

Kinetic transfer coefficients for radionuclides in estuarine waters: Reference values from ^{133}Ba and effects of salinity and suspended load concentration

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In estuarine environments there are important spatial and temporal changes in both salt and suspended load concentrations. An experimental procedure have been developed to produce kinetic parameters being representative of the natural environment studied, and we have investigated the effect of salinity and suspended load concentration to the kinetics of the uptake. These results are encouraged by recent advances in environmental modelling concerning to radionuclide dispersion in aquatic natural systems and involving non-equilibrium processes. Experiments are carried out with unfiltered water samples from the Odiel estuary (Southwest of Spain), with ^{133}Ba tracer to illustrate experimental procedures.

Introduction

Estuarine areas are of relevant environmental interest. The reason being essentially that important industrial and human concentrations locate around them. Thus, significant wasting activities take place there and, consequently, an important environmental impact appears on both the neighbouring marine environment and the human population living in the vicinity.

Coastal areas and estuarine environments are usually dominated by tidal dynamics, and they are characterized by large spatial and temporal (tidal and seasonal) changes in physico-chemical parameters. The uptake of radionuclides by suspended particulate matter depends, among other factors, on the suspended load concentration, on its mineralogic and granulometric structure, and on the electrical conductivity.^{1,2} Consequently, the uptake of radionuclides will take place typically under non-equilibrium conditions, what is described in terms of kinetic transfer coefficients: a coefficient k_1 governs the transfer from water to the solid phase and a coefficient k_2 the inverse process.

Interest in measuring transfer coefficients has increased recently, since they are important parameters required to understand and reliably model the dispersion of non-conservative radionuclides in small coastal regions or estuaries.^{3,4} NYFFELER et al.⁵ obtained kinetic transfer coefficients for several elements from seawater onto particulate matter and surface sediments. BENES and CERNICK⁶ and BENES et al.⁷ studied the effects of changes in suspended solid concentration and temperature on the kinetic transfer coefficients of Co and Sr. Although other authors⁸⁻¹¹ have not obtained transfer coefficients, their studies have included the effect of sediment type, temperature, colloids, particle size and particle concentration on the uptake of radionuclides by sediments.

At present, there is not a suitable set of experimentally determined parameters to support environmental models. Even the available experimental results are often apparently surprising or contradictory amongst themselves. Thus, different experiments have shown an increase, a decrease, a passage through a maximum or a null effect on uptake kinetics as a function of increasing suspended load.^{7,12} Some theories on surface-electrolyte interactions state only for equilibrium conditions, as the Couy-Chapman diffuse double layer and the Stern double layer theories.^{13,14} Consequently they cannot properly account for the effect of salinity concentration in the kinetics of the uptake of radionuclides by suspended particulate matter.

For environmental modelling studies, concerning the dispersion of radionuclides in tidal estuarine waters, it is necessary to investigate:

(1) The kinetics transfer coefficients for each studied radionuclide, corresponding to the most representative environmental conditions.

(2) In estuarine waters there are important spatial and temporal (tidal and seasonal) changes in salt and suspended load concentrations; thus, the effect of such physical parameters in these kinetic coefficients must be studied.

In References 1 and 2 a basic microscopic theory of the uptake kinetics of dissolved radionuclides by suspended particulate matter is presented. The most relevant contribution is the assumption that exchange of radionuclides occurs in a certain surface layer in particles, with a non zero thickness. Then, the governing time evolution equation for a dissolved radionuclide, whose concentration in water is a_w (in Bq/l), is written as:

$$\frac{da_w}{dt} = -k_1 a_w + k_2 a_{sp} \quad (1)$$

where a_{sp} is the specific activity, in Bq/kg, of the studied

radionuclide in the surface layer of particles, where uptake takes place. Thus, the k_1 and k_2 coefficients are written as:

$$\begin{aligned} k_1 &= f_c S_p v_m P_r \\ k_2 &= \alpha m_{sl} \times 10^{-6} \end{aligned} \quad (2)$$

where S_p is the free external surface of particles, in m^2/m^3 , f_c is a correction factor accounting for the Stern double layer effect and for the proportionality factor to v_m . This last is the averaged module of the velocity, that depends on temperature and on ion mass. Finally, P_r is a probability of adsorption. In k_2 expression, α is the probability of desorption, and m_{sl} the mass of the external surface layer of particles (in mg/l).

