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Journal of Environmental Radioactivity 87 (2006) 325–334

JOURNAL OF ENVIRONMENTAL RADIOACTIVITY

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Diffusion of ^{226}Ra and ^{40}K radionuclides reproduced in underwater sedimentary columns in laboratory

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Received 28 July 2005; received in revised form 21 December 2005; accepted 21 December 2005
Available online 20 February 2006

Abstract

The potential radiological impact of the increase of radioactive substances in the environment makes interesting the study of the migration of the contaminant radionuclides in soils and sediments, which are the last receiver system of these substances. By using a battery of sedimentary columns controlled in the laboratory, the diffusion of the ^{226}Ra and ^{40}K radionuclides has been studied, assessing their respective effective diffusion coefficients in a similar sedimentary medium. A decreasing temporal evolution is obtained, associated to the progressive 'fixation' of the radionuclides by the clay minerals of the sediment, followed by a constant tendency. A timescale of the 'fixation' by the sediment is determined, being of the order of days for ^{226}Ra and of the order of months for ^{40}K, so the progressive 'fixation' of ^{40}K by the clay minerals of the sediments is slower than in the case of ^{226}Ra.

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Keywords: Radionuclides; Sedimentary columns; Diffusion; Effective diffusion coefficient

1. Introduction

In addition to the radionuclides coming from the atmosphere, the littoral strip receives through the rivers and the rainfall most of the radioactive pollution, natural or anthropogenic. After the time they remain on the water layer and by means of different mechanisms, the radionuclides end up being deposited in the sediments of the bottom of the sea. Once in the

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doi:10.1016/j.jenvrad.2005.12.010

water–sediment interface, the radionuclides can migrate from the more superficial to the deeper layers. In order to assess this type of pollution in the aforementioned coastal strip it is necessary to know the most relevant phenomena of incorporation and subsequent mobilisation of the radionuclides in the underwater sediments. This is important since the long-term radiation dose received by man due to the presence of radionuclides in the sediments depends on the migration of the radionuclides in the surface layers.

The transference of radioactivity concentration from the aquatic environment to the sediments can occur through different processes, such as uptake by suspended matter and sedimentation, direct precipitation of colloidal forms, direct uptake by adsorption, deposition of organic waste which previously had incorporated the radionuclides.

In porous media, such as sediments, diffusion is one of the main transport mechanisms that causes the postdepositional mobility of the radionuclides. It is a molecular process resulting from the presence of radionuclides in the pore water of the sedimentary medium and also as a consequence of the Brownian movement of the reduced size particles (<1 μm) that had previously incorporated radionuclides. In general, diffusive transport is characterised by a random movement of the atoms. Such movement causes a net flux that is proportional to the gradient of the radionuclide concentration, with direction from the higher concentration areas to the lower ones.

The diffusion of radionuclides through porous media is affected by both physical and chemical factors (Atkinson, 1983; Atkinson and Nickerson, 1988). The physical contribution is characterised by factors such as the tortuosity and porosity of the medium, both inversely related. In sediments, the small pore size and the tortuosity reduce the diffusion with regard to the diffusion that the radionuclide would have in free water. On the other hand, the chemical processes that affect diffusion can be very varied: precipitation/solubility and adsorption/desorption among others. Certain chemical processes contribute to the 'fixation' of the isotopes by the sediment, which causes a reduction of the diffusion. This 'fixation' phenomenon depends on variables associated to the sediment, such as the grain size, the mineral composition or the organic content (De Meijer et al., 1985; Elejalde et al., 1996); it can also be influenced by the physicochemical state of the radionuclides (Cundy and Croudace, 1995).

To carry out long-term predictions in environments polluted by radionuclides, it is important to know the temporal evolution of the migration of radionuclides in that medium in question. There are different works which study this temporal evolution, particularly for radiocaesium, both in real environments (Bunzl et al., 1995; Rosén et al., 1999; Isaksson et al., 2001) and in laboratory (Konoplev et al., 1996; Smith et al., 1999). Some works focus on the study of the aforementioned diffusion phenomenon, assessing the temporal dependency for different radionuclides and different types of porous media (Duursma and Eisma, 1973; Jannasch et al., 1988; Smith et al., 2000).

