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Modeling sensitivity of biodiffusion coefficient to seasonal bioturbation

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ABSTRACT

Biodiffusion coefficient is the predominant parameter used to constrain biological activity in marine sediments. Bioturbation characterization is important because of the dominant role it plays on the flux determination through the sediment-water interface. Biological mixing is quantified through models of radionuclides diagenesis by both a biodiffusion coefficient (Db) and a mixed depth (L) under the basic steady-state assumption. Based on a new global compilation of radionuclide data in marine sediments and on previously published modeling results, we show that short-live radionuclides are perfectly devoted to quantify biological mixing for sediments associated with $\lambda L^2/Db$ lower than 125, λ representing the decay constant of the radionuclide. 75 % of the ^{234}Th-derived Db, and 79 % of the ^{7}Be-derived Db are concerned by this result. However, as transient regimes prevail within marine sediments, especially at a seasonal time scale and within the coastal and shelf environment, it is necessary to model their impacts on Db calculations. A transient model of radionuclide decay and transport is therefore used to perform extensive sensitivity tests of Db calculations in respect to seasonal mixing. Numerical tests of seasonal sensitivity indicate that ^{234}Th and ^{7}Be are the most sensitive tracers to seasonal biological mixing: the steady-state assumption remains valid and applicable for most of natural marine environments. However, systematic tests reveal that incorrect seasonal sensitivity of ^{234}Th is detected for marine environments with $\lambda L^2/Db$ lower than 10 and greater than 1000. In these cases, the apparent seasonal variations of the biological activity need to be corrected. The main parameter in selecting the appropriate radionuclide for field analyses is the dimensionless pulse, which defines the relative importance of decay time scale relative to the seasonal time scale. This pulse controls the relative extension of the domain of satisfactory sensitivity. Consequently, long-lived radionuclides (^{210}Pb and ^{228}Th) are not appropriate for predicting seasonal mixing, except for specific environments which display an unexpected sensitivity to seasonal mixing. These marine environments are characterized by a moderate biological mixing and a deep mixed-layer.

1. Introduction

Bioturbation is defined by the mixing of surface sediments by organism activity, which can introduce significant modifications to the physical, biological and chemical properties of sediment. Bioturbation can redistribute particles within the surface layer of marine

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sediments, dispersing the stratigraphic record (Ruddiman and Glover, 1972; Guinasso and Schink, 1975; Wheatcroft, 1990) and influence the spatial extent of the different reactions of organic matter mineralization. Through the redistributing reactive particles, bioturbation can significantly affect the gradients of pore-water constituents and consequently their fluxes across the sediment-water interface (e.g. Schink and Guinasso, 1977). Bioturbation can accelerate pollutant burial or, inversely, remobilization of buried contaminants (e.g. Bosworth and Thibodeaux, 1990). In the absence of physical sediment reworking, i.e. currents, bioturbation constitutes the major process affecting sedimentary particles. Therefore, quantification of the biological mixing rates is fundamental. The most widely used mathematical method to evaluate bioturbation is based on the analogy of eddy diffusion for which the biological mixing is characterized by both a biodiffusion coefficient (Db) and a mixed depth (L) (Goldberg and Koide, 1962; Guinasso and Schink, 1975; Berner, 1980; Aller, 1982; Boudreau, 1997). Extensive developments concerning the conditions of this biodiffusive approach were exposed by Meysman *et al.* (2003) who concluded that biodiffusion has proven to be a valuable empirical model for sediment mixing. Recently, Reed *et al.* (2006) suggested that short-lived radionuclides are not suitable to be used with the biodiffusive model. This analysis will be discussed later. The biodiffusion parameters are commonly estimated by modeling depth profiles of naturally occurring or anthropogenic radionuclides added to the sediment in association with fine-grained particles. Naturally occurring radionuclides, such as ^{234}Th, ^{7}Be, ^{228}Th or ^{210}Pb are commonly used to calculate the biodiffusion coefficient. Their half-life extends from 24.1 day (^{234}Th) to 22.3 year (^{210}Pb). The Db calculation from natural radionuclides-depth profiles is based on the steady-state approximation that stipulates the lack of time variations of both biological activity and radionuclide flux through the sediment-water interface. However, detailed examinations of natural sedimentary environments exhibit strong temporal variations related to a number of variables, i.e.(i) Seasonal biological activity, (ii) Meteorological conditions and (iii) Transient flux of matter through the sediment-water interface (e.g. Martin and Bender, 1988; Sun *et al.*, 1991; Soetaert *et al.*, 1996a; Gerino *et al.*, 1998; Schmidt *et al.*, 2002, 2007). Many authors have extensively discussed conditions of steady-state application (see review in Boudreau, 1997, p. 191) and concluded that transient-state regimes are the rule in the marine sediments instead of the exception. Moreover, the steady-state approximation remains valid, firstly, when fluctuations imposed on the system have a mean period significantly shorter than the other process-time scales and, secondly, when fluctuations exhibit longer periodicity than the other process-time scales. In these cases, a sedimentary system can be described by a series of steady-states (Boudreau, 1997). Transient regime can exert significant perturbations on the Db calculation process. For example, using a diagenetic model of short-lived radionuclides, Lecroart *et al.* (2007) have shown that the transient regime imposed by variations of radionuclide flux to the sediment-water interface can generate significant error (up to a factor of two) upon the calculation of the Db coefficient.

Due to the few studies that deal with bioturbation variations in marine environments

Table 1. Commonly used radioisotope tracers of biodiffusion.

Db tracer	Half-live	Decay constant λ (y^{-1})	Pulse γ
^{234}Th	24.1d	10.51	0.095
^{7}Be	53.3d	4.75	0.211
^{228}Th	1.9y	0.365	2.74
^{210}Pb	22.3y	0.0311	31.74

(e.g. Gerino *et al.*, 1998; Schmidt *et al.*, 2007), the literature has not explored, up to now, the sensitivity of the biodiffusion coefficient to seasonal biological activity. Therefore, a modeling approach is clearly required to solve this problem. We used a non-steady-state and dimensionless model of radionuclide diagenesis to test the sensitivity of the Db calculation scheme to seasonal variations of biological activity. This model simulates transient tracer profiles affected by seasonal bioturbation. This study precisely characterizes how the traditional method, used to compute Db, is able to follow the transient biological activity. This work is particularly relevant for the natural radionuclides traditionally used to constrained biodiffusion coefficient (Table 1), which are divided into two classes: the short-lived radionuclides (^{224}Th and ^{7}Be) and the long-lived ones (^{228}Th and ^{210}Pb).

2. Model equations and numerical solution

a. Model equation

Bioturbation is classically described as an eddy-diffusive transport process occurring within a surface mixed layer (Goldberg and Koide, 1962). Biodiffusion is a local transport associated with spatially random and small scale sediment displacements between each mixing event (Boudreau, 1986a; Meysman *et al.*, 2003). Nonlocal mixing, which implies exchange of material between non-adjacent points, exists in natural environments (Boudreau, 1986b). Biodiffusion and non-local mixing are generally treated separately. This study is only concerned by radionuclides sensitive to seasonal biodiffusive mixing events, but does not assess their sensitivity to nonlocal bioturbation. Consequently, complementary field investigations and modeling studies are needed to address this additional problem.

