Atti Accademia Nazionale dei Lincei

Classe Scienze Fisiche Matematiche Naturali

Rendiconti

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Essay of a method of absolute dating of the coastal marine sediments by means of the vertical distribution of the fallout radionuclides

Atti della Accademia Nazionale dei Lincei. Classe di Scienze Fisiche, Matematiche e Naturali. Rendiconti, Serie 8, Vol. **45** (1968), n.6, p. 515–521.

Accademia Nazionale dei Lincei

<http://www.bdim.eu/item?id=RLINA_1968_8_45_6_515_0>

Atti della Accademia Nazionale dei Lincei. Classe di Scienze Fisiche, Matematiche e Naturali. Rendiconti, Accademia Nazionale dei Lincei, 1968.

Oceanografia. — *Essay of a method of absolute dating of the coastal marine sediments by means of the vertical distribution of the fallout radionuclides* (*). Nota di BRUNO SCHREIBER (**), presentata (***) dal Corrisp. S. RANZI.

RIASSUNTO. — La grande discontinuità nella ricaduta radioattiva dovuta alle esplosioni nucleari effettuate nell'atmosfera dal 1958 al 1961 ci ha indotti a pensare di poter utilizzare tale fatto per un tentativo di datazione assoluta dei sedimenti marini costieri.

L'osservazione di base dalla quale siamo partiti è che la misura dell'attività beta totale in sezioni successive di carote di sedimento marino (La Spezia) denotava sempre la massima intensità nei primissimi strati, mentre in quelli successivi si aveva una rapida caduta.

Lo studio radiochimico ha dimostrato che i primi strati di sedimento marino costiero contengono i radionuclidi del fallout come Sr^{90}, Cs^{137}, Ce^{144}, Pm^{147}, Eu^{155}, Sb^{125}, insieme a quelli naturali come K, Ra, U, Th; gli strati sottostanti contengono solo radioattività naturale. Inoltre da ricerche di laboratorio circa il comportamento dei suddetti radionuclidi è emerso che, mentre Sr^{90} e Cs^{137} diffondono lungo la colonna del sedimento, Ce^{144}, Pm^{147}, Eu^{155} restano fissati stabilmente all'argilla dello strato in cui si trovano e non diffondono.

Sulla base di tali reperti e dal parallelismo esistente tra l'andamento delle ricadute radioattive e quello della radioattività nei sedimenti da noi considerati è possibile dare i seguenti valori approssimati per la velocità di sedimentazione:

1 cm/anno per la platea continentale del Mar Ligure (La Spezia).
2 cm/anno per il fondo dell'Alto Adriatico vicino al delta del Po.

The great discontinuity shown by the radioactive fallout from 1961 to 1968, suggested our trying an absolute dating of the coastal marine sediments.

The gross beta radioactivity stratification in the latter shows a series of values the trend of which is comparable with that of radioactive fallout.

The previous statement for the validity of this method consists in the possibility of using, as instruments for dating, the radionuclides having particular characteristics. They must be beta emitters of enough energy and in particulate form, adsorbable on sedimentary clays; they must moreover have a half life in accordance with the sedimentation rate of the zone under examination. The radiochemical analysis has luckily shown the presence of Ce^{144} in a large quantity. This nuclide suits this kind of research because it is present above all in particulate form, has a half life of 285 days and a beta energy sufficient for being revealed by a simple gross beta measure-

(*) A summary of this work was presented at the XXI Plenary CIESM meeting (Marine radiochemistry Committee) held in Monaco Principality Sept. 23–28, 1968.

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(***) Nella seduta del 14 dicembre 1968.

ment. Of course other nuclides, even if in a minor proportion, are responsible for the gross beta radioactivity value.

From 1963 to 1968 we analysed the vertical distribution of gross beta radioactivity in about 65 cores collected on the continental shelf of the Ligurian and North Adriatic Seas. The techniques of the research have been already described in our previous works [1, 2]. The graphs of gross beta radioactivity values reported here have been referred to by us as " stratigraphic curves " because they show in ordinate the depths in cm and in abscissa the corresponding gross beta radioactivity values. A true visual representation of the " stratigraphy " of sediment is made possible, emphasizing in the meanwhile the sedimentation phenomenon further occurring in the clayey materials suspended in water, onto which the particulate radionuclides are adsorbed, thus resulting as coeval to the layer they are a part of.

1.—The accumulation of fallout in the first layers.