Due to the solubility of radium and potassium elements, the ^{226}Ra and ^{40}K radionuclides move relatively easily in the porous medium by diffusion. The normal levels of both radionuclides in the environment can be increased as a consequence of certain human activities. As an instance, the treatment and use of phosphate fertilizers can concentrate in very localised regions great amounts of ^{40}K and ^{226}Ra radioactivity, finding activities up to 4800 Bq/kg in phosphatic rocks compared with normal values of 25 Bq/kg (UNSCEAR, 1988). The objective of this work is to study the phenomenon of the diffusion of ^{226}Ra and ^{40}K in sedimentary columns, making the diffusive process of both radionuclides independent of the composition of the sedimentary medium, like its mineralogic, granulometric and organic composition. For this, some sedimentary columns have been reconstructed with the same type of sediment under overlying water rich in ^{226}Ra and ^{40}K. In this way, the diffusion of both radioisotopes has been studied during one year, which has allowed us to interpret the temporal evolution of the diffusion coefficient for each radionuclide in the sedimentary medium and to make a comparison between them.

2. Experimental method

The experimental system comprises 12 PVC cylindrical tubes with a 12.5 cm diameter and 1.60 m height, in which clay sediments were placed by re-packing. The sediments were extracted in the same geographical area with an illite content of 62%. Therefore, all the columns are granulometrically and mineralogically alike. The sediments were saturated with pure seawater by gradual flooding until a stable liquid layer of 30 cm was kept over the sedimentary interface. The height reached by the damp sediment column inside each tube was approximately 1 m. Five hundred Bequerel of ^{226}Ra were dumped to the overlying water of each core by pouring 400 g of phosphogypsum. It can be assumed that ^{226}Ra was dissolved in the overlying water (Bolívar et al., 2002). In order to make the experiment come close to the natural conditions, a vertical movement of the supernatant was caused by the immersion of fizzers in the liquid layer. In this way, the most relevant phenomenon of postdepositional mobility of ^{226}Ra at the sedimentary core is diffusion.

To study the diffusion of ^{40}K, an experiment similar to the previous one was carried out. However, in this case the pollutant used to dump ^{40}K to the overlying water of the sedimentary cores was potassium chloride (KCl), thus, it can be considered that ^{40}K will be in soluble form. Six thousand six hundred Bequerel of ^{40}K were dumped by pouring 400 g of KCl in each core.

The analysis of the columns started two months after dumping the pollutants. At a monthly interval one column after another was analysed, obtaining the mobility of both radionuclides in their respective columns along one year. Each sediment core was frozen at a temperature of −10 °C to ensure that the interstitial water remained at its corresponding level. Later, the sediment columns were sectioned into 1 cm thick slices. Each sample obtained was dried in an oven at 80 °C and its porosity was measured. Once the sample was dried, it was milled and sieved with a grain size under 250 μm to ensure its homogeneity for the spectrometry measurements, and its apparent density was determined by a gravimetric method. A proportional part of each sample was preserved in order to determine its organic carbon content, while the rest of the sample was placed into polyethylene containers, cylinder-shaped, of 4.6 cm diameter, keeping them sealed in the container during at least one month before carrying out the spectrometric measurements, in order to ensure the secular equilibrium between ^{226}Ra and ^{222}Rn.

The organic carbon content was determined by using the technique described by Gaudette et al. (1974), modified by El-Rayis (1985), which consists of the oxidation of the sediment sample with potassium dichromate in a strongly acid medium at 135 °C. The organic carbon content is calculated by an assessment of the excess of dichromate not consumed in the oxidation with ferrous ammonium sulphate. The percentage of organic carbon detected by this method depends on the type of sediment and is between the 80% and the 95%. With this technique, the elementary carbon is virtually not attacked and the carbonates do not display interference problems.