Surface sediments are characterized in one dimension by a mixed layer of thickness L with a constant porosity. The classical advection-diffusion model equation adapted for radionuclides is

$$\frac{\partial A}{\partial t} = \mathrm{Db}(t)\,\frac{\partial^2 A}{\partial z^2} - \mathrm{w}\,\frac{\partial A}{\partial z} - \lambda A \tag{1}$$

where A is the activity of the radionuclide within the mixed layer, z the depth in the mixed layer relative to the sediment-water interface, Db the biodiffusion coefficient , w the burial

rate, and λ the decay rate of considered radionuclide (Table 1). By assuming steady-state, Eq. (1) can be solved and Db computed

$$\mathrm{Db} = \frac{\lambda z^2}{(\ln(A_0/A_z))^2} - \frac{\mathrm{w}z}{\ln(A_0/A_z)} \tag{2}$$

Db is then characterized from depth profiles of radionuclide activity measured in the sediment cores. In field investigations, steady-state approximation is often validated by the observation of a significant correlation between the logarithm of the radioisotope activity and the depth. This analysis is not always sufficient because transient profiles can easily mimic steady-state ones in a semi-logarithm diagram (Lecroart *et al.*, 2007). However, when transient biodiffusion occurs, the computed Db does not only characterize the biological activity just before core extraction, but integrates also past bioturbation variations. In this study, several seasonal variations are imposed to the biodiffusion coefficient. The advection-diffusion model Eq. (1) is used to calculate the apparent Db (Eq. 2), which is compared to the expected one initially introduced into the model. Negligible difference between the apparent and the expected values implies that the selected radionuclide is perfectly adapted to follow seasonal biological activity. The close comparison of the transient profile with the steady-state one is regularly examined. This condition is necessary to adopt the classical Db calculation.

The temporal dependence of biodiffusion is approached by an input function of prescribed sine shape

$$\mathrm{Db}(t) = \mathrm{Db}_\mathrm{m}\left(1 - \frac{\alpha}{100}\sin(2\pi t/\mathrm{T})\right) \tag{3}$$

where T represents the period of biological oscillations, α the oscillation amplitude, and Db_m the mean value of the biodiffusion coefficient. The use of a different function shape gives similar results for comparable input parameters.

Eq. (1) is simplified by introducing dimensionless variables (Guinasso and Schink, 1975 and Boudreau, 1986a). This transformation proposes a more general approach, reducing the number of free tested parameters and correlatively the number of simulations needed to be performed. Therefore, Boudreau's (1986a) approach to make Eq. (1) dimensionless was followed except for the dimensionless time variable which is expressed relative to the decay rate i.e. $\tau = \lambda t$. In transient study, it is intuitive that sensitivity may depend on the decay rate of the radionuclide. Sensitivity of biodiffusion to seasonal mixing is dominated by the balance between biodiffusion and radioactive decay. Other choices are possible depending on the objects of the study. For example, Boudreau (1986a) expressed time variable relative to the characteristic diffusion time to test the influence of the depth distribution of benthic infaunal organisms within the mixed layer. According to dimensionless approach, Eq. (1) becomes

$$\mathrm{Da}\,\frac{\partial\theta}{\partial\tau}(\varepsilon,\tau) = \mathrm{f}(\tau)\,\frac{\partial^2\theta}{\partial\varepsilon^2} - \mathrm{Pe}\,\frac{\partial\theta}{\partial\varepsilon} - \mathrm{Da}\;\theta \tag{4}$$

where θ is the dimensionless activity defined by dividing A by the initial activity at the sediment-water interface, ε is the nondimensional depth, that is $\varepsilon = z/L$. Pe and Da are the dimensionless Peclet and First Damkohler numbers, respectively (see details in Boudreau, 1986a).

$$Pe = wL/Db_m \tag{5}$$

$$Da = \lambda L^2/Db_m \tag{6}$$

Pe defines the relative influence of burial on biodiffusion in surficial sediments. For example, sediment environment with large Pe value are particularly relevant for the burial of paleoceanographic signal. Da measures the relative influence of decay compared to biodiffusion. Perfect radionuclide is characterized by a decay characteristic time ($1/\lambda$) as close as possible to the biodiffusion time (L^2/Db_m) that means Da is close to 1. The confidence level around 1 is not as restricted as predicted by the theory (see discussion). Excessive decay rates imply that the radionuclide is only detectable in a narrow part of the mixed-layer. Db calculation will be imprecise. Inversely, radionuclides associated with reduced decay rates generate homogeneous radioactive profiles in the mixed-layer. Decay is insufficient relative to biodiffusion to exhibit a radioactive decrease with depth. The classical procedure adopted to compute Db is not relevant in this case. f in Eq. (4) is the dimensionless biodiffusion function defined by

$$f(\tau) = 1 - \frac{\alpha}{100}\sin(2\pi\gamma\tau) \tag{7}$$

where γ is the dimensionless pulse defined by $\gamma = 1/\lambda T$. γ characterizes the importance of the decay period of the tracer relative to the period of the biodiffusion variations (Table 1). Dimensionless approach reduces the number of free parameters from six with the dimensional equation (w, α, Db_m, L, T, λ) to four (Pe, Da, α, γ).

b. Boundaries and initial conditions

The boundary condition at the sediment-water interface ($\varepsilon = 0$) is a fixed flux condition

$$\psi = Pe\ \theta - f(\tau)\frac{\partial\theta}{\partial\varepsilon} \tag{8}$$

ψ is the dimensionless radionuclide flux to the sediment. For convenience, ψ was fixed to 1 which did not affect the results of this study. Boundary condition at the base of the bioturbated layer ($\varepsilon = 1$) is a no gradient condition. Biodiffusion theory assumes a semi-infinite mixed layer with a no-gradient boundary condition at the base of the bioturbated layer. In the classical papers, which deal with modeling of bioturbation with radionuclides (e.g. Boudreau, 1986a; Reed *et al*., 2006; Meysman *et al*., 2007), the mixed-layer depth is fixed. This choice is justified for short-lived radionuclides that do not reached the base of the bioturbated layer. This assumption is only acceptable for the two

long-lived radionuclides that reach the base of the bioturbated layer, (see details in Reed et al; 2006). The error introduced by this assumption is negligible for ^{228}Th and ^{210}Pb.

