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Studies of ^{137}Cs absorption-desorption processes in sediments, using γ -ray spectroscopy

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Abstract

The description of ^{137}Cs behaviour during water to sediment transport generally involves the characterization of the sorption process in a particular hydro-geological system by certain prescribed kinetics parameters. In the present work the kinetics of radiocesium sorption and desorption and the corresponding radiocesium distribution profiles in lake sediment cores were investigated. Radiocesium reaches rapidly (< than 5 d) at a depth which does not exceed 4 cm. The desorption of radiocesium was found to depend upon cation concentrations. Empirical laws were derived both for cesium adsorption and desorption.

1 Introduction

To study the migration of a radionuclide in an aquatic environment, it is necessary to quantify the behaviour of a particular solute species relative to the carrier lake or river due to physico-chemical sorption processes. Since in most of the cases it is impractical to study the sorption processes in-situ, we resort to the results of laboratory simulation experiments and then extrapolate to predict the behavior of the real system.

In the aquatic environment, radioisotopes are carried by water. The migration of radionuclide is influenced by various processes, predominantly by reversible and irreversible fixation to sediments. Thus sediments are an important component of the aquatic environment, functioning either as a sink or as temporary repository for radionuclides. Of the most important nuclides for radiation protection is radiocesium (^{137}Cs). Once in water, ^{137}Cs migrates to bottom sediments by direct sorption or by sorption on suspended solids that later deposit

on the bottom. The sediments usually act as a sink for radiocesium, but they may also act as a source, when they are resuspended in periods of turbulence or when desorption occurs.

The redistribution of ^{137}Cs in lake sediments was investigated by Brobberg [1] and Maubert et al [2]. Kirikopoulos et al [3] studied experimentally the kinetics of ^{137}Cs sorption in lake sediments. The main conclusion of these investigations is that radiocesium is trapped in the first 2-4 cm of the sediment.

The objective of the work presented here was to simulate experimentally the kinetics of radiocesium sorption-desorption processes in lake sediments, with weight given to the study of the desorption process as a secondary source of contamination. Of the possible desorption processes, we consider the exchange with K^+ , since it has been shown by laboratory studies that K^+ is the primary cation in competition with Cs^+ for sorption studies [4].

2 Experimental methods

^{137}Cs was introduced into a tank containing 28 L of lake water. The sediments were put in cylindrical containers, simulating sediment cores, and placed in the bottom of the water tank. Only the tops of the cores was exposed. The activity concentration in water was measured daily by γ -ray spectroscopy.

The profile of radiocesium adsorbed in the sediments was determined with a non-destructive scanning arrangement, described in detail elsewhere [3]. In short, the arrangement comprised a lead collimator in front of a $3'' \times 3''$ NaI(Tl) scintillator detector. The detector was allowed to view radiation coming from the region in front of the collimator's slit. Since the width of the slit was 5 mm, it was possible to measure the radioactivity emitted from 5 mm sections.

In a series of batch experiments, studying the ^{137}Cs adsorption-desorption process in sediments, 1 g quantities of powdered dried sediment matter were placed in bags made of filtered paper and suspended in 400 mL containers filled with solutions of Cs salts (to study adsorption) and K salts (to study desorption) in various concentrations. In the adsorption study, ^{137}Cs was added as a tracer to each of the containers. The activity of the sediments and the activity concentration of the solutions was monitored until equilibrium was reached. In this way, the adsorbed cesium was correlated with the salt concentration of the solutions and the adsorption-desorption isotherms were inferred.

Moreover, the desorption of radiocesium was studied as a function of potassium concentration. In this study, sediment cores were placed in tanks filled with

water of selected K^+ concentrations. The desorption process was monitored with measurements of radioactivity profiles with the scanning -ray arrangement.

The samples from the batch experiments were measured for radiocesium activity with a 1.9 keV resolution (for the 661.65 keV line of ^{137}Cs), 18% efficiency, intrinsic Ge detector. The detector was shielded with 5 cm of lead against background radiation. Standard electronics were used and the spectra were accumulated in 1024 channels. The analysis was performed with a personal computer. The detector was calibrated for efficiency versus energy with a standard $129.5 \text{ kBq L}^{-1} \text{ }^{152}\text{Eu}$ solution. Counting time required to accumulate a spectrum with adequate statistics ranged from 1,000 to 10,000 s.