The gamma radiation measurements were performed for the radionuclides ^{40}K and ^{226}Ra with a coaxial detector HPGe of 90 cm^3 active volume, with a relative efficiency of 20% (with respect to a NaI(Tl) detector 3×3 in) and a resolution of 2 keV, for the photon of 1.33 keV of the ^{60}Co. This detector is sensitive within the energetic range of 50 keV–10 MeV. With the aim of minimising the external radiation, the measurement system is surrounded by a 10 cm thick lead shielding, which is at the same time covered with 1 mm of Cd and 1 mm of Cu. The measurement of the background radiation was performed periodically. The efficiency of the equipment, which is a function of the gamma radiation energy and of the sample height and density (Barrera et al., 1999; Ramos-Lerate et al., 1998), was calibrated. To obtain meaningful statistics in the measurements, the time of spectrometry for each sample was between 24 and 48 h. The measurement of the ^{40}K has been performed through the detection of its own gamma emission of the energy 1460.8 keV and intensity 10.7%. The measurement of the ^{226}Ra has been performed by the detection of the gamma emission of energy 609.3 keV and intensity 46%, emitted by ^{214}Bi, which is in secular equilibrium with ^{222}Rn and consequently with ^{226}Ra.

3. Diffusion model

To describe the transport equation in our experiment, it is considered that the flux of activity of the radionuclides in the sedimentary columns is due to the diffusive flux: $J_{\mathrm{Dif}} = -D(\partial a/\partial z)$, where a is the activity per unit volume, z is the depth and D ($\mathrm{L}^2\,\mathrm{T}^{-1}$) is the effective diffusion coefficient, which is reduced with respect to its value in free water due to physical hindrance to contaminant mobility, caused by factors like the porosity and tortuosity of the medium, and due to chemical processes that take place, such as sorption. Certain authors (e.g. Berner, 1980) propound an approximation for D using the water–sediment distribution coefficient, which is out of the objective of this study. There were no benthic organisms observed in the sedimentary cores, therefore, we do not consider the existence of vertical mixing by bioturbation.

For the transport equation we also have to take into account that our experimental system of sedimentary columns is closed, which means that after dumping the pollutants there are neither losses nor contributions of activity. Although a possible cause for a loss would be the radioactive decay, in this case we consider it negligible as a consequence of the elevated half-lives of ^{226}Ra and ^{40}K (1602 and 1.3×10^9 years, respectively).

Therefore, by taking into account only the diffusive flux, the evolution of the concentration of activity of the radionuclides in the sedimentary columns along the time t is obtained from the equation

$$\frac{\partial a}{\partial t} = \frac{\partial}{\partial z}\left(D\frac{\partial a}{\partial z}\right) \tag{1}$$

The effective diffusion coefficient D could depend on the depth and the time, however, in order to simplify the migration models, parameters like D tend to be considered constant (Bossew and Kirchner, 2004). To consider the spatial dependence implies to use more complex models which introduce problems of overparameterisation (Beck and Arnold, 1977). We have mentioned above that two fundamental factors in the diffusion of radionuclides in the sedimentary column are porosity and the content of organic matter. Fig. 1 shows that, for the penetration that takes place along a year, both parameters have a limited variation. Due to this, in our model we use an average effective diffusion coefficient D in depth, as it has already been done in

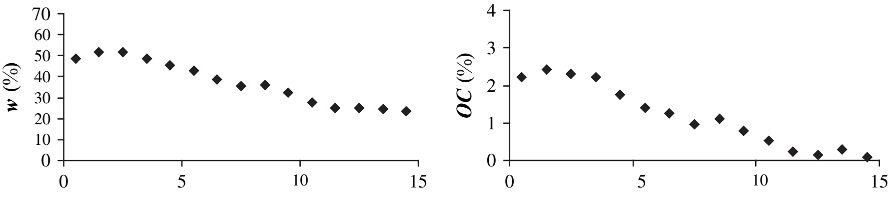

Fig. 1. Profiles at depth of porosity, w, and organic carbon content, OC, averaged for the experimental sediment columns.

others works (Barrera, 2002; Ligero et al., 2005a, 2005b). The possible temporal dependence of D will be deduced through the analysis of the successive sedimentary columns, which allows us to know the advance in depth of the radionuclides considered at monthly intervals. Therefore, considering D as constant Eq. (1) remains

$$\frac{\partial a(z,t)}{\partial t} = D\frac{\partial^2 a(z,t)}{\partial z^2} \tag{2}$$