For typical aquarium experiments Eq. (1) can be transformed by multiplying by the total water volume. Thus a_w converts into A_w , in Bq, and $m_{sl} \times 10^{-6} a_{sp}$ converts into A_s , the total activity bound in particles, also in Bq:

$$\frac{dA_w}{dt} = -k_1 A_w + A_s \quad (3)$$

In estuarine waters, the uptake of dissolved radionuclides by suspended particulate matter can be regarded as a Langmuir adsorption with two components: the first one being the naturally occurring ions in solution, which provokes a partial saturation of free active areas over the surface layer of particles, and the second one being the dissolved radionuclide that rarely provokes any further saturation phenomena. Thus, from ABRIL:^{1,2}

$$P_r = P_{r_o} \frac{a_{sp}}{a_{sp} + a_o} \quad (4)$$

where subindex "o" refers to the ideal situation of zero concentration of ions in solution.

From this author,^{1,2} experimental procedures to determine kinetic transfer coefficients useful for environmental modelling, have to be carefully designed. Thus, the use of higher suspended load concentrations than in natural systems, leads to kinetic coefficients being also higher than environmental ones. The use of bottom sediments cannot characterize the behavior of naturally occurring suspended loads due to their different particle size spectra and their different physico-chemical properties. Changes in the concentration of naturally occurring ions in solution also affect the kinetic transfer coefficients, as well as the use of relatively large amounts of tracer. The use of alternative radioactive tracers (as assuming that ^{133}Ba is representative of the environmental behavior of ^{226}Ra) is not advised, since they have different ion mass and ion size.

In this work, we present a basic experimental procedure (experiment 1) able to determine kinetic transfer coefficients representative of environmental conditions. A modified procedure (experiment 2) is then used to investigate the effect of both suspended load and salt concentrations, in the uptake of dissolved radionuclides by suspended particulate matter. Especially, the use of Eq. (4) is discussed in some detail. Finally, conclusions are presented.

Unfiltered water of the Odiel river has been used, an estuarine system at the south-west of Spain where a phosphate fertilizer processing factory releases part of its wastes. Although our initial interest was the environmental behavior of the ^{226}Ra , this isotope cannot be used as radioactive tracer in our experiments because it is already present in natural samples. Thus, we used ^{133}Ba as the most suitable alternative.^{3,4,16} The possibility of using γ -spectrometry to measure this isotope supposes an additional advantage, if we take into account the large amount of required measurements, and the complexity of the experimental procedure. Consequently, results have to be considered as representative of ^{133}Ba and, but only approximately, of ^{226}Ra behavior. Nevertheless, experimental procedure 1 and 2 have a general character. Thus conclusions concerning the effect of salinity and the suspended load concentration are of the same generality.

Experiment 1

Experimental procedure: A 50 l Odiel river water sample was collected in summer 1994. The water was introduced into two plastic bottles and stored in dark for a very short time (few days). The pH, electrical conductivity and the suspended matter concentration (by nephelometry) in the water were measured in the laboratory.