3 Results and discussion

A typical radiocesium concentration profile measured in a sediment core with the scanning apparatus is given in Fig. 1. From the inspection of Fig. 1, it is inferred that the absorption of cesium proceeds to a depth of 2-4 cm. The absorption of ^{137}Cs was observed to advance rapidly during the first 3 to 5 d. Most of radiocesium is adsorbed in the sediments during the first 3 d. Moreover, it was observed that the distribution of ^{137}Cs could be described by the exponential function

$$C(x) = c(0)e^{-\lambda x} \quad (1)$$

where the parameter was estimated as $= (0.340 \pm 0.005) \text{ cm}^{-1}$. Since the cesium distribution displayed an exponential behaviour, the total adsorbed radiocesium activity in the sediment core was calculated measuring the counting rate R , which the 661.65 keV peak of ^{137}Cs induces to the Ge detector, following the procedure described in ref. 3. The kinetics of the absorbed ^{137}Cs in the sediment core is presented in Fig. 2.

The ^{137}Cs adsorption isotherm is presented as the logarithm of the adsorbed cesium per g of sediment vs. the logarithm of cesium concentration (Fig. 3). From these results, it was deduced that the adsorption of cesium in sediments follows Freundlich's empirical law:

$$y = kc^{\frac{1}{n}} \quad (2)$$

where k and n are Freundlich's empirical constants. These constants were determined as $n=1.9 \pm 0.1$ and $k=0.9 \pm 0.6 \text{ kg}^{-1} \text{ L}$.

The Cs desorption in the 1 g sediment samples is presented in Fig. 4, as the remaining radiocesium in the sample is near equilibrium conditions. The

remaining radiocesium, expressed as the Cs activity desorbed per unit of Cs activity absorbed was found to follow the empirical law

$$\ln(C_{s,rem}) = A \ln(C_K) + B \quad (3)$$

where C_K is the K^+ concentration and the empirical constants are $A = -216 \pm 46$ and $B = 991 \pm 213$.

Typical radiocesium concentration profiles measured with the scanning apparatus in sediment cores immersed in K solutions are presented in Fig. 5. The kinetics of the total activity in the cores were also calculated. Fig. 6 is a typical plot for the presentation of desorption kinetics in a sediment core, immersed in a K^+ solution of $10^{-3} \text{ kg L}^{-1}$.

According to the present results, radiocesium is immobilised at a distance x from the water-sediment interface, not greater than a few cm. Although cesium is very soluble, its migration in aqueous environments is retarded by the interactions with clay minerals in soils and sediments. The reversibility of the cesium sorption on the clay minerals determines the ability of Cs to be discharged into water by changing environmental conditions.

Concerning the migration of Cs in the sediments, a purely diffusion process should be excluded. For every data set, the correlation of the logarithm of the radiocesium concentration distribution to the square of distance x^2 , as expressed by the correlation coefficient r was not greater than the correlation of the same logarithm to distance x . Also the radiocesium profiles did not evolve dramatically with time, maintaining their shape after the first 5 d.

Although there is no universal selectivity order of cation displacement by ion exchangers, the exchange with K^+ was chosen as an object of the present study, since the observed order of cation displacement of ^{137}Cs was previously found significant [4] as $\text{NH}_4^+ ; \text{H}^+ ; \text{Cs}^+ = \text{K}^+ \gg \text{Ca}^{+2} ; \text{Na}^+ = \text{Mg}^{+2} = \text{Fe}^{+2}$. In our experiments, it was found that ^{137}Cs can be removed from sediments to the limit of $\approx 20\%$ by extraction with solutions of K^+ ion.