To solve this differential equation it is necessary to establish the initial and boundary conditions. It is considered that $t = 0$ is the instant at which the pollutant is dumped in the sediment cores coming in contact with the water–sediment interface, and therefore, in order to establish the initial condition we take into account the mineralogical homogeneity of the sedimentary columns before the pollutant dumping, so the concentration of activity will be constant at $t = 0$: $a(z, t = 0) = cte$. For the boundary condition at $z \to \infty$ we take into account the limited penetration of the pollutant, in a way that the initial values will be reached: $a(z \to \infty, t) = a(z, t = 0) = cte$. The boundary condition at $z = 0$ is built taking into account the conservative nature of the experimental system designed, that is, at the sediment cores the activity concentration is to be preserved due to the absence of losses and contributions, therefore it is verified:

$$a(z = 0, t) = a_0 - \sum_z (a(z,t) - a(z, t = 0)) \tag{3}$$

with $a_0 = a(z = 0, t = 0)$ being the initial condition in the water–sediment interface. In Eq. (3) we suppose a null activity net balance in the exchange between the overlying water and the interface.

The differential equation (2) is solved using a numerical method. The finite differences method is used, expressing Eq. (2) in a discretised form (Haberman, 1987). The solution that is obtained depends on the parameters D and a_0. The determination of these parameters for each radioactive profile is carried out by minimising the quadratic difference, χ^2, between the theoretical and experimental data. Fig. 2 schematises the proposed model.

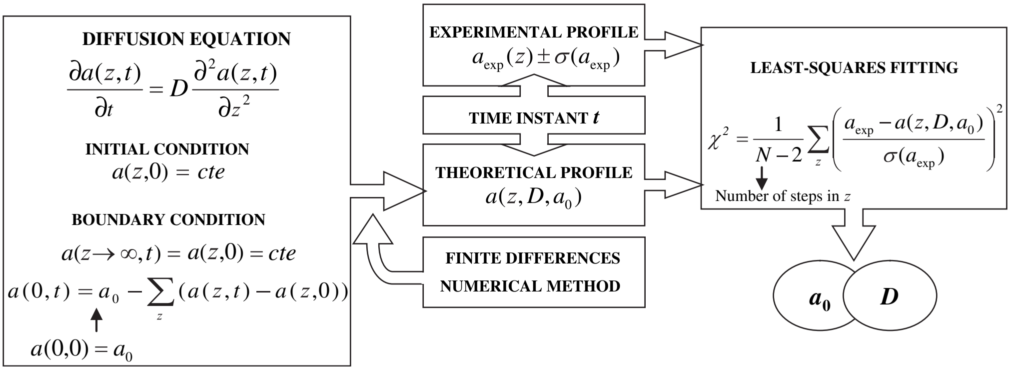

Fig. 2. Outline of the algorithm to determine the parameters for the diffusion model.

4. Results and discussion

Determining the in-depth profiles of porosity (*w*) and organic carbon content (OC) in the sedimentary columns, we appreciate that they display a similar behaviour in all the cases, which ratifies that all the columns reproduce the same sedimentary medium in which analysing the temporal evolution of the radionuclides is under study. The aforementioned Fig. 1 shows the averaged profiles of *w* and OC for all the sedimentary columns.

Applying the diffusion model to the experimental profiles of activity concentration of ^{226}Ra and ^{40}K obtained in each sedimentary column, we determine the effective diffusion coefficient of both radionuclides corresponding to each month of analysis. In order to establish the initial condition in the model, a non-polluted sediment sample is measured. In all the fittings carried out there is a good correlation between the theoretical function provided by the model and the experimental data. Fig. 3 shows, as an example, the fittings of some of the sedimentary columns measured.

Fig. 4 shows the representation of the obtained D values of ^{226}Ra and ^{40}K against the analysis time. A decreasing performance for both radionuclides can be appreciated within the first months of analysis, with a tendency to constant values from a certain value of t. Therefore, D parameter presents a degree of temporal dependence, so the 'assumption' of D constant along time carried out in Eq. (2) is erroneous both for ^{226}Ra and for ^{40}K, at least at the initial months of the experiment. This implies that in order to obtain a reliable effective diffusion coefficient, temporal dependence in the model has to be considered.

In this experiment the reduction of the diffusion coefficient along time can be justified, as a consequence of the clayey composition of the sediment used, since as deduced from other studies (Comans and Hockley, 1992; Chamard et al., 1993), ^{226}Ra and ^{40}K can experiment a progressive 'fixation' by clay minerals of the sediment. This produces an increase in sorption that causes a decrease of the activity diffusion rate along time (Smith and Comans, 1996).