The initial condition is defined by the analytical steady-state solution of Eq. (4) with $f(\tau) = 1$ (formulae derived from the literature, *e.g.* Boudreau, 1997). These conditions are classically encountered in modeling studies of diagenesis.

c. Model resolution and apparent biodiffusion function calculation

Eq. (4) is solved using a finite element approach implemented with the PDE Modes module of *COMSOL Multiphysics*®. The simulation output consists of depth profiles of radioactive tracers affected by seasonal biological activity. Space step is fixed to 1/30 of the mixed layer. Depth profiles are computed during a five-year period of time with a time step of 1/16 year. In dimensionless analysis, these parameters correspond with a time scale included between 0 and 5λ and a λ/16 time step. From each simulated profile, the apparent biodiffusion function (f_{app}) is computed using the following dimensionless formula:

$$f_{app}(\tau) = \frac{Da}{sl^2} + \frac{Pe}{sl} \tag{9}$$

sl defines the slope of the profile in a semi logarithmic diagram. The standard approach is justified by the similarity between transient and steady-state solutions of Eq. (4). It is clear that detection of transient state is extremely difficult to visualize from measured radioactive profiles in the case of marine sediments dominated by biological activity. Validity of the linear regression that results to the steady-state assumption is nevertheless continuously checked by calculating the correlation coefficient between the logarithm of the activity and the depth along the profile. The length of the profile selected to compute biodiffusion function is defined by the distance between the sediment water interface and the depth for which radioisotope activity is equal to 1/10 of that observed at the sediment-water interface. When the tracer reaches the base of the bioturbated layer, the top half of the bioturbated layer is only considered to compute the apparent biodiffusion function. This process minimizes the influence of the boundary condition at the base of the system. For profiles with insignificant correlation (99 percent confidence level), the length of the profile is reduced in order to try to obtain a more significant correlation. When the distance becomes too small (lower than 1/10 of the mixed-layer depth), the profile is excluded.

Relative difference between f_{app} and f, initially introduced into the model, is explained by insufficient sensitivity of the tracer to seasonal biological mixing. The sensitivity is then quantitatively defined by

$$S = \alpha_{app}/\alpha \tag{10}$$

α_{app} is the apparent amplitude of the temporal variations of the biodiffusion function. Perfect sensitivity of the radionuclide to seasonal mixing implies $S = 1$ whereas insensitivity is associated with $S = 0$. Overestimation (under) of seasonal bioturbation is related to $S > 1$ ($S < 1$, respectively).

3. Pe and Da within marine surface sediments

Prior to performing time-expensive simulations, it is pertinent to determine the actual range of Pe and Da variations in natural environments. To do that, computed Pe and Da values are obtained from a comprehensive database of transport parameters (Db, w and L) reported in the literature. This compilation is based on existing ones extracted from previously published papers (Boudreau, 1994; OMEX database, van Weering *et al.*, 1998; Middelburg *et al.*, 1997; Smith and Rabouille, 2002) updated with recently published data (references in Table 2). Biodiffusion coefficient was calculated from depth profiles of ^{210}Pb, ^{228}Th, ^{7}Be or ^{234}Th assuming validity of both the diffusion-advection model and the steady-state approximation. The mixed-layer depth is extracted from literature or directly determined from published ^{210}Pb depth profiles. In this later case, mixed-layer depth is defined either as the depth of the maximum radionuclide penetration for deep-sea environments associated with small burial rate or, for shelf or coastal environments, as the depth of the strong slope break-up observed along the radioactive profile. Published burial velocities were computed from the long-lived radionuclides (^{210}Pb and ^{14}C). This new compilation offers a comprehensive characterization of transport processes extracted from 387 sediment cores. Database encompasses coastal to deep-sea environments (water-depth ranging from 0 to 5654 meters) with an uneven geographical distribution that depends on either the half-life of the radionuclide, or the proximity of marine research laboratories (Table 2 and Fig. 1). ^{7}Be is only detectable in the shallowest marine sedimentary environment and the Southern hemisphere is poorly constrained. Pe and Da display ranges of five and six magnitude orders, respectively. The range of mixed layer depth is more restricted (from 1 cm to 41 cm, Table 2). So, for a given radioisotope, Pe and Da variations are mainly controlled by Db and w variations.

A convenient way to illustrate the relative importance of transport and decay processes in surficial sediment is to plot the two dimensionless parameters Pe and Da in a logarithmic diagram (Fig. 2 a, b, c). Four sectors can be defined by the lines Pe = 1 and Da = 1. The top left-hand sector (sector 1) corresponds with marine sediments dominated by bioturbation over burial. In this sector, the tracer decay presents a time scale lower than the bioturbation time scale. For convenience, we have chosen the long-lived ^{210}Pb radioisotope to illustrate the influence of the parameters Pe and Da on the shape of the depth profile of radioisotope. The tracer profiles can be modelled with a simple steady-state model of diagenesis that has two layers, the upper one affected by biodiffusion and burial and the lower one by burial only. Biodiffusion coefficient, burial rate and porosity are assumed to be constant. In this simple case, an analytical solution exists for the diagenetic equation. Formulae are derived from the literature (e.g. Boudreau, 1997). In sedimentary environments located within sector 1, the tracer-depth profile presents a clear decrease in the mixed layer and a strong slope break-up located at the base of the mixed layer (Fig. 3a). In this sector, Db and L determination process is robust. In sector 1, short-lived radioisotopes display rapid decay that generally inhibits their penetration beneath the mixed layer. The majority of points associated with ^{234}Th falls inside this sector. On the other hand, the sector 2 characterizes

Table 2. Comprehensive Peclet and Damkohler numbers compilation derived from published biodiffusion coefficients Db, mixed-layer depth L, and burial velocities w′.

Db tracer	Number of data	Water depth (m)	Db ($cm^2\ yr^{-1}$)	L (cm)	w ($cm\ yr^{-1}$)	Pe	Da	Sources
^{234}Th	135	6–4426	0.1–263	1–41	0.001–3	$5\ 10^{-4}$–5.4	1–3200	1-30
^{7}Be	25	0–15	1–40	3–11	0.05–0.5	$1.6\ 10^{-2}$–1.45	4.45–478	7; 21; 31-33
^{228}Th	9	200–3104	0.025–107	3–15	0.0015–0.3	$7\ 10^{-3}$–0.7	0.08–810	14; 23
^{210}Pb	218	0–5654	$3\ 10^{-3}$–370	1–40	10^{-4}–2.4	10^{-4}–9.3	$2.4\ 10^{-3}$–414	3; 4; 7; 11; 13; 14; 16; 23; 24; 29; 34-61