The observation from which we started is that the gross beta radioactivity measurements showed the highest values in the previous layers, while in the further ones the values are lower.

In the cores collected in 1963, after the maximum values recorded in the previous layers and a sharp decrease, it was still possible to note fluctuations with some peaks, while below 12 cm the gross beta radioactivity values are nearly constant. Secondly we must point out that—based on the decay shown by radioactivity in the previous layers being unnoticeable in the further ones—below 3 cm the values measured successively proved to be constant.

The conclusion drawn is that the radioactivity of the first layers had to be attributed to artificial radionuclides with longer or shorter half life; that of the layers below was due to the natural radioactivity (K, Ra, U etc.).

The radiochemical analysis carried out on mixed samples of more cores collected at the same time have in fact proved the presence in them of fallout radionuclides, particularly Ce^{144} (in about 60 %) together with the other rare earths and Sr^{90} [3, 4, 5].

This fact established, i.e., that the first layers of marine sediments hold and fix a certain number of fallout components, particularly the ones in particulate and not soluble form, we have thought to use this finding for the sedimentation rate evaluation.

2.—Correlation between the radioactivity of the first layers and the fallout values.

Nevertheless we had first to prove the exactness of such a supposition. The first criterion used by us has been the comparison of the gross beta radioactivity values of the first layers with those of the radioactive fallout recorded

from the beginning of the atomic age. If we observe the diagram of fig. 1. we can see that since 1961, the period of the big Russian and American explosions, there has been a progressive increase with a maximum in 1963-64, while from these years there is a rapid decrease.

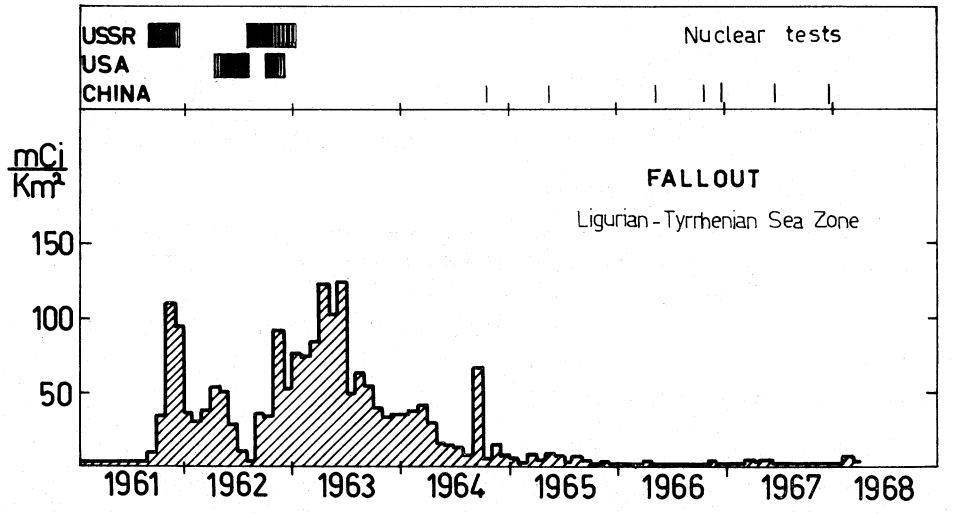

Fig. 1. – Comparison between the frequency of nuclear tests and the trend of radioactive fallout in the collection zone of the Ligurian Sea sediments (La Spezia).

This fact is perfectly shown by the gross beta values in the first layers of the sediment and therefore confirms our supposition.

If we look at the complete diagrams of the radioactivity distribution in depth in the various cores in some of which we give also the radioactivity decay, we can complete the general picture of the phenomenon [6]. Each of them shows confirmation of the constancy of values in the layers below the 12th cm and the gradual decrease of those of the first layers in correspondence with the world fallout decrease.

3.—COMPARISON OF PEAKS.

If we observe in particular some cores, we can see that below the high decrease of the first layers, for instance, in samples of 1963, we note a distinct increase more or less in the 10th-12th cm, while below it the values become again like those of the background.

We have interpreted these higher values found in depth as the remains of the high fallout peak of 1958-59 which obviously, for the time elapsed, had in the meanwhile decayed. Fig. 2 shows this parallelism.

These facts are particularly evident in a collection made in the Po delta in January 1966, especially if compared with a core collected in the Lago Maggiore in 1961 [2, 7].