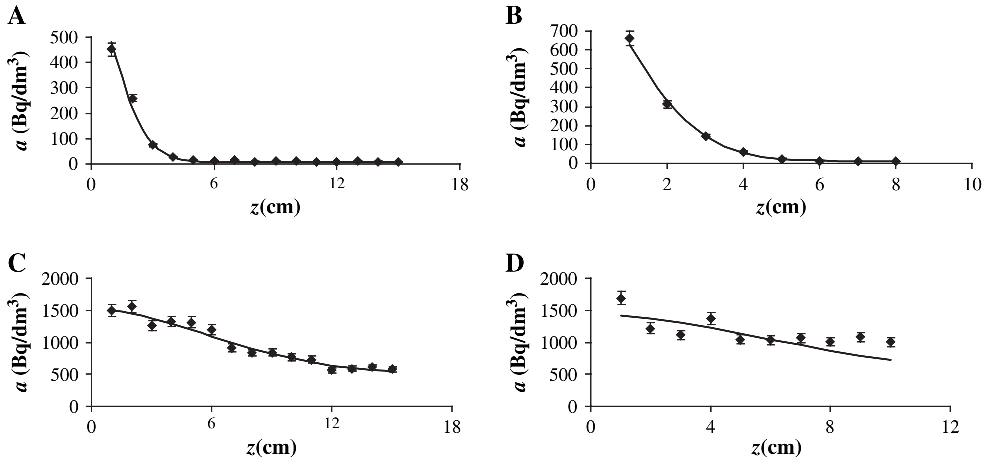

Fig. 3. Adjustments between the experimental profiles of activity concentration and those provided by the model for some of the cases under study: (A) month 2 for ^{226}Ra, (B) month 9 for ^{226}Ra, (C) month 2 for ^{40}K, and (D) month 6 for ^{40}K.

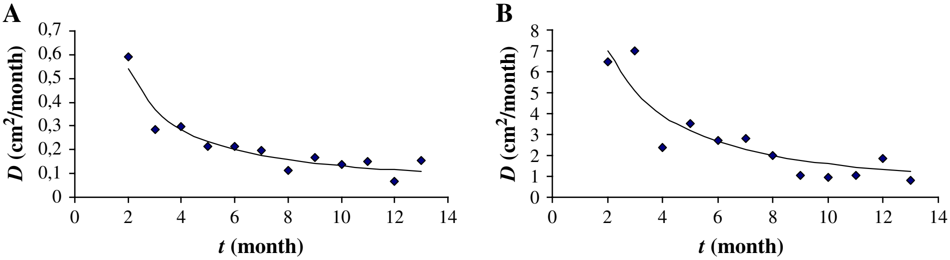

Fig. 4. Temporal evolution of the data obtained for the effective diffusion coefficient, D, for (A) ^{226}Ra and (B) ^{40}K. The adjustment line shows the fitting carried out by Eq. (5).

To study the temporal dependence of the effective diffusion coefficient with a greater approximation, Smith et al. (2000) propose the function

$$D'(t) = D'(0)[\eta + (1-\eta)\exp(-kt)] \tag{4}$$

where η ($0 < \eta < 1$) and k are constants. This function represents diffusion at an initial rate $D'(0)$, which decays, on a timescale $1/k$, to the constant value $\eta D'(0)$. It can be seen how the rate of decay k is approximately equal to the rate of 'fixation' to the sediment. In addition, Smith et al. (2000) prove that the effective diffusion coefficient D, obtained with a diffusion model similar to that developed in Section 3 (assuming D as constant), is related to the parameters in Eq. (4) by the expression

$$D = D'(0)\left[\frac{(1-\eta)}{kt}(1-\exp(-kt)) + \eta\right] \tag{5}$$

In this way, the values of $D'(0)$, η and k corresponding to our experiment can be obtained in a simple way by an adjustment of function (5) to the values of D obtained previously with the diffusion model. Fig. 4 shows the adjustments carried out. The corresponding adjustment coefficients are shown in Table 1. Approximating the timescale of 'fixation' to the sediment to $1/k$, the authors have obtained values of approximately 5 days for ^{226}Ra and 30 days for ^{40}K. According to this, after a time over 5 days after the beginning of the experiment, the diffusion of ^{226}Ra in the sediment takes place at a uniform rate, with little evidence of 'fixation', while the diffusion rate of ^{40}K takes more than 30 days to reach this uniformity. Fig. 5 shows the temporal evolution of the diffusion coefficient D' obtained for both radionuclides, normalised to the initial value $D'(0)$.