References: (1) Aller and Cochran (1976); (2) Aller *et al.* (1980); (3) Aller and DeMaster (1984); (4) Alperin *et al.* (2002); (5) Bentley and Nittrouer (2003); (6) Boer *et al.* (2006); (7) Boudreau (1994); (8) Chaillou *et al.* (2002); (9) DeMaster *et al.* (1994); (10) Gerino *et al.* (1998); (11) Green *et al.* (2002); (12) Jaeger and Nittrouer (2006); (13) Kim and Burnett (1988); (14) Legeleux *et al.* (1994); (15) McManus *et al.* (1995); (16) Nittrouer *et al.* (1983/1984); (17) Pope *et al.* (1996); (18) Santschi *et al.* (1980); (19) Schmidt *et al.* (2001); (20) Schmidt *et al.* (2002); (21) Schmidt *et al.* (2007); (22) Schmidt et al. (submitted, 2007); (23) Silverberg *et al.* (1986); (24) Smith *et al.* (1993); (25) Smith and Rabouille (2002); (26) Sun *et al.* (1991); (27) Sun *et al.* (1994); (28) Thomas *et al.* (2002); (29) Turnewitsch *et al.* (2000) (30) van Weering *et al.* (2002); (31) Krishnaswami *et al.* (1980); (32) Rice (1986); (33) Smoak and Patchineelam (1999); (34) Anderson *et al.* (1988); (34) Benninger *et al.* (1979); (35) Berg *et al.* (2001); (36) Brand and Shimmeld (1991); (37) Carpenter *et al.* (1985); (38) Cochran (1985); (39) Cochran *et al.* (1990) (40) Crusius and Kenna (2007); (41) DeMaster and Cochran (1982); (42) Henderson *et al.* (1999); (43) Kershaw (1985); (44) Kuehn *et al.* (1993); (45) Li *et al.* (1985); (46) Lynch and Officer (1984); (47) Masqué *et al.* (2002); (48) Mulsow *et al.* (1998); (49) Nozaki *et al.* (1977); (50) Peng *et al.* (1979); (51) Sanchez-Cabeza *et al.* (1999); (52) Sayles *et al.* (2001); (53) Smith *et al.* (2000); (54) Soetaert *et al.* (1996b); (55) Stordal *et al.* (1985); (56) Suckow *et al.* (2001); (57) Tanaka *et al.* (1991); (58) Thomson *et al.* (1993); (59) Thomson *et al.* (2000); (60) van Weering *et al.* (1998); (61) Yang *et al.* (1986).

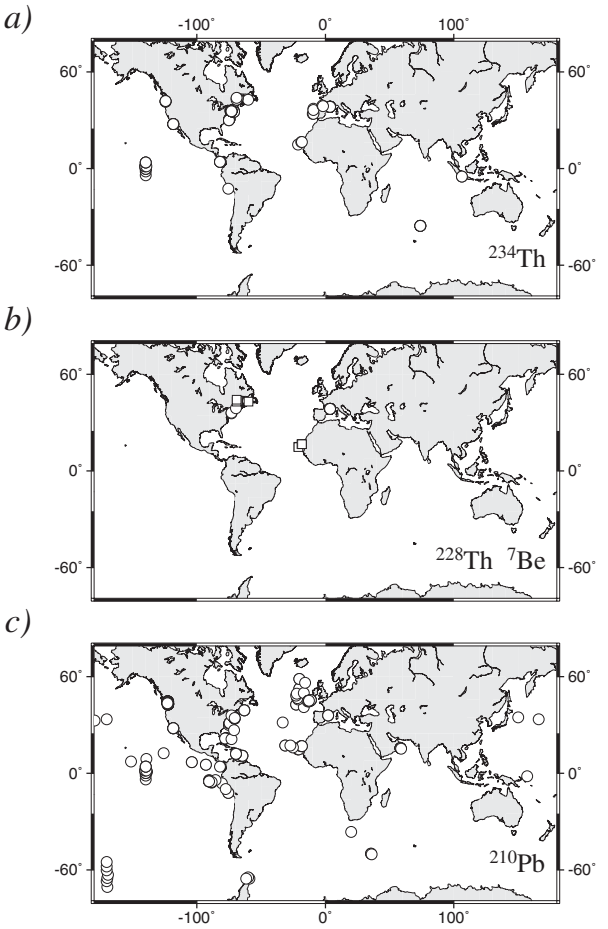

Figure 1. Location map of the sites where a comprehensive description of surficial sediment transport parameters exists. Db values were characterized from (a) ^{234}Th profiles, (b) ^{7}Be (circles) or ^{228}Th profiles (square) and (c) from ^{210}Pb profiles.

natural sediments in which bioturbation dominates both the burial and decay processes. In this case, the mixed layer depth does not display significant radioactive decrease and Db calculation can be submitted to uncertainty due to profile slope inaccuracy (Fig. 3b). Most of the ^{210}Pb points fall inside the sector 1 but a significant quantity is located within sector 2. However, caution must be taken for Db calculation from the smaller Da values located within the sector 2. Sector 3 is associated with sediment dominated by burial relative to biodiffusion. Biodiffusion time scale dominates decay time scale and consequently, radioactive profiles cannot display significant break-up at the base of the mixed layer (Fig. 3c). Db and L determination are thus subjected to caution because of the predominant burial processes. In natural sedimentary environments, this caution only concerns a few

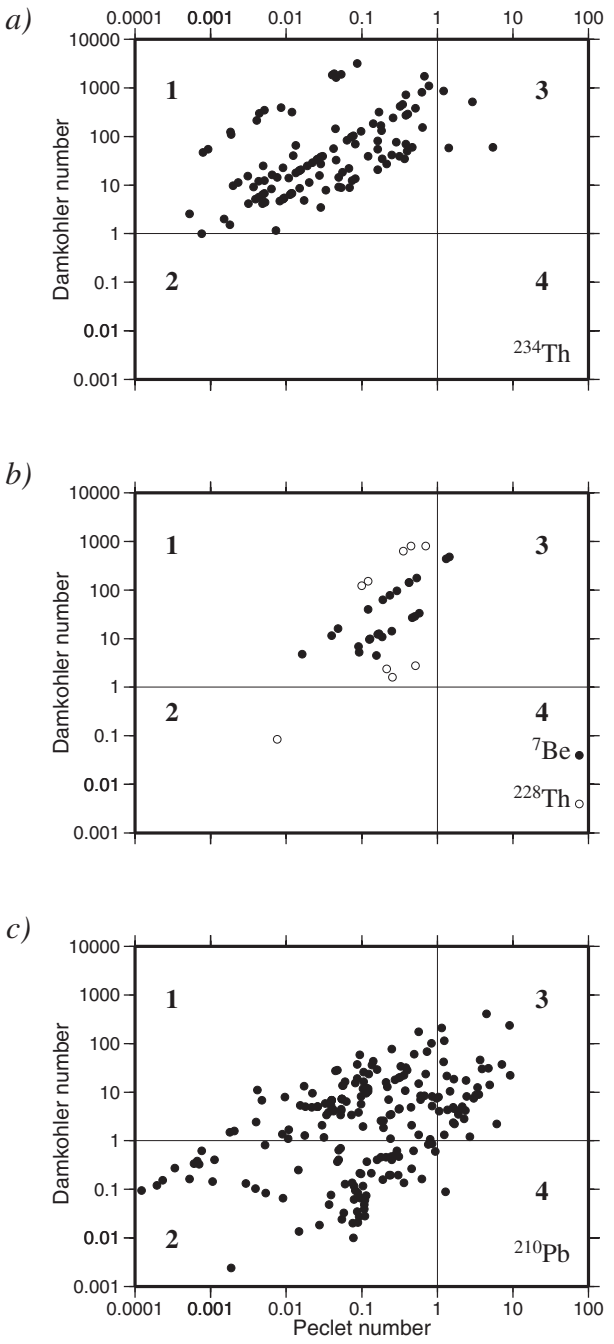

Figure 2. Peclet number *versus* Damkohler number for the different surficial sediment sites. Db used to the dimensionless analysis is obtained from (a) ^{234}Th profiles, (b) ^{7}Be (close circles) or ^{228}Th (open circles) and (c) ^{210}Pb profiles. See text for the signification of the four sectors.