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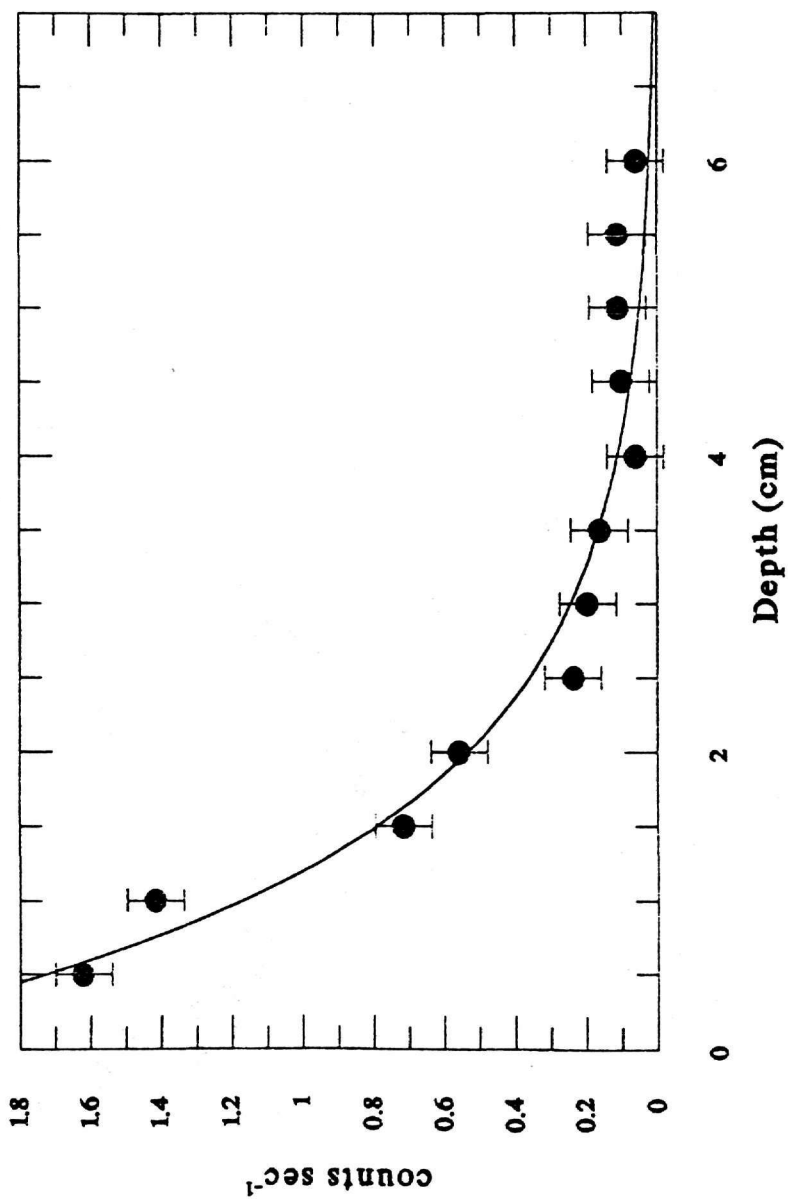


Fig. 1 Typical radiocesium sorption profile in a sediment core.