A series of 5 experiments was carried out. Each experiment was performed with 5 l of unfiltered water. The suspended matter concentration was measured again before each experiment for control purposes. The water was spiked with some 400 Bq of $^{133}\text{Ba}^{2+}$. The water was stirred during the experiment using a magnetic stirrer, and room temperature was maintained constant during the experiment. After the relevant time period had elapsed, the filtration of water started. Nucleopore filters of 0.45 μm pore size were used. Filtered water was measured to verify all the solid phase had been recovered on the filter. Conversely, 1 l of filtered water was introduced in a Marinelli beaker. ^{133}Ba activity was measured by gamma-spectrometry (in a Canberra HPGe) in both the suspended matter and water. The last measurement is a test to check that no activity is lost during the experiment (by interaction with the glass walls) nor the filtration process. Subsequently, the filters

were found to retain negligible activity, by filtering distilled water traced with $^{133}\text{Ba}^{2+}$ and measuring the filter activity.

Table 1. Elapsed time, ^{133}Ba activity incorporated into suspended matter and suspended matter concentration, m_s , for experiment 1. Errors (1σ) of the activities come from counting statistics. Error in m_s is of a 3% in all the cases. A_s^* is the activity in suspended matter renormalized at $m_s = 30$ ppm (see text)

Time, hour	A_s , Bq	m_s , ppm	A_s^* , Bq
14.1	4.54 ± 0.12	40.0	3.42 ± 0.09
41.9	5.30 ± 0.08	36.0	4.42 ± 0.07
89.8	6.92 ± 0.16	36.0	5.78 ± 0.13
141.2	3.64 ± 0.12	22.3	4.88 ± 0.16
190.0	3.00 ± 0.13	24.6	6.0 ± 0.3

Efficiency calibrations were carried out by preparing samples of known ^{133}Ba activity and the same geometry as the real samples to be measured. In the case of filtered water, a known activity was dissolved into a 1 l Marinelli beaker filled with distilled water. For the filters, a known activity of ^{133}Ba was added to 0.5 l of distilled water. Then Ba was precipitated with SO_4H_2 , by the same method used to extract ^{226}Ra from a water sample (see for instance References 15 and 16). After filtration, a ^{133}Ba sample with the same geometry as the filters measured was obtained. The experiment is repeated for different times to obtain the time evolution of A_w and A_s shown in Fig. 1. For all experiments, temperature was constant, 20°C , and electrical conductivity was 39.1 ± 0.6 mS. The averaged suspended load concentration was around 30 ppm, but significant changes were found from one subsample to another. This problem arises when handling natural samples. Although, we tried to homogenize the water sample before taking each subsample, by agitation of the plastic beaker where water was stored, we got suspended load concentrations ranging from 22.3 to 40.0 ppm. Consequently, some mathematical treatment of data has been necessary to interpret results, as discussed in the following section.

Results and discussion: In Table 1 we present the results for experiment 1.

The analytical solution of Eq. (3) gives A_s at time t as:

$$A_s(t) = \frac{k_1}{k_1 + q} A_w(0) (1 - e^{-(k_1 + q)t}) \quad (5)$$

where $A_w(0)$ is the activity in the dissolved phase at $t=0$. Since in our experiment 1 the natural equilibrium between salt ions in solution and particulate matter is not perturbed, and since the amount of tracer is small, and other physico-chemical changes do not take place, P_r , the probability of capture, will be almost constant. Thus, k_1 will be proportional to m_s since the specific free surface, $S_{sp} = S_p/m_s$, does not depend on m_s :

$$k_1 = f_c v_m P_r S_{sp} m_s = \chi_1 m_s \quad (6)$$

This will allow us to renormalize the measured A_s at different m_s values and, consequently, corresponding to different solutions of Eq. (5) to the averaged m_s value (30 ppm). The renormalization is complex and will be briefly described below.

The variation of A_s with m_s at a given time is:

$$\frac{A_s}{m_s} = \frac{k_1}{k_1 + q} \frac{A_w(0)}{m_s} (1 - e^{-(k_1 + q)t}) + \frac{A_w(0)}{m_s} e^{-(k_1 + q)t} \quad (7)$$

From the crude A_s values in Table 1 an estimation of k_1 and q is performed by a standard fitting procedure to Eq. (5). By applying the renormalization of Eq. (7) we obtain, by integration, new values of A_s . The procedure is repeated till mathematical convergency to the solution values, A_s^* , is reached. These are presented in Table 1 with the corresponding propagation errors.