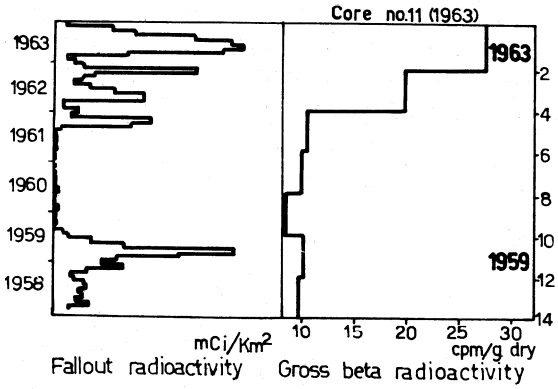

Fig. 2. – Comparison between the beta values of fallout and core fractions of core no. 11 collected in the Ligurian Sea (1963).

In this case and unlike most of the Ligurian cores, the highest radioactivity value was at a depth of 5 cm, while both above and below this one it decreased rapidly as shown in fig. 3.

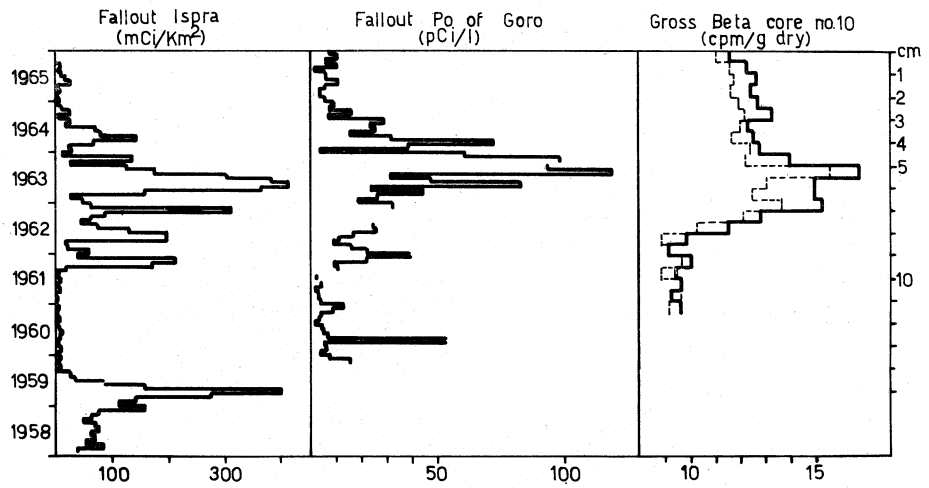

Fig. 3. – Comparison between the beta values of fallout and core fractions of core no. 10 collected in the Adriatic Sea near the Po (1966).

(From B. SCHREIBER et AL., Arch. It. Oceanogr. Limnol., in the press).

The parallelism between the trend of these data and those of Po water radioactivity is very evident.

Unfortunately the thickness of the measured sediment did not allow us to measure the deepest radioactivity values which on the contrary appear in the sediments of the Lago Maggiore collected in 1961 (fig. 4). Here the highest value is at 1.5 cm of depth and the trend of the values above and below this is exactly like that of the fallout from 1958 to about 1960. The two observations above mentioned made both at the end of an intense radioactive fallout, allow to complete the series from 1958 to 1966. If we

arbitrarily make the two maxima in the cores coincide with those of the general fallout curve, we can date, from the time values of this latter, the respective strata of the sediments.

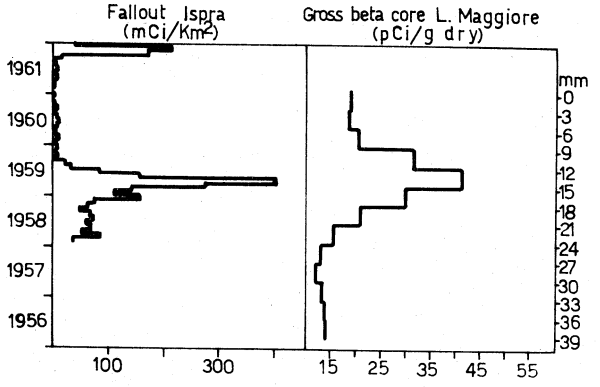

Fig. 4. – Comparison between the beta values of fallout and core fractions of a core collected in the Lago Maggiore (1961).
(From B. SCHREIBER et AL., Arch. It. Oceanogr. Limnol., in the press).