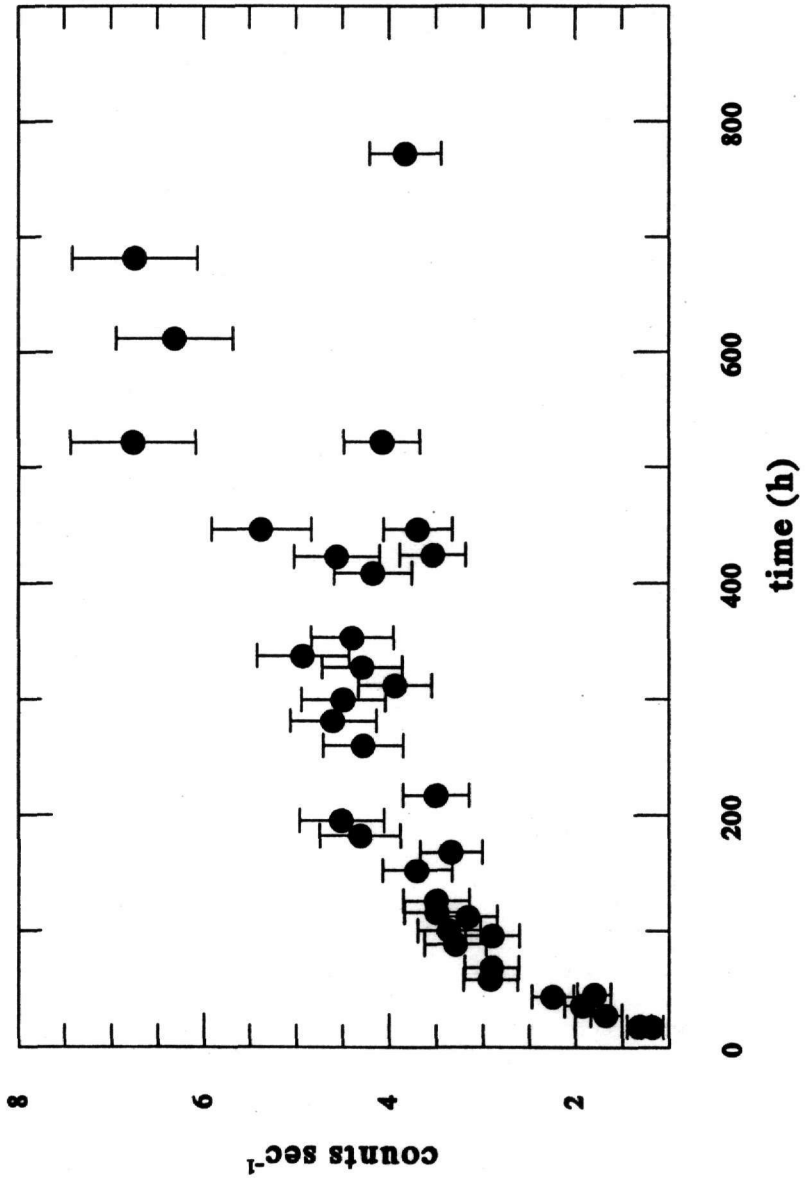


Fig. 2 Kinetics of the absorbed ¹³⁷Cs in a sediment core.