These A_s^* values are used in the final fit to Eq. (5) (see Fig. 1) in order to get the following parameter values:

$$k_1 = (8.9 \pm 1.0) \cdot 10^{-7} \text{ s}^{-1} \quad (8)$$

$$q = (1.35 \pm 0.17) \cdot 10^{-5} \text{ s}^{-1}$$

where errors in the fitted coefficients have been evaluated by BENES and CERNIK.⁷

The transfer coefficients allow us to evaluate the equilibrium k_d . Effectively, from Eq. (3), at equilibrium $dA_w/dt=0$, and $k_1 e^{A_w} = q e^{A_s}$; and converting activities into specific activities we get

$$k_d = \frac{1}{m_s^*} \frac{k_1}{q} \quad (9)$$

where m_s^* is 30 ppm. Substituting values we obtain $k_d = 2.2 \pm 0.8$ l/g, in agreement with other published values¹⁷ of k_d for ^{226}Ra .

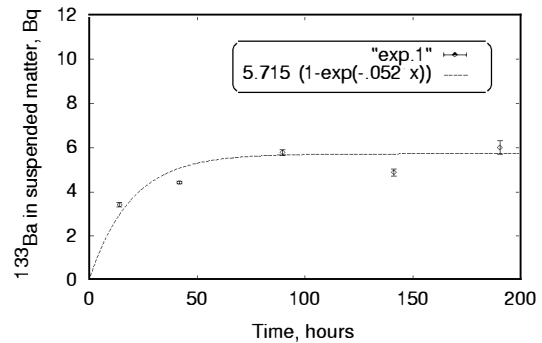


Fig. 1. Time evolution of corrected A_s^* values. Dotted line corresponds to the best fit to the analytical solution

Table 2. Characteristics of the four solutions used in experiment 2

Solution	m_s , ppm	Conductivity, mS
(1)	110	68.4
(2)	445	51.8
(3)	445	93.9
(4)	445	32.2

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Experiment 2

This second experiment has been designed to investigate the effect of salinity in the uptake kinetics. The effect of suspended load concentration will be also checked, although it has been well established by other authors.^{1,2}

Experimental procedure: Some 200 l of estuarine water was collected, in summer 1995, at two different locations in the Odiel estuary (Spain). Location 1 was close to location sampling of experiment 1, and location 2 was in the Tinto river, which connects with Odiel river, but having higher suspended loads concentrations. By a decantation process the suspended matter was concentrated, producing some 10 liters of suspension with m_s values up to 110 and 445 ppm, for the first and second location samples, respectively. This decantation process preserves the equilibrium with natural ions (this is, there are not changes in electrical conductivity). Four series of experiments were performed by using these concentrate: (1) A subsample from concentrate of 110 ppm, (2) a subsample from concentrate of 445 ppm, (3) an additional decantation of a subsample of the 445 ppm concentrate was carried out. A part (a half, approximately) of the supernatant water was removed and replaced by a solution of higher conductivity. This solution was prepared from filtered estuarine water reducing its volume by evaporation (till a 30%, approximately). Final solution had 445 ppm and an electrical conductivity of 93.9 mS, and (4) an additional decantation of a subsample of the 445 ppm concentrate was carried out. A part (a half, approximately) of the supernatant water was removed and replaced by distilled water. In this way resulting m_s is 445 ppm by electrical conductivity is reduced (from 51.8 to 32.2 mS).

By this means we got a range of electrical conductivity from 32.2 to 68.4 mS (see Table 2). In Table 2, solution 1 corresponds to site 1, and solutions 2 to 4 to site 2.

