^{239,240}Pu IN ATLANTIC SEDIMENTS : NON-STEADY STATE INPUT

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Abstract

A pulse model to interpret published data on the ^{239,240}Pu distribution in oxic sediments is proposed. We consider that a rapid injection of Pu (mainly III,IV) to the sediment occurred just after the principal nuclear tests in the atmosphere. A slower steady input of Pu (mainly V,VI) has been added to the sediment since that period. The model predicts that less than 10% of the global Pu inventory in sediments can be attributed to a steady input.

Résumé

Les distributions de ^{239,240}Pu dans les sédiments oxyques sont interprétées par la théorie de la diffusion d'une source impulsionnelle. L'injection "instantannée" qui correspond aux principaux essais nucléaires dans l'atmosphère (1963), serait complétée par un apport ultérieur continu de moindre importance. Les valeurs de diffusivité trouvées sont dans la gamme de 1.10^{-8} à 7.10^{-8} cm^2 s^{-1}, les carottes étant prélevées entre 200 et 5000 m.

Introduction

Contrary to other models, this paper assumes that the main source of ^{239,240}Pu in continental slope and deep sea sediments is a "pulse" corresponding to the 1963 maximum fallout. Data from Livingston and Bowen (1979) and Livingston (unpublished) on 12 N. Atlantic cores have enabled to test our basic assumption and to calculate further integrated inputs.

In the present paper, we try to determine which proportion of plutonium in the sediment came from a single instantaneous pulse in 1963 (the year of maximum fallout).

Mathematical treatment

The solution for the diffusion equation without extraneous sources, sinks or transport (the sedimentation rate being negligible) is for a plane diffusing only downwards :

$$C = \frac{2Qo}{\sigma}\left[\frac{1}{\sqrt{2\pi}}\exp -\frac{1}{2}\left(\frac{z}{\sigma}\right)^2\right] \quad (1) \qquad \text{with } \sigma = (2Dt)^{1/2} \quad (2)$$

where Qo is the inventory of Pu in the core per unit surface, σ the standard deviation, D the diffusivity, C(z, t) the concentration along the vertical axis

Qo is measured from the experimental curve (fCi.cm^{-3}), together with surface value C(o,t). The time interval, t, between the source injection and the coring, is known. Hence "D" can be deduced.

Results

Experimental values covering ± 50 % of the core length down to zero concentration values (at about 25cm) enabled the calculation of D with its variance. The following remarks can be made from our numerical results :

a) The values of D all lie within the range of 1 10^{-8} to 7 10^{-8} cm^2 s^{-1}, for the coring depths in the 200 - 5,000 m interval. This order of magnitude

is consistant with known values of diffusivity for other elements (eg. ^{210}Pb : R.A. Berner, 1980 "Early diagenesis, a theoretical approach" Princeton Univ. Press).