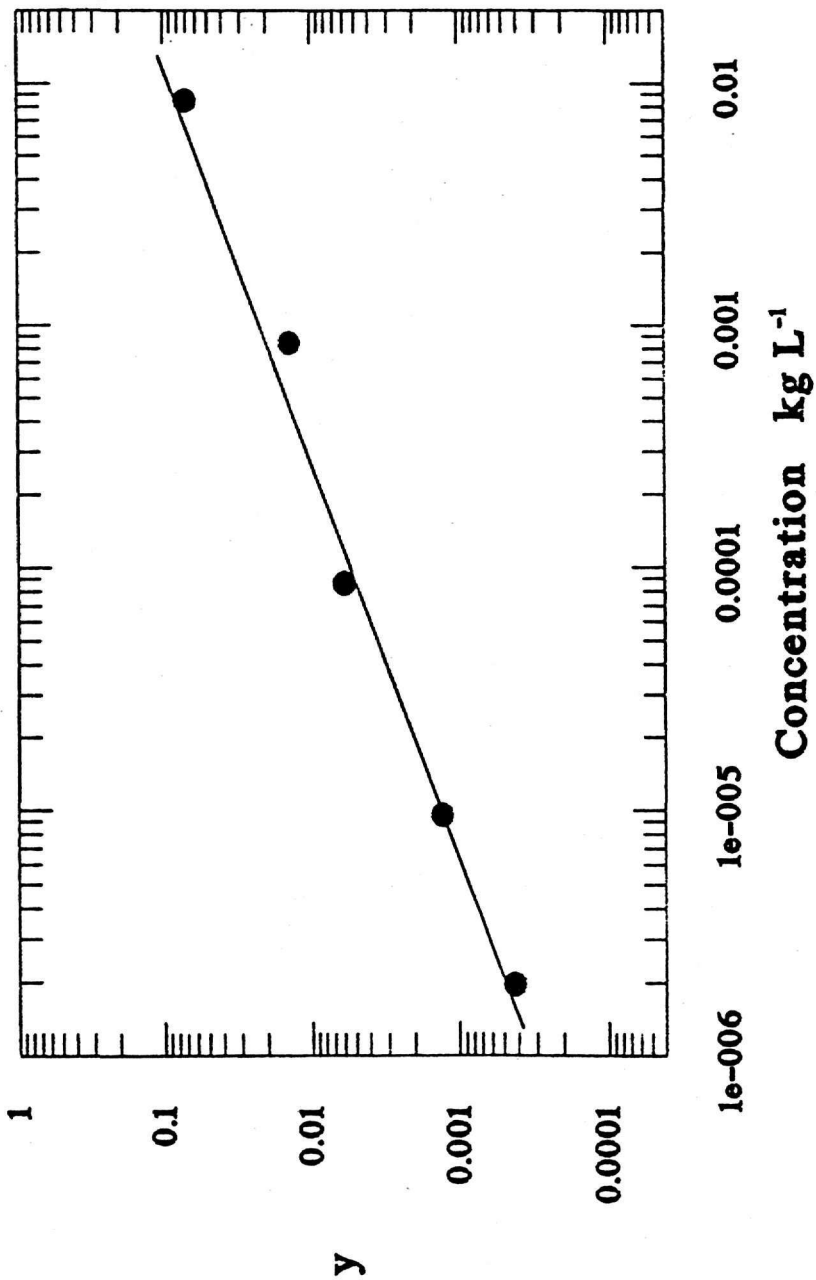


Fig. 3 The ^{137}Cs adsorption isotherm. y is the ratio of radiocesium adsorbed per unit mass of sediment.