The large amount of suspended loads allows us to have a faster kinetics (and, consequently, shorter elapsed times) and the use of centrifugation instead of filtration to separate water and suspended matter. Thus, each series of experiments started with a 2-litre solution, spiked with a known activity of $^{133}\text{Ba}^{2+}$ and continuously stirred. At every selected time, a subsample of 50 ml was obtained and centrifugated for 15 minutes, after which supernatant was separated and

HNO₃. Both samples, particles and supernatant water, were measured by gamma-spectrometry after preparing the corresponding calibration samples. This procedure has been checked in order to ensure that particle separation by centrifugation was complete.

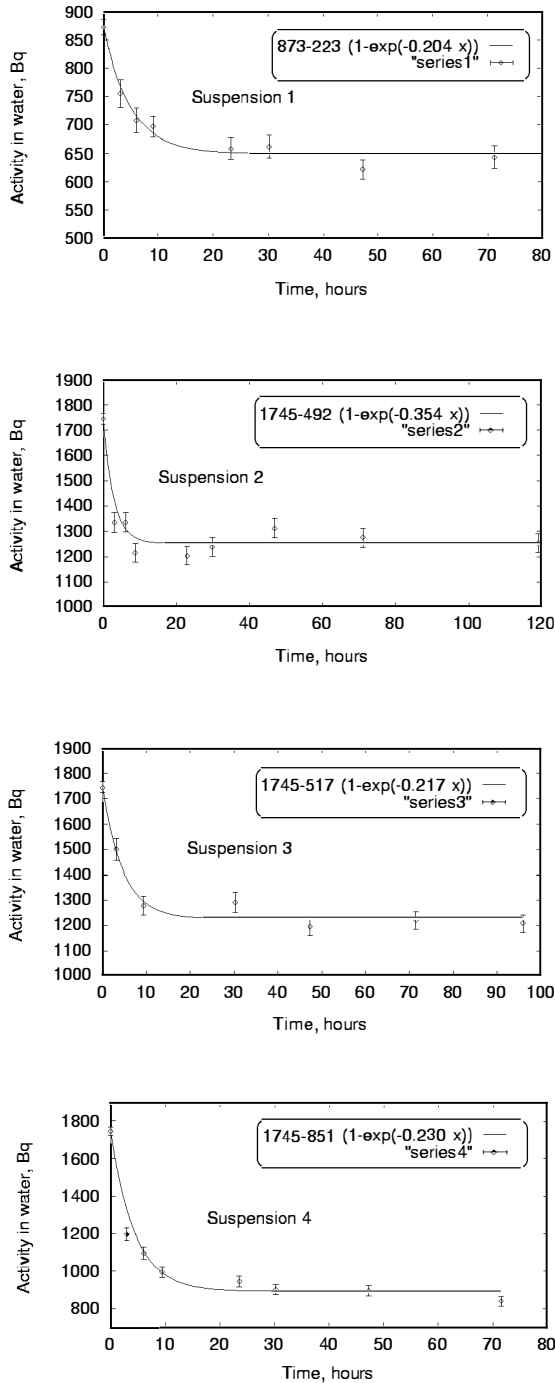


Fig. 2. Time evolution of the measured A_w values for the four series of experiment 2. Continuous lines correspond to the best fit to the analytical solution

Table 3. Calculated values of the transfer and distribution coefficients

Series	k_1, s^{-1}	k_2, s^{-1}	$k_d, l/kg$
(1)	$(1.45 \pm 0.15) \cdot 10^{-5}$	$(4.2 \pm 0.4) \cdot 10^{-5}$	3180 ± 470
(2)	$(2.8 \pm 0.8) \cdot 10^{-5}$	$(7.1 \pm 2.0) \cdot 10^{-5}$	880 ± 120
(3)	$(1.8 \pm 0.2) \cdot 10^{-5}$	$(4.2 \pm 0.4) \cdot 10^{-5}$	950 ± 120
(4)	$(3.1 \pm 0.2) \cdot 10^{-5}$	$(3.3 \pm 0.2) \cdot 10^{-5}$	2140 ± 160

This was carried out by means of nephelometric measurements prior and after centrifugation in several test samples. Temperature is controlled, and kept constant (20 °C) during the whole duration of the experiment.