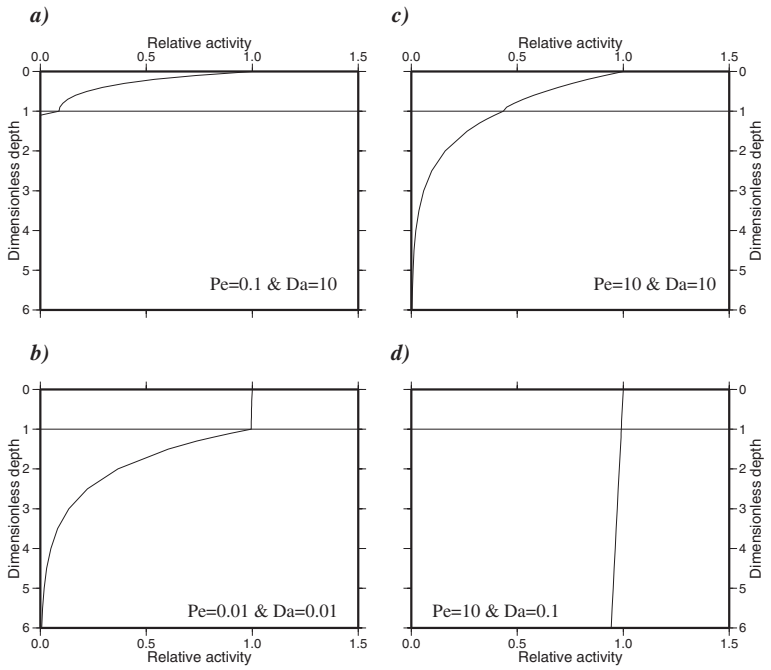

Figure 3. Samples of modeled steady-state depth profiles of ^{210}Pb for different values of Pe and Da. (a), (b) , (c) and (d) profiles from the sectors 1, 2, 3 and 4, respectively. See text for signification of these sectors. Horizontal line represents the base of the bioturbated layer. The diagenetic model is a simple two layers model: the upper layer is affected by biodiffusion and burial and the lower one by burial only. Biodiffusion coefficient, burial rate and porosity are assumed to be constant. In this simple case, an analytical solution exists for the diagenetic equation. Formulae are derived from the literature (e.g. Boudreau, 1997).

^{210}Pb-Db determinations (Fig. 2). Finally, in sector 4, burial overshadows biodiffusion which itself dominates decay. This sector is also characterized by any slope difference along the radioactive profile (Fig. 3d). Additional difficulties arise due to slope uncertainties because sediment is well mixed by biodiffusion and decay rate is too slow to constrain this transport process. Fortunately, any point from natural environments falls inside this problematic sector.

Age-dependent mixing process (Smith *et al.*, 1993; Fornes *et al.*, 2001) or decay period effect (Reed *et al*., 2006) can explain the distribution difference of the Pe and Da values obtained from ^{234}Th and ^{210}Pb. Figure 2 shows that decay rate effect dominates age-dependent mixing on the Da distribution in natural sedimentary environments. Considering both the relationships proposed by Middelburg *et al.* (1997) that relate Db and w with water depth and a constant mixed-layer depth (9.8 cm) according to Boudreau (1998), the shallower sedimentary environments are located toward large values of Pe and small values of Da. Inversely, the deeper environments trend toward lower Pe and larger Da.

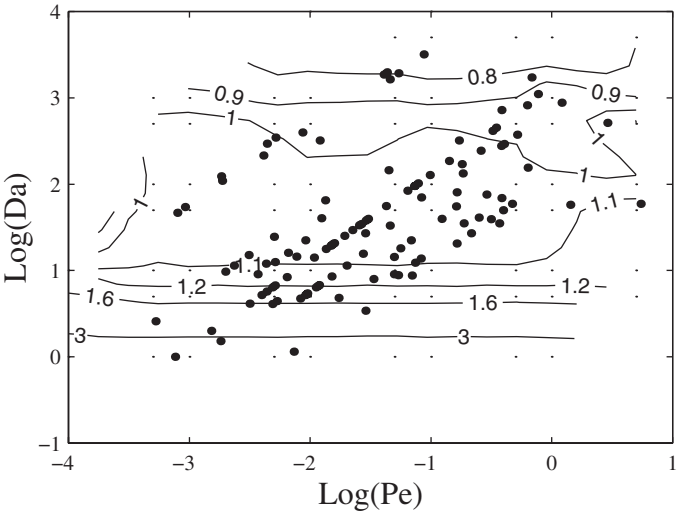

Figure 4. Sensitivity (S) of ^{234}Th to seasonal biodiffusion as a function of the parameters Pe and Da. A perfect sensitivity is defined by S = 1. Satisfactory sensitivity corresponds with $0.9 < S < 1.1$. Dots locate the selected numerical run for which a significant correlation exists between the depth along the profile and the logarithm of the dimensionless activity (> 99 percent significant level). Filled circles represent Pe and Da values for natural sedimentary environments (Table 2). The parameter γ is equal to 0.095.

Nevertheless, it is important to notice that the observed variability in Figure 2 is poorly controlled by global relationships, but mainly by local variations of the Db, w and L parameters. The Pe-Da diagram will be adopted in the next part of the paper to characterize sensitivity of the biodiffusion to seasonal bioturbation.

4. Results

According to the aims and objectives of this study, the period of sinusoidal oscillations is fixed to one year. As λ is a constant value for each selected radioisotopes (^{234}Th, ^{7}Be, ^{228}Th and ^{210}Pb), the number of free parameters is finally reduced to three (Pe, Da, α). A sufficiently strong value for α (80 percent) is selected. This value is important to allow for sensitivity of biodiffusion within seasonal bioturbation.