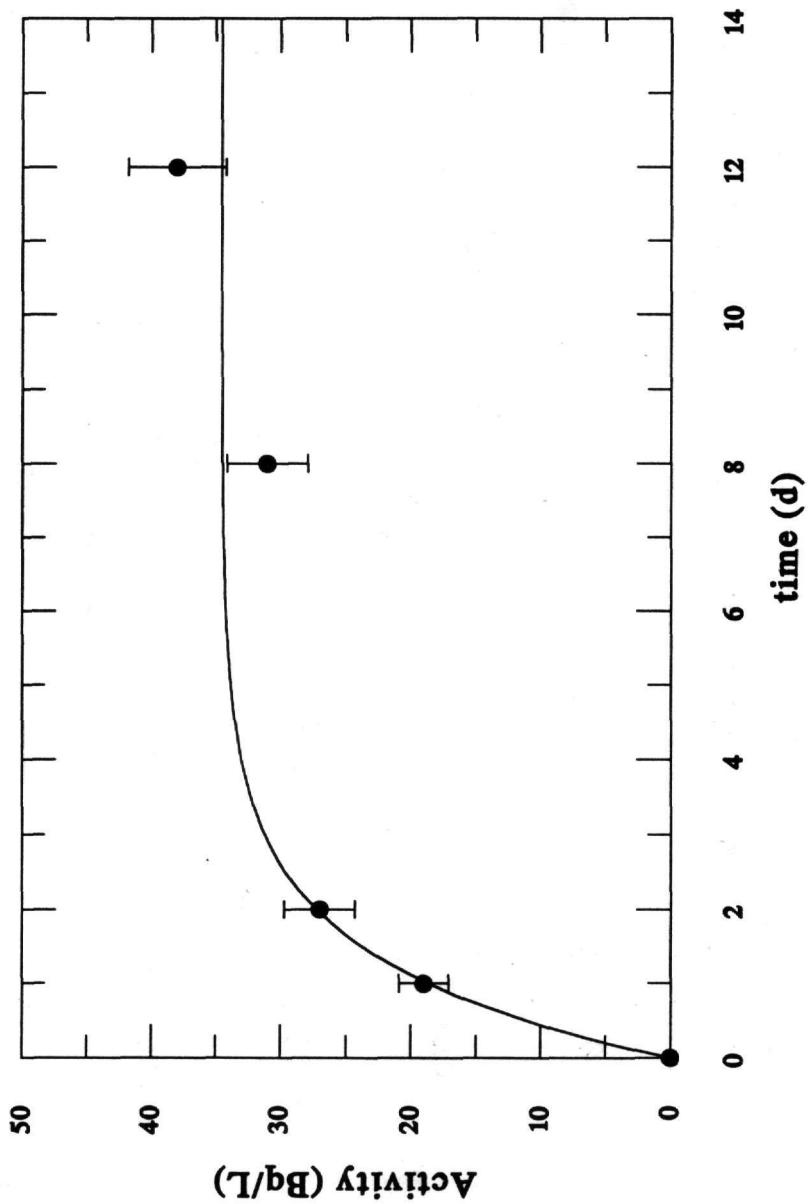


Fig. 4 Cs desorption in the 1 g sediment samples suspended in K^+ solutions as a function of K^+ concentration.

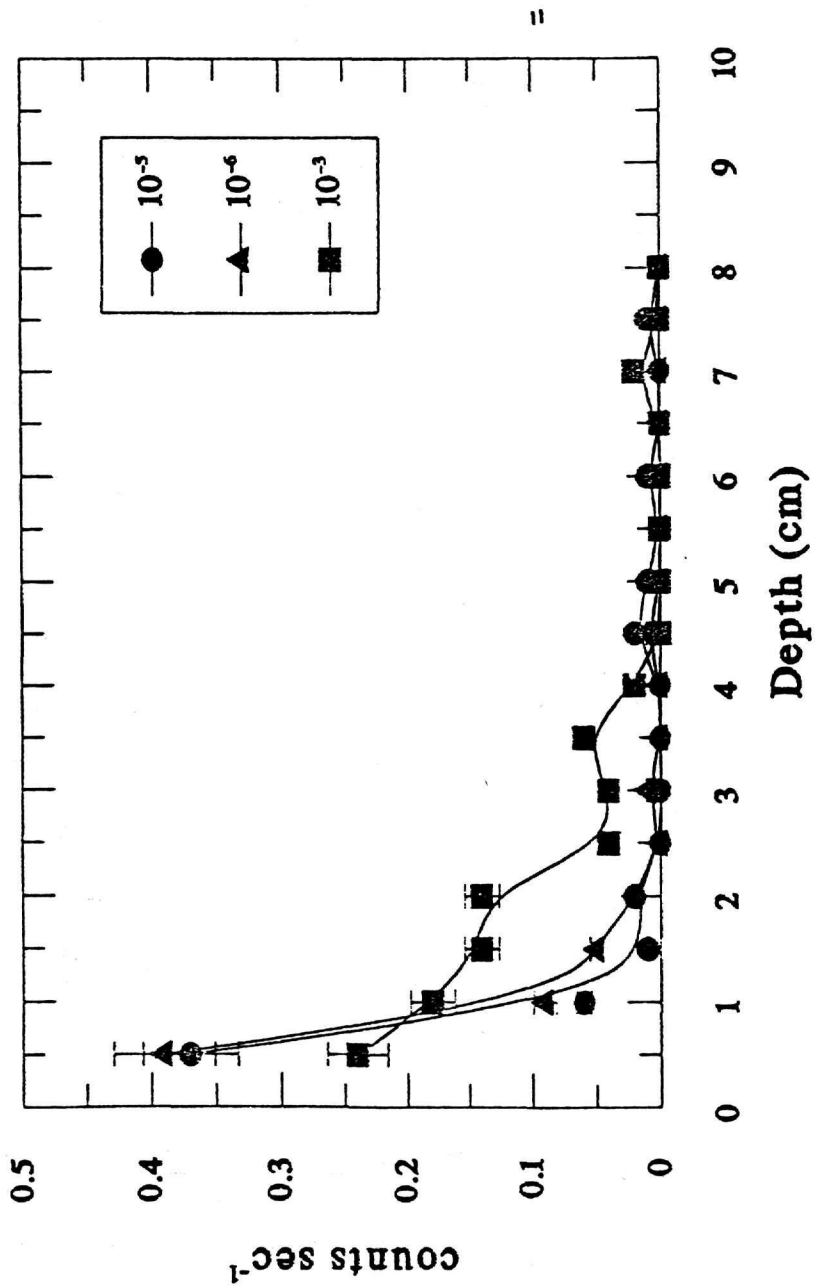


Fig. 5 Radiocesium equilibrium concentration profiles of sediment cores immersed in solutions of different K⁺ concentration.