Results: For each series of experiments, we applied the same mathematical treatment presented for experiment 1. Thus, in Fig. 2 we plot the fitted curves from Eq. (5) to the measured $A_w(t)$ activities ($A_w(t) = A_w(0) - A_s(t)$). In this new situation the continuous stirring ensure a reasonable homogeneity in m_s in all subsamples, thus no renormalization has been applied. The so obtained k_1 and η coefficients, and the corresponding k_d values are shown in Table 3, with their corresponding errors.

It can be seen, from Table 3, that k_d 's are in the range of variation that can be found in literature,¹⁷ which is $0.5 \cdot 10^3$ to $50 \cdot 10^3$ l/kg with a mean value of $5 \cdot 10^3$ l/kg. The transfer coefficient η is of the same order of magnitude than that obtained by NYFFELER et al.⁵ for Ba. k_1 is different due to the different conditions (temperature, salinity, suspended matter concentration) in which the experiments by NYFFELER et al.⁵ were performed. Thus, it seems that k_1 is the relevant parameter controlling the geochemical behavior of elements.

Analysis of results

k_1 coefficient

The dependence of k_1 with salinity can be studied from series 2 to 4, since no other parameter than electrical conductivity changes within these series. Thus, for a small amount of tracer (this is, no further saturation phenomena), at equilibrium, and from Eqs (2) and (4), k_1 can be written as:

$$k_1 = \delta k_1 (1 - \delta) \quad (10)$$

where δ is the fraction of occupied sites (active areas for ion uptake), and is related with the electrical conductivity (this is, with ion concentration in water), c , through the Langmuir isotherm:

$$\frac{\delta}{1 - \delta} = \frac{c/c_0}{1 + c/c_0} \quad (11)$$

where c_0 is a parameter, the ratio of the coefficients

for the reverse and forward reactions, and corresponding to the ion concentration in water for which $\delta = 1/2$ (see Reference 14).

In Fig. 3 we plot k_1 versus electrical conductivity for series 2, 3 and 4. The curve-line corresponds to a solution of Eq. (10). We note that changes in salinity could affect the state of aggregation of the floc fraction of suspended particles (and therefore the reactive surface area), what surely affects δ . Nevertheless, the continuous stirring maintains a mechanically induced level of defloculation which apparently does not change with electrical conductivity in a significant way, as it can be guessed from Fig. 3. This mechanically induced defloculation represents the most frequent situation in estuarine systems, due to tidal water currents. Nevertheless, with only 3 points and with large error bars definitive results cannot be derived. In the curve-line, the value of ${}_o k_1$ is of $5.34 \cdot 10^{-5} \text{ s}^{-1}$, and corresponds to k_1 if all active sites were unoccupied. The 50% of saturation takes place when the electrical conductivity (related with the concentration of ions in solution) is of some 50.5 mS. In this situation ${}_o k_1$ is also ${}_o k_1/2$.

If we introduce now the effect of suspended load concentration, under the condition that increase/decrease of m_s is always in equilibrium with the same salinity value, and that particle size spectrum does not change (see References 1 and 2 for the effect of particle size in the uptake kinetics), then ${}_o k_1$ for location 2 (Tinto River) will be:

$${}_o k_1(m_s, c) = \frac{{}_o k_1 c}{c_o + c} \frac{m_s}{m_{s, \text{exp}}} \quad (12)$$

with the parameter values stated above. This expression gives us ${}_o k_1$ as a function of the suspended load concentration, m_s and the electrical conductivity, c .