For each numerical run characterized by a given couple of Pe and Da values, the sensitivity of the radioactive tracer is determined to the imposed seasonal biological mixing. The Pe and Da values are regularly selected inside the range of the compiled natural values (Table 2, Fig. 2). The transient solution of Eq. (4) is numerically solved and biodiffusion function computed from the slope of the radioactive profiles in a semi-logarithm diagram (Eq. 9). The main tests allow the computation of the biodiffusion coefficient through the classical process because the shape of the radionuclide profile is very close to the exponential one which constitutes the solution of the steady-state

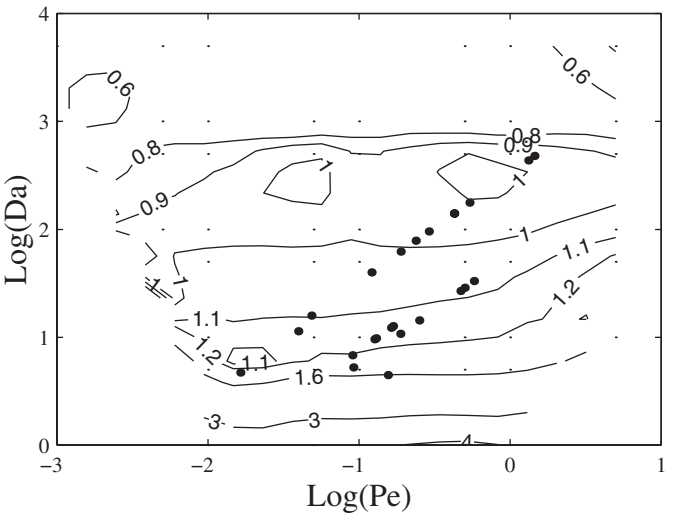

Figure 5. Sensitivity (S) of ^{7}Be to seasonal bioturbation as a function of Pe and Da parameters. Same graphical codes as in Figure 4. $\gamma = 0.211$.

equation. Then, the apparent amplitude of the biodiffusion function is characterized and compared to the actual one initially introduced in the transient model to compute the sensitivity of the radionuclide. Using *Matlab*® routines, a regular grid of sensitivity is interpolated from the (Pe, Da, S) points and then contours of equal value are drawn.

a. Sensitivity of ^{224}Th and ^{7}Be to seasonal bioturbation

The first simulation investigates the sensitivity of short-lived radioisotopes to seasonal biological activity. Two natural tracers are included in this category: ^{234}Th and ^{7}Be. Corresponding pulses (γ) are equal to 0.095 and 0.211, respectively, which implies that seasonal time scale remains dominant over decay time scale. Due to the small penetration of ^{234}Th and ^{7}Be in the sediment, apparent biodiffusion function is then computed along a depth interval comprised between the first quarter and the first tenth of the mixed-layer depth. Sensitivity of ^{234}Th to seasonal bioturbation varies between 0.63 and 3.93 (Fig. 4). For Pe lower than 1, lines of equal values of sensitivity are parallel to the Pe axis. So, sensitivity appears essentially to be dependent on the Da parameter in marine sediments dominated by bioturbation. For Pe greater than 1, sensitivity depends on both Pe and Da values. Satisfactory sensitivity, that can be defined as S inside the 0.9 and 1.1 range, occurs in a large domain included between the lines Da = 10 and Da =1000. For ^{7}Be, sensitivity values are included inside a more extended range comprised between 0.38 and 4.15 and also depend on the Da parameter (Fig. 5). The satisfactory sensitivity domain is observed between the line Da =10 and Da = 500 and is more restricted than for ^{234}Th.

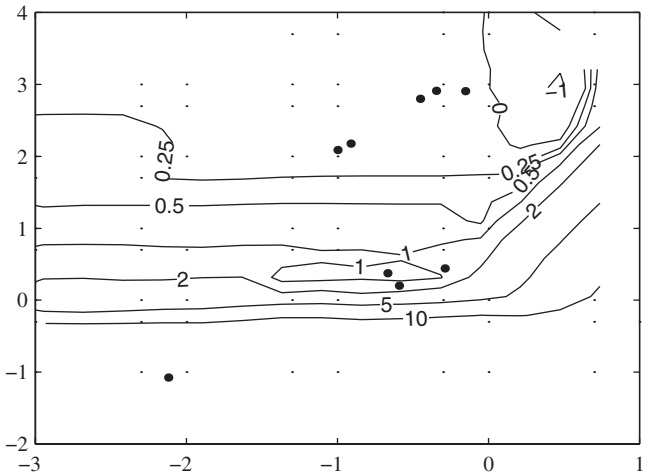

Figure 6. Sensitivity (S) of ^{228}Th to seasonal bioturbation as a function of Pe and Da parameters. Same graphical codes as in Figure 4. $\gamma = 2.74$.

b. Sensitivity of ^{228}Th and ^{210}Pb to seasonal bioturbation

Pulse values are equal to 2.74 and 31.74, respectively. In this case, decay time is larger than seasonal time scale. Sensitivity ranges between 0.02 and 43.16 for ^{228}Th (Fig. 6) and between 0.0071 and 51 for ^{210}Pb (Fig. 7). Pe parameter plays a minor role on the sensitivity of biodiffusion function within the domain defined by Pe lower than 1. The domains

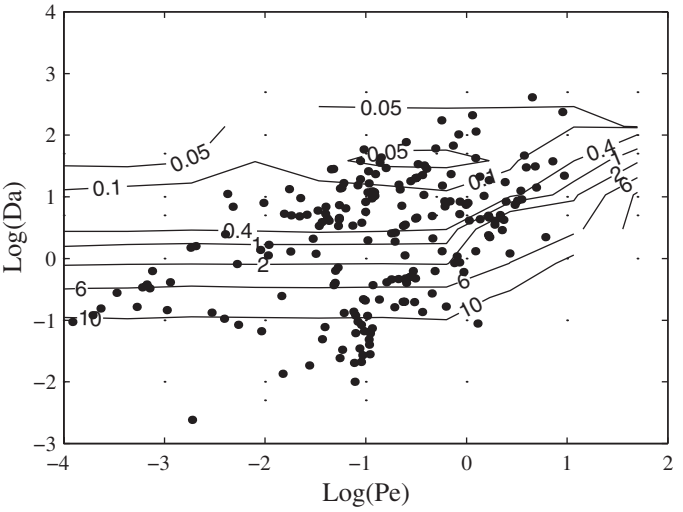

Figure 7. Sensitivity (S) of ^{210}Pb to seasonal bioturbation as a function of Pe and Da parameters. Same graphical codes as in Figure 4. $\gamma = 31.74$.

associated with underestimated or overestimated biodiffusion intensity dominate the diagrams. This result is directly related to the large pulse value associated with these two tracers. As expected, these tracers are clearly inappropriate for modeling seasonal biological activity in marine sediments. This result is valid whatever the selected Pe and Da values except for an extremely small and unexpected domain located in the vicinity of the domain defined by S = 1 (Figs. 6 and 7). This satisfactory domain is included between the lines Log(Da) = 0.7 and Log(Da) = 0.8 ($5.01 \leq Da \leq 6.31$) for ^{228}Th and between the lines Log(Da) = 0.13 and Log(Da) = 0.25 ($1.35 \leq Da \leq 1.78$) for ^{210}Pb. Satisfactory sensitivity domain of ^{228}Th is more extended than for ^{210}Pb. Figure 8 displays the results of a numerical simulation that illustrates the satisfactory sensitivity of the radionuclide ^{210}Pb to seasonal mixing with this particular set of dimensionless parameters. The existence of a satisfactory sensitivity of the long-lived tracers to seasonal mixing is unexpected because ^{210}Pb and ^{228}Th are associated with a decay time scale greater than the time scale of both biodiffusion and seasonal time scale. Nevertheless, the satisfactory domain exists and assumes the necessary transition between the two major domains of under and overestimated biodiffusion. For short-lived radionuclides, this transition dominates the Pe-Da diagram whereas for long-lived radionuclides it is reduced. The greater the radionuclide pulse, the more this transition is reduced. Three sediment cores, for which a complete description of the mixing parameters exists, falls inside the satisfactory sensitivity window of ^{210}Pb: cores 183KG and 215KG from Peru Basin (Suckow *et al.*, 2001) and core KTB14

