins (Cuney, 1974). The uranium mining extraction began in 1955 and ceased at the beginning of the 1980's.

Treated waters coming from the former uranium mine deposits are discharged into the river Besbre, which supplies the lake of Saint-Clément, located approximately 20 km downstream. Dissolved ^{228}Ra in the Besbre waters sampled downstream from the former mining site are similar to those determined in other tributaries (1–3 mBq L^{-1}, unpublished). These results sustain the fact that ^{228}Ra in the Besbre river are primarily of natural origin.

2.2. Sediment sampling

The two coring sites, C2 (N 46.085597° E 3.688483°) and C4 (N 46.086111° E 3.688250°), were located within the deepest body of the lake (Fig. 2).

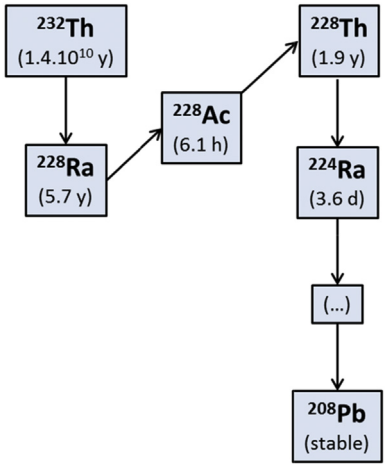

Fig. 1. Simplified decay chain of thorium 232 and his daughter products.

C4 was the deepest sampling point (13–14 m of overlying water) close to the Saint-Clément dam. The sampling area is crossed by the former river bank, and the sediment coverage is highly variable, following the topography. A set of sediment cores were sampled with an UWITEC© hand corer equipped with 60 cm to 3 m long PVC sampling tubes at different depths. The four sediment cores described in this paper, displayed a homogeneous mud composition. The first two cores, 2011C2 and 2013C2, were 195 and 90 cm long, respectively. The other two cores were shorter: 2014C2 (25 cm length), 2014C4 (40 cm length).

Sediment cores were horizontally sliced and stored at 4 °C before discrete sampling. In the laboratory, selected sediment slices were dried at 110 °C and grounded in an agate mortar, and sealed in polyethylene tubes ready for gamma-counting.

2.3. Sampling of lake's suspended particulate

Two 14 L samples of overlying waters at 1 and 9 m depth were collected with a Ninskin© bottle and stored in a polyethylene tank for transportation to the laboratory. Back in the laboratory (less than one hour after sampling), the water samples were mixed and filtered through 0.45 μm Millipore membrane filters, which were retained for analysis.

2.4. Gamma spectrometry

Aliquots of 6–7 g of sediment samples were measured in the wells of very low background high efficiency Ge detectors (Canberra©) at the "Laboratoire Souterrain de Modane" (LSM) in the French Alps (Cazala et al., 2003; Reyss et al., 1995). Protected from cosmic radiations by 1700 m of rock overburden, backgrounds as low as 0.5 counts per minute from 40 keV to 3000 keV were measured. ^{210}Pb, ^{234}Th (assumed in equilibrium with ^{238}U), ^{214}Bi and ^{214}Pb (^{226}Ra), ^{228}Ac (^{228}Ra), ^{212}Pb and ^{208}Tl (^{228}Th), ^{137}Cs and ^{7}Be were generally analyzed after counting times of one day for sediment samples. Given that the amount of suspended particles recovered after water filtration was low (a few mg in 14 L) the activities of radionuclides were expected to be as low as a few mBq L^{-1} (Cazala et al., 2003; Dominik et al., 1989). Consequently, measurements required 3–4 days to determine activities with a one sigma statistical error lower than 10%. The gamma detectors were calibrated using several standards from IAEA (RGTh-1, RGU-1, 375, Soil 6) and from National Institute of Standard and Technology (NBS4350b and NBS4353). The use of standards for the isotopes of interest measured in similar geometry and weight for the efficiency calibration rule out bias in the measurements.

A specific measurement was performed at LSCE (Gif-sur-Yvette) with a high resolution GEM-FX Ortec© detector for a precise quantification of the ^{241}Am gamma ray at 59.5 keV in the presence of ^{234}Th at 63.3 keV.

2.5. Alpha spectrometry

Sediment and suspended particles samples were totally dissolved using HCl, HF and $HClO_4$ (suprapur Merck©). ^{229}Th and ^{236}U spikes (Amersham) were added and standard procedures employed for measuring ^{238}U, ^{234}U, ^{230}Th and ^{232}Th (Ku and Broecker, 1969). Alpha spectra were obtained by counting in α grid chambers at LSCE.

2.6. Ion coupled plasma-mass spectrometry (ICP-MS) analyses

U and Th concentrations were also measured at the SARM-CRPG