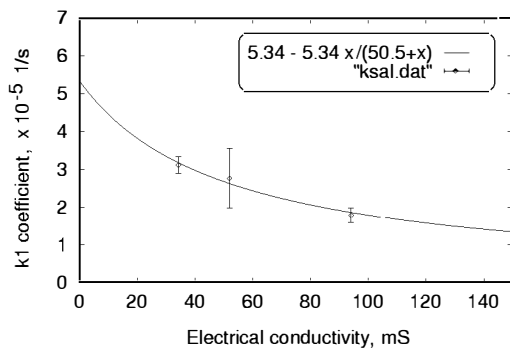


Fig. 3. Dependence of ${}_o k_1$ coefficient with electrical conductivity. Experimental data corresponds to series 2 to 4, the curve-line corresponds to the theoretical fit

In this expression appears the ratio ${}_o k_1/m_{s, \text{exp}}$, where $m_{s, \text{exp}}$ is the value of m_s corresponding to the experimental conditions in which ${}_o k_1$ was measured (445 ppm in our situation).

This formula has to be corrected by the factor $S_{\text{sp}}/S_{\text{sp, exp}}$ that accounts for the change in specific surface accompanying a change in m_s . This is the usual situation in estuarine waters, where the amount of suspended matter depends on water velocity, and larger size particles can be resuspended from the sediment during high current situations.^{3,4,18}

This formula does not account, in principle, for ${}_o k_1$ from series 1 and 2, since there are, at least, three parameters changing from place to place: m_s , S_{sp} and the maximum exchange capacity, here measured through the c_o parameter. Thus, if we apply Eq. (12) to the renormalized ${}_o k_1$ value from series 2 to 4 to the same conductivity as in experiment 1, and we make the correction by the same factor $(1-\delta)$ in series 1, we get the ${}_o k_1$ versus m_s plot shown in Fig. 4. The plot fits approximately a linear relationship, and discrepancies from this trend have to be interpreted in terms of differences in S_{sp} and c_o values. Figure 4 can also be interpreted as reflecting a good level of confidence that is achieved when extending the application of Eq. (12) to other locations within the same estuary. This supposes an interesting simplification for environmental models. Thus it seems to be enough to calibrate Eq. (12) at a few sample sites defining subzones or domains within the studied estuarine environment. On the other hand this supposes another interesting finding, since faster experimental procedures (as experiment 2) can be used instead of more orthodox ones (as experiment 1). Although this result is very interesting, we recognize that it is actually tentative, and some further experimental confirmation is required.

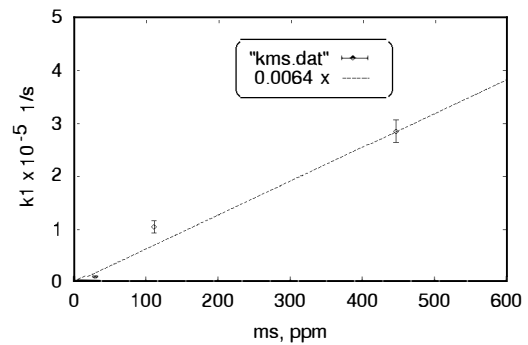


Fig. 4. Dependence of ${}_o k_1$ coefficient with suspended matter concentration. Experimental data have been corrected by electrical conductivity effect (see text)

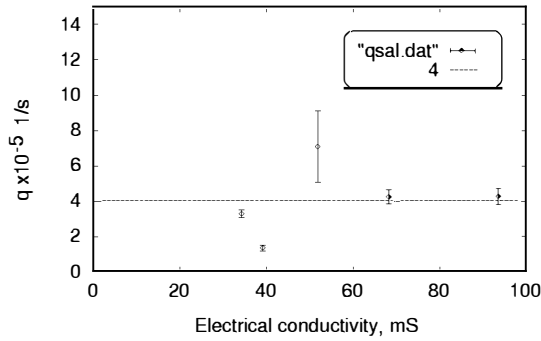


Fig. 5. Dependence of q coefficient with electrical conductivity. Experimental data corresponds to series from experiment 1 and 2, the line corresponds to the averaged value