These are found to be 1.5 cm in one year and a half in Lago Maggiore and 5 cm in two years and a half in the Adriatic Sea. In the Ligurian Sea there should be values of about 2 cm in two years. As may be seen therefore the sedimentation rate value should be in the Adriatic Sea Po delta twice that of the Tyrrhenian shelf and of the central Lago Maggiore.

On this subject we have to state that what is called "sedimentation rate" must be understood as " rate of annual deposition " and must not be confused with the " sedimentation speed ".

In a previous work [6] we have considered also this factor which more precisely corresponds with the one that other authors call " residence time " of the particles in the water.

Moreover the values we give on the " annual deposition " have not to be interpreted as a corresponding increase of the bottom level. The values of such an increase are much lower from the well-known phenomena of " compactness " of the actual sediments due to many causes, i.e. pressure, modification of the physical state of the clays and geological sliding.

4.—THE Ce^{144} DECAY.

These values could be indirectly confirmed with a different method and i.e. the one of the theoretic evaluation of the Ce^{144} decay in the quantity present in the superficial layers. The zero point of this curve, in view of the natural radioactivity values of sediments and the sensitivity of the measurement methods, would lead to a zero point after about 5 half lives, i.e. 4 years. In all our cores such a zero point is found in the Ligurian Sea at a depth of about 4 cm.

Our interpretation of radioactivity vertical distribution in marine sediments, as a consequence of the coeval sedimentation of clay and fallout, coincides perfectly with the observations on the sediments of the Clinch River basins where the radioactive wastes are released [8]. Carrigan and Pickering demonstrated the close parallelism between the radioactivity vertical distribution in the sediments of Clinch River basins and the releases of known quantities of radionuclides (fig. 5). It happens the same in the Columbia River with the radioactive wastes of the Hanford reactor [9]. The sedimentation rate values appear in the first case 12.5 cm/year and in the second 10 cm/year.

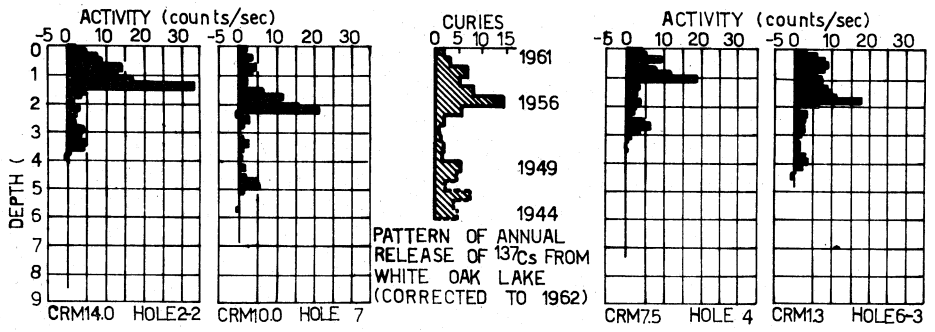

Fig. 5. – Comparison of pattern of variation with depth of gross gamma radioactivity in four bottom sediment cores to variation in annual release for Cs^{137} to Clinch River.
(With the permission of the Chairman of the Clinch River Study Steering Committee).

We want moreover to point out that a similar dating method has been already applied in nature by Wilgain and Picciotto [10], by comparing the radioactivity peaks in the glacial antarctic stratifications and by Lanser, Franz and Knie in the glacier stratification in the Austrian Alps [11].

An interesting parallelism is remarked in what has been seen in glacial and post-glacial depositions.

On the basis of the absolute chronology of the varve deposits which, as is well-known, permits the annual calculation of the deposited layers, Heikki values the " annual deposition " in the Post-Glacial Period in thicknesses from 0.2 to 2 mm/year in cores of the Baltic Sea.

Moreover, Tutin, by means of direct measures of the actual annual deposition at Windermere, estimates thicknesses of 2.6 mm/year.

These values are not very far from the ones resulting from our researches.

Lastly we want to mention briefly the results obtained in our last researches. We have traced with Sr^{89}, Cs^{137}, Ce^{141}, Pm^{147} and Eu^{152} some cores collected in Ligurian Sea and we have noticed that Sr^{89} and Cs^{137} diffuse with different rate along the core, on the contrary the rare earths remain firmly fixed on the first layer on which they precipitated [12].

This last research confirms the exactness of the definition of the method.

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