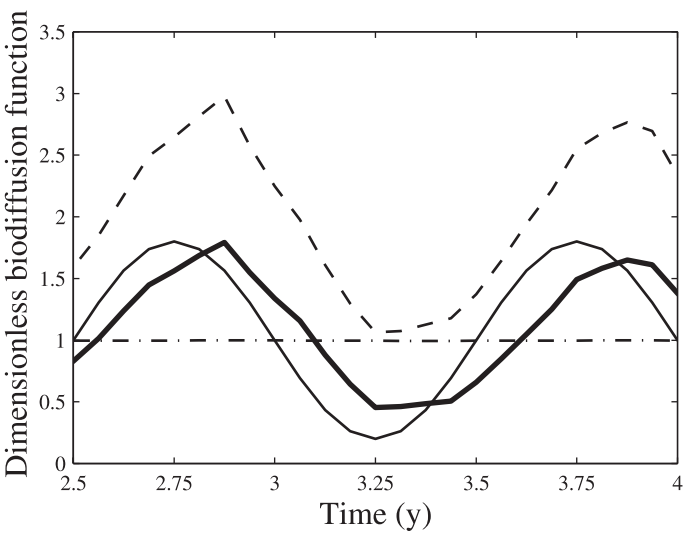

Figure 8. Sensitivity of ^{210}Pb to seasonal biological mixing. The fine line represents the expected variations of the biodiffusion function initially introduced inside the transient model. Bold line is the apparent biodiffusion function calculated from the classical procedure employed on the transient profiles. Model parameters are Pe = 0.1, Da = 1.35, α = 80 percent. For comparison, the dashed line corresponds with a Da value of 1 and the dot-dashed line with a Da value of 100.

from the northeast tropical Atlantic (Legeleux *et al.*, 1994). Other sediment cores are in close approximation: core KTB11 (Legeleux *et al*., 1994), cores Puget Sound E#5 and 89 (Carpenter *et al.*, 1985), core Mid Atlantic Bight station 3 (Tanaka *et al.*, 1991) and core OMEX D8609 (van Weering *et al.*, 1998).

5. Discussion

a. Short-lived radionuclides sensitivity to bioturbation

Reed *et al.* (2006) employed a lattice-automaton model of bioturbation to derive two estimates of the mixing intensity: a radionuclide-derived Db obtained by the standard biodiffusion model and a particle-tracking Db directly derived from the statistics of the particle movements. With a particular set of parameters, they showed that the two derived Db can be significantly different for short-lived radionuclides. The Db derived from short-lived tracers was systematically overestimated compared to the long-lived ones. They concluded that short-lived radionuclides are not appropriate to quantify mixing intensity. Additional investigations with the lattice automaton model are still needed to definitively exclude short-lived radionuclides:

First, the typical marine settings chosen by Reed et al. (2006) are not so typical. The selected sedimentary environments were characterized by an anomalously large $Da = \lambda L^2/Db$ value included between 10^3 and 10^5. These values are rarely observed in natural sedimentary environments (Fig. 4) even if an error of one or two order is affected to Db, as suggested by these authors. In this case, the tracer decay is too fast compared to the biodiffusion process.

Second, Reed et al. (2006, Fig. 2) showed that the sensitivity of radionuclides to mixing intensity is improved when the half-life of the radionuclide increases. For coastal parameter sets, for example, the error between the particle-derived and the tracer-derived biodiffusion coefficients vanishes when the radionuclide half-life reaches 0.5 y^{-1}. They concluded that the short-lived radionuclides are not suitable to correctly quantify bioturbation intensity. Correlatively, the Da parameter decreases from the initial value of 2.10^3 to the final value of 125 where the Db error becomes negligible. Additional tests with lattice automaton model of bioturbation are needed concerning the different parameters that control the system because the error on Db is clearly dependent on the three parameters λ, Db and L and not only on λ: The Db error depends on the competition between the bioturbation intensity and the tracer decay that can be quantified through the $Da = \lambda L^2/Db$ parameter. Reed et al. (2006) explored the influence of λ on Db error, other choices exist that give satisfactory Db values, and especially with the short-lived radionuclides: for example, if bioturbation in the sediment is fixed to a greater value, the Da value can reach the boundary value of 125 and, in this case, the error on Db finally disappears. Increasing sediment mixing improves the sensitivity of the short-lived tracer to bioturbation before decaying. This new situation gives a parameters set compatible with the Da range observed in our compilation (Fig. 4). The third possible track to reduce the Db error consists in decreasing the mixed-layer depth.

This analysis is valid whatever the considered short-lived radionuclide and can be applied to both slope and abyssal environments. In practice, biodiffusion analogy constitutes a satisfactory hypothesis for quantifying sediment mixing even if biological mixing violates the mathematical assumptions of biodiffusion. Short-lived radionuclides are devoted to quantify biological activity for natural environments characterized by a Da parameter lower than 10^2. This result validates 75 % and 79 % of the compiled ^{234}Th-Db and ^{7}Be-Db, respectively. Meysman et al. (2003) used the term 'biodiffusion paradox' to qualify this apparent contradiction between theory and practice. The best way to resolve this contradiction is to suppose that the complex mathematical formalism associated with biological mixing in marine sediment possesses a solution that clearly mimics the profile generated by the biodiffusion analogy. Correlatively, these results highlight the pertinence of the sensitivity study of short-lived radionuclides to seasonal bioturbation, especially in coastal systems where usually ^{210}Pb presents a mixed layer and is not appropriate to such determination.

b. Short-lived radionuclides sensitivity to seasonal biological mixing

The domain of ^{234}Th sensitivity encompasses most of marine surface sediments (Fig. 4). The steady state assumption assumed to compute the biodiffusion coefficient can therefore be extended to the seasonal regime imposed on the system. ^{234}Th is then adapted to follow strong seasonal variations of biological mixing. Among natural radionuclides, ^{234}Th constitutes the more sensible radioisotope able to follow seasonal bioturbation. Its rapid decay rate allows rapid adjustments of the radioactive profiles to seasonal bioturbation variations. For example, the tests reveal that the amplitude of the time variations of the biodiffusion coefficient is perfectly constrained with ^{234}Th during the PULSE study (Gerino *et al.*, 1998). This result supports a few exceptions and some marine sedimentary environments are clearly excluded from the satisfactory sensitivity domain. They correspond with a Da parameter greater than 1000 or lower than10. The marine environments associated with Da greater than 1000 underestimate the seasonal variations of Db. Some examples of such environments have been revealed by the data compilation: they are characterized by either a small biological activity (e.g. cores MC19, MC66 and MC71 from Pope *et al.*, 1996 or core PE138-99-1 from Schmidt *et al.*, 2002) or a large mixed-layer depth (e.g. L = 41 cm for the core 6 from Boer *et al.*, 2006). This configuration of transport parameters limits the tracer penetration and correlatively the opportunity to constrain seasonal biological activity.