q coefficient

This coefficient is a probability of desorption. It can be expanded as the summation of probabilities of desorption from sites with different energies or by different processes. The way in which q can be affected by salinity is rather complex. The ionic strength of the solutions changes the electrical potential at the surface of particles, which affects the desorption of the physico-adsorbed ions, but with a no clear effect over the chemico-adsorbed ones. On the other hand, the concentration of ion in solution affects the surface charge in the colloidal fraction and the state of aggregation of the floc fraction,¹⁸ but the surface charge in mineral particles depends on several contributions (see Reference 19). Also, changes in particle size spectra and, of course, mineral composition and nature and size of overcoatings, will produce some differences in q values.

Results from experiments 1 and 2 (see Table 2) show q values of the same magnitude. Data do not show any trend of variation neither with m_s nor with conductivity. This is clearly shown in Fig. 5, where q values are around an averaged value of $(4 \pm 2) \cdot 10^{-5} \text{ s}^{-1}$.

k_d values

From ABRIL,^{1,2} k_d value does not depend on m_s if initial equilibrium with natural salt ions is preserved, and if S_{sp} does not change with m_s . Thus, the k_d values from experiment 1 and from series 4 are almost identical, even being water samples collected at different sites and times.

k_d value will depend on electrical conductivity through the dependence of k_1 above discussed, while q parameter is expected to remain constant. Thus, in Fig. 6

we plot the measured k_d values for series 2 to 4 versus electrical conductivity. A fit is obtained by using the following equation:

$$k_d = \frac{e k_1}{q m_s \times 10^{-6}} = \frac{E}{G} \frac{k_1 c}{c_o + c} \frac{1}{m_s \times 10^{-6}} \quad (13)$$

In Fig. 6, the curve line has been obtained from this equation by using the already known parameter values and the averaged value for m_s . The fit is essentially the same as for k_1 . Apparently larger discrepancies appear because we used the averaged value of q in the theoretical fit while each particular q value is used for experimental data.

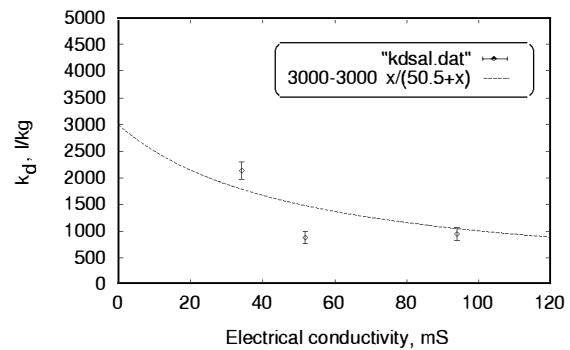


Fig. 6. Dependence of k_d coefficient with electrical conductivity. Experimental data corresponds to series 2 to 4, the line corresponds to the theoretical value

Conclusions

An experimental procedure is presented to obtain kinetic transfer coefficients for a given radionuclide that are useful for environmental modelling (this is, corresponding to natural environmental conditions). This has been applied to ¹³³Ba in estuarine waters from the Odiel river (Spain).

A second experimental procedure is presented to investigate, with shorter elapsed times, the effect of electrical conductivity in both, the kinetics parameters and the equilibrium distribution coefficient.

The obtained pattern of variation of k_1 with electrical conductivity is in reasonable agreement with previous theoretical considerations.

A general formula is proposed to give the k_1 dependence with electrical conductivity and suspended matter concentration, which are two of the governing parameters in estuarine waters. The use of this formula can be extended to "natural environmental conditions".

k_d does not depend on suspended matter but on electrical conductivity, following the same trend that k_1 (if some conditions are preserved).

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