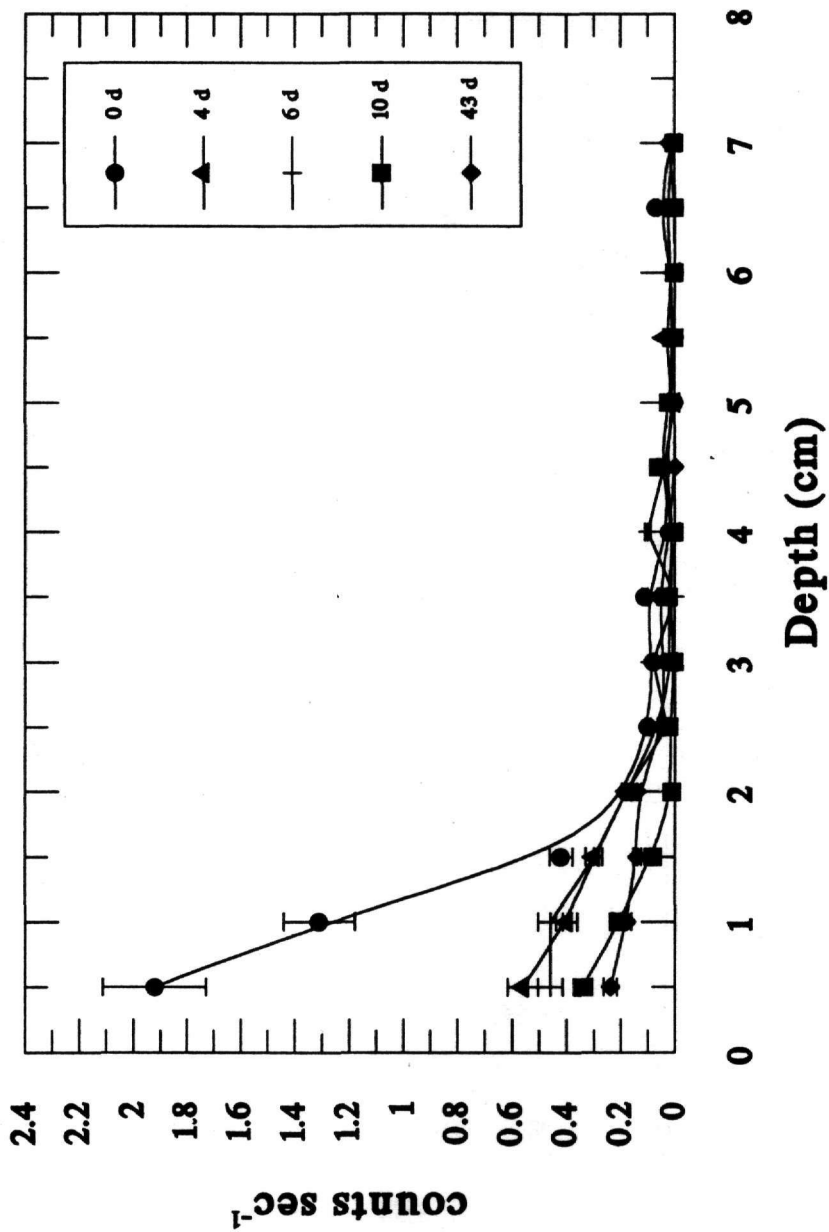


Fig. 6 Evolution of the activity profile of a sediment core immersed in a K⁺ solution of 10⁻³ kg L⁻¹.