Marine environments associated with a Da parameter lower than 10 overestimate the sensitivity of the biodiffusion coefficient to seasonal biological activity. Such environments are generally characterized by either a Db value higher than 40 $cm^2\ y^{-1}$ (e.g. core PULSE 1 from Gerino *et al.*, 1998 and core 1777 from Smith *et al.*, 1993) or a mixed-layer depth lower than 3 cm (e.g. cores C4 from Schmidt *et al.*, 2007). The decay time scale becomes insufficient in relation to the biodiffusion time scale to allow correct discrimination between the seasonal variations of the biological mixing. The mixed layer is saturated

by the radionuclides. The slope of the radionuclide profile converges to zero and the Db value converges to infinity (Eq. 9). The radionuclide profile adopts a conservative shape when biodiffusion dominates decay. The seasonal oscillations are then completely hidden. In some cases, physical reworking associated for example to storms (Gerino *et al.*, 1998), can significantly affect the profile shape and correlatively explain the strong observed biological mixing. The question that remains posed by this study concerns the reliability of the Db value, determined by field data analyses, to the actual biological activity intensity in the case of environments dominated by biodiffusion upon decay.

We can reduce the domain of satisfactory sensitivity to seasonal bioturbation to the domain for which the tracer-derived Db does not overestimate the bioturbation activity (Reed *et al*., 2006). With this consideration, the satisfactory range for ^{234}Th to seasonal mixing sensitivity is included between the Da range of 10 and 125. This domain encompasses most of the natural sediments (Fig. 4).

The domain of satisfactory seasonal sensitivity is slightly more restricted for ^{7}Be than for ^{234}Th. The smaller the pulse γ, the more the domain of satisfactory sensitivity of radionuclide is extended. The majority of natural environments fall inside this domain of perfect sensitivity. The small number of cores, where ^{7}Be have been measured, limits the discussion concerning the natural marine environment for which this tracer is not well adapted to constrain the seasonal variations of the biological mixing. The discussion previously developed and concerning ^{234}Th can be extended to ^{7}Be, but until now no marine environments, characterized by field analyses, correspond with underestimated seasonal biodiffusion amplitude (Fig. 5). Some natural marine environments fall inside the domain with overestimated seasonal oscillations. Biodiffusion dominates radionuclide decay and the seasonal variations of the biodiffusion are not correctly constrained by ^{7}Be.

^{234}Th and ^{7}Be are satisfactory for the great majority of studied marine environments. The classical procedure followed to compute Db is adapted even in the case of the transient regime associated with the seasonal bioturbation. Based on the steady-state assumption, the classical method to compute Db is valid in cases of seasonal biological mixing. It appears more robust to seasonal variations of biodiffusion than to variations of imposed flux of the radionuclide through the sediment-water interface (Lecroart *et al.*, 2007). Such a simple model offers sensitivity quantification for seasonal biodiffusion that can be used to correct the apparent amplitude of seasonal biological activity determined by field analyses. This approach could be applied to more complicated environments with some model adjustments.

c. Long-lived radionuclides sensitivity to seasonal biological mixing

The previous discussion concerning the origin of the imperfect seasonal sensitivity of the short-lived radionuclides can be extended to the long-lived ones. The incorrect sensitivity of these radionuclides was qualitatively expected from a simple time-scale consideration. The important result of this study is to reveal the presence of a small window in which seasonal biological activity can be detected from these radionuclides.

This window is extremely restricted and its position depends on the selected radioactive tracer (Figs. 6 and 7). When a marine environment meets these transport characteristics, ^{210}Pb can be a useful radionuclide to constrain seasonal mixing. Without competing ^{234}Th that constitutes the more adapted tracer, ^{210}Pb has the advantage of a longer half-life, which allows measurements to be obtained after several years. A few natural environments fall inside this satisfactory window (Figs. 7 and 8). They are associated with a special combination of Db and L mixing parameters: Generally, they correspond with marine environments with moderate mixing affecting a deep mixed layer.

The question that can be asked now is to identify the period of biological activity that can be perfectly followed by the long-lived radioisotopes. Assuming that this period corresponds with a γ value lower than 0.1 then this implies that the selected oscillation period must be larger than 3225 years for ^{210}Pb and 27 years for ^{228}Th. ^{210}Pb does not appear to be useful in detecting variations in biological activity in marine surface sediments, except for a small window of correct sensitivity. However, ^{228}Th is perfectly adapted to detect variations of the biological activity related to imprint of human activity on marine sediments at a ten-year time scale.

6. Conclusion

The biodiffusion coefficient is the most commonly used parameter to constrain biological activity in marine sediments. Bioturbation characterization is important because this transport process is dominant in marine sediments and plays a major role in flux determination through sediment-water interface. Different radionuclides are available to compute Db from depth-activity profiles. This method is based on the basic steady-state assumption, but transient regimes are the rule in marine sediments, especially at the seasonal time scale and for coastal to shelf environments.

For most of the sedimentary settings, the short-lived radionuclides are suitable to quantify biological activity. Based on a new global compilation of radionuclide data and on previously published modeling results, we show that short-live radionuclides are perfectly devoted to quantify biological mixing for sediments associated with $\lambda L^2/Db$ lower than 125. 75 % of the ^{234}Th-derived Db, and 79 % of the ^{7}Be-derived Db are concerned by this result.

This study presents extensive sensitivity tests of Db calculation to seasonal mixing and quantitatively reveals that ^{234}Th constitutes the more sensitive tracer to detect seasonal biological mixing. In this case, the steady-state assumption remains valid and applicable for most of natural marine environments. This conclusion can also be extended to ^{7}Be. However, these short-lived radionuclides are not always perfectly reliable for detecting seasonal biological mixing. In some marine environments, defined by particular values of the Da parameter, they incorrectly detect seasonal mixing amplitude. In these cases, the apparent seasonal variations of the biological activity can be corrected with the sensitivity parameter constrained by our study. The major parameter that determines the radionuclide choice is the dimensionless pulse that defines the relative importance of decay time scale

relative to seasonal time scale. The pulse controls the relative extension of the domain for satisfactory sensitivity. With these considerations, long-lived radiotracers are not adapted to follow seasonal mixing processes in marine surface sediments. They are more adapted to detect biological mixing at a longer time scale, except for a specific domain which displays a satisfactory sensitivity to seasonal mixing. Marine environment characterized by moderate biological mixing and deep mixed-layer are concerned by this unexpected conclusion. In that case, ^{210}Pb can be perfectly adapted to follow seasonal change in biological activity in marine sediment.

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