With the values obtained for $D'(t \to 0)$ we observe the retardation that the diffusion coefficient of ^{226}Ra and ^{40}K undergoes in the sedimentary medium with respect to its value in free water, D_0 (23 cm^2/month for ^{226}Ra and 50.8 cm^2/month for ^{40}K at 25 °C; Li and Gregory, 1974). If we calculate $D_0/D'(t \to 0)$ for both radionuclides, we obtain a retardation factor of the order of 3.7 for ^{226}Ra and 3.1 for ^{40}K, so that in the initial instants of diffusion we can

Table 1
Coefficients of the adjustment carried out for the effective diffusion coefficient D by Eq. (5), applied to the data obtained for ^{226}Ra and ^{40}K

	$D'(0)$ (cm^2/month)	η	k (1/month)	r
^{226}Ra	6.3	0.005	6.10	0.95
^{40}K	16.5	0	1.04	0.90

r is the correlation coefficient.

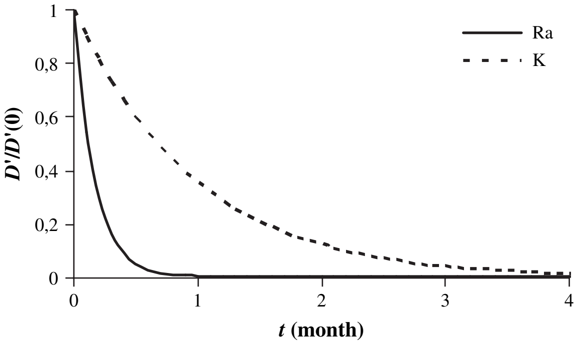

Fig. 5. Temporal evolution of the effective diffusion coefficient resulting from Eq. (4) for the diffusion of the ^{226}Ra and ^{40}K radionuclides in the experiment. The effective diffusion coefficient has been normalised to its value at the initial instant.

confirm that there is little evidence of 'fixation'. With this result we can corroborate that in the sedimentary columns built in the experiments the initial conditions (the physical and chemical factors that affect the retardation factor) are similar.

From a qualitative point of view, the results obtained for the diffusion of ^{226}Ra and ^{40}K in this experiment are in accordance with the result obtained by Smith et al. (2000) for the diffusion of ^{134}Cs in sediments with clayey composition dominated by illite, in which the initial value of the diffusion rate also experiments a reduction in a time interval of a few days in this case. This confirms the affinity of the cations Ra^{2+}, K^{+} and Cs^{+} for the sediments with a clayey composition.

5. Conclusions

The study that has been carried out shows that, during the first year after the contamination in a similar sedimentary medium by ^{226}Ra and ^{40}K, the effective diffusion coefficient of both radionuclides is time dependent, showing an initial decay behaviour, followed by a roughly constant tendency, which is in accordance with previous studies carried out (Smith et al., 2000). This decrease in time is justified by a slow but progressive 'fixation' of the radionuclides by clay minerals of the sediment. We observe that the effective diffusion coefficient of ^{226}Ra gains the independence in time more quickly than ^{40}K, and therefore, we conclude that the progressive 'fixation' of ^{40}K by clay minerals of the sediments is slower than in the case of ^{226}Ra.

It has been shown that the studied diffusion coefficients tend to constant values very close to zero (zero in the case of ^{40}K) in a time interval of days for ^{226}Ra and months for ^{40}K, what limits the capacity of penetration of these radionuclides in the sediments. This result could be very useful in actions to remedy a possible polluting episode caused by these radionuclides in sedimentary media with clayey composition, in which the dominant phenomenon of postdepositional mobility is molecular diffusion.

Acknowledgement

This study has been possible through the financing obtained from the Ministerio de Ciencia y Tecnología (Project REN2000-1261).

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