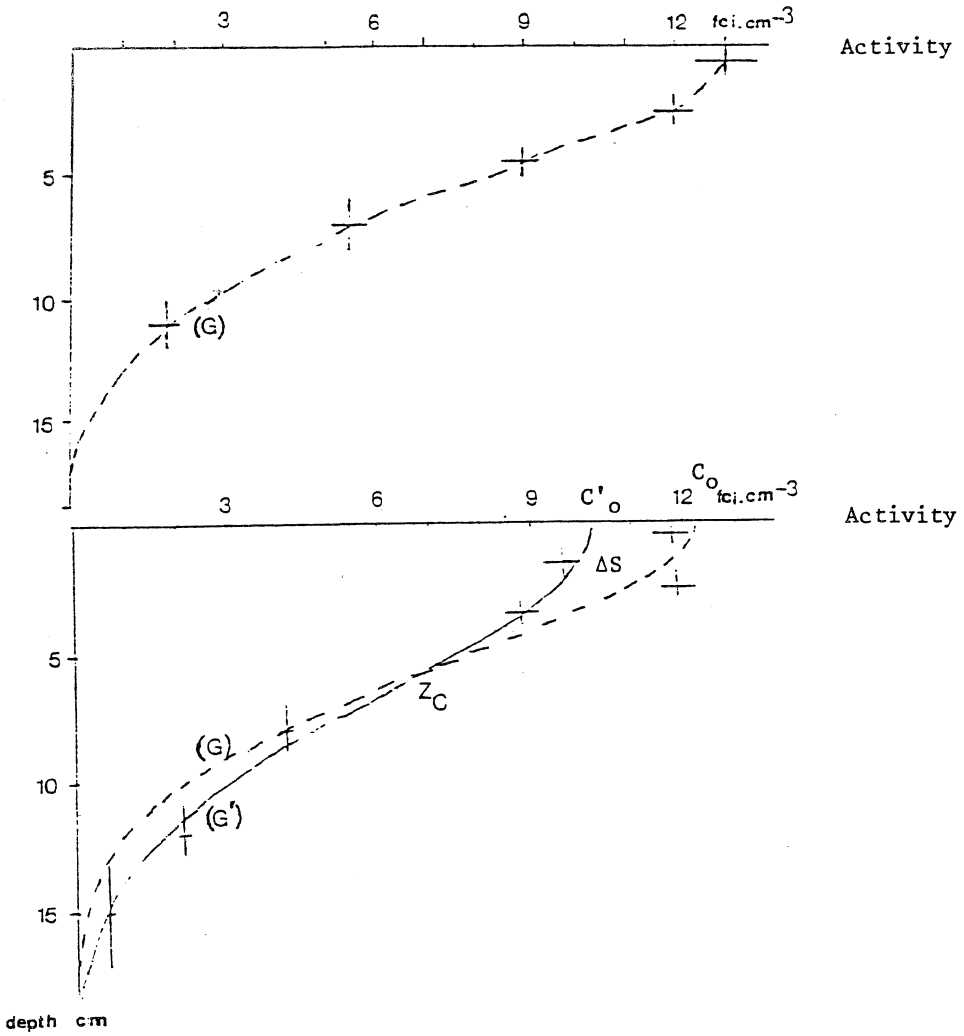

Figure 1A Distribution of Pu-239,240 vs depth in NW Atlantic ocean core 86-6 (1975) 224 m. G = Gauss best fit. D = 4 ± 0.5 cm s^{-1} (10^{-8})

Figure 1B Distribution of Pu-239,240 vs depth in the same zone. core T 13 (1977). G = Gauss best upper fit. G' = best lower fit. D = 3.3 ± 0.6 cm s^{-1} (10^{-8}). ΔS = 8.4 %

b) Surprisingly, the influence of depth on D is relatively small, most values lying in the range of 1 10^{-8} to 3 10^{-8} cm^2 s^{-1} for abyssal sediments and in the range of 3 to 7 10^{-8} cm^2 s^{-1} for shallow sediments (200-300 m). The slightly higher values obtained in the latter case may be interpreted as an effect of greater biological activity.

c) D values do not vary much whether the experimental curves are smooth or perturbated.

Determination of the primary input of Pu to the sediments

A single Gauss curve, G, is obtained for the estimated Co from the experimental curve. It corresponds to a single D value. G was plotted for each of the 12 cores examined, with the following observations :

a) Although most of the data sets show an approximately good fit with the model, the test could only apply to 6 cores where there was enough information.

b) In 2 cases, the G curve was found satisfactory.

c) In 4 cases, the G curve falls too rapidly. Another curve, G', was determined amongst curves having area Qo. It fits the lower part of the experimental points and has a lower surface value, C'o, than G. At first approximation, G' is the solution for a single initial pulse which would have been obtained with a constant D, and G is the altered solution with additional inputs of Pu that shifted the origin upwards from C'o to Co. These are estimated equal to area ΔS between G and G' from the origin to the crossing point z_c.

This surface was found to be at most 25%, the mean value for 6 cores being 7%. The basic assumption that the Pu in the sediments can be treated as the result of a single injection and a small additional input, seems to be justified. One effect of this secondary input is to give an apparent variation of D that could be interpreted as an increase of D with depth (e.g. Fig. 1B). This is contrary to the concept that bioturbation should be proportional to the density of living organisms in the sediment.

Conclusions

Firstly, the value of the diffusivity found for Pu is close to that for other metals (e.g. ^{210}Pb), indicating a common mechanical process.

Secondly, the diffusivity increases slightly with biological activity, leading to a ratio of 2 to 4 between the diffusion in slope and deep-sea sediments.

Finally, the apparent increase of D with depth in the sediment can best be interpreted by a constant D process superimposed on variable inputs of Pu.

Acknowledgments

We are extremely grateful to Dr H.D. Livingston for generously accepting our work on unpublished data he obtained.

Discussion

J.-C. GUARY: Je pense tout d'abord que l'hypothèse impulsionelle choisie pour le modèle n'est pas très réaliste dans le cas présent. Par ailleurs, avez-vous tenu compte du taux de sédimentation dans votre modèle? Enfin, pensez-vous qu'une simple équation de diffusion suffise à expliquer les transferts de plutonium (essentiellement lié à la phase particulaire) dans la colonne sédimentaire?

G. LAPICQUE: Je crois avoir fourni les éléments de réponse: Je suis moi même surpris qu'un modèle si simple puisse rendre compte d'un phénomène très complexe. Quant à l'hypothèse de la prépondérance de l'impulsion, je l'ai explicitée au début de mon exposé. Elle paraît a priori réaliste. Par ailleurs, a postpriori, le bon accord avec l'expérience de

la courbe Gaussienne solution de l'équation justifie l'hypothèse et permet aussi d'estimer l'apport supplémentaire secondaire. Bien sur aucun modèle ne fourni une explication sur la véritable nature d'un phénomène, mais c'est le seul moyen d'obtenir comme je l'ai souligné, une information quantitative de caractère général et objectif. Quant à l'objection sur la non formulation de la vitesse de sédimentation elle n'est pas à retenir ici car ce paramètre est négligeable (e.g. according to H. Livingston and F. Sayles <5 cm pour 1000 ans).