45

Piotr SZEFER, Stanisław OSTROWSKI
Medical Academy in Gdańsk
Department of Analytical Chemistry

ON THE OCCURRENCE OF URANIUM AND THORIUM IN THE BIOSPHERE OF NATURAL WATERS. PART II. URANIUM AND THORIUM IN CORALS, MOLLUSCS AND FISH

1. INTRODUCTION

Attention of research workers has been focused on the distribution of uranium and thorium not only in aquatic plants but also in some aquatic animal organisms including corals, molluscs, Echinodermata and fish. As with plankton and marine algae, in these organisms also lower levels of both elements were found than in the bottom sediments.

This survey of the literature concerning the occurrence and levels of uranium and thorium in some components of the biosphere of natural waters supplements the data presented in Part I of the present study.

2. URANIUM AND THORIUM IN CORALS

2.1. General remarks

The extent of coral reefs is restricted to warm seas only, as below 18°C corals perish. Another important factor affecting the formation of coral reefs is the cleanness of the water. Suspended particles of impurities may concentrate on the surface of corals and impede their growth. In turn, the irradiation of the water depends on the concentration of impurities. The light intensity, in turn, affects symbiosis of corals with a group of microscopic brown-yellow algae, so-called zooxanthelles. These single-cell plants live inside coral cells and occur in such enormous numbers (about 30,000 individuals per mm^3 of coral tissue) that their colour affects that of the corals themselves. Consequently, zooxanthelles live in a safe environment of corals and utilise their metabolic products, i.e. carbon dioxide, phosphates and nitrates, which provide food for them. On the other hand, corals use up the excess oxygen produced by the algae during photosynthesis. Recent observations suggest the ability of zooxanthelles to stimulate the metabolism of corals without contributing to their feeding process. Corals grow much faster with zooxanthelles than without them.

The fauna of the Atlantic coral reef consists of 35 species belonging to 26 genera, whereas reefs of the Indo-Pacific fauna consists of 700 species and 80 genera. The most abundant are the Acropora (3 Atlantic and 150 Indo-Pacific species) and Porites (3 and 30 species respectively). The size of the particular species depends on the depths at which they grow, strength of current and mass of detritus covering them.

The main components of corals, branching in some cases to form original shapes, are skeletons made from calcium carbonate, which remain after the wasting away and falling off of the living tissue covering them.

2.2. The levels of uranium and thorium in living corals

The level of uranium in living corals ranges from 2×10^{-4} to 4×10^{-4} per cent [13, 22, 24, 29, 31, 40, 45, 50, 61, 63]. The capacity to accumulate uranium in these organisms differs and depends on the species and locality. For instance, mean levels of uranium in species inhabiting the same basin are 4.5×10^{-4}, 3.6×10^{-4} and 3.0×10^{-4} in Madracis asperula, Cladocera patriarca and Bathycyathus maculatus, respectively [31]. A similar relationship was found by Broecker and associates [33] between the coral species and accumulation factor of uranium. Of the Pacific corals the highest levels of uranium were found in the Acropora (2.63—3.03 ppm). Porites (2.6 ppm) and Leptastrea (2.7 ppm) genera [61, 63]. The lowest level of uranium was found in Favia (1.28 — 1.67 ppm) [63].

Among the Atlantic species the highest level of uranium was found in Desmophyllum cristogalli (4.0 — 4.7 ppm). Meandrina braziliensis (4.2, 4.8 ppm), Madracis asperula (4.5 ppm), Trochocyathus sp. (4.0 ppm), Madracis sp. (4.0 ppm), Dendrophyllia sp. (4.6 ppm) [31], and Mussa (4.4 ppm) [40]. The smallest quantities of uranium are accumulated by the Diploria labirinthiformis [50], Montastrea annularis (2.5 ppm) [31, 50], Scolmia cubensis (2.6, 2.7 ppm) and Meandrina areolata (2.7 ppm) corals [31].

In the Red Sea corals, Favia and Stilophora pistillata, 2.24 and 2.46 ppm of U were detected [22].

An inverse relationship was found between the growth rate of corals and their uranium content. In the rapidly growing species the uranium level was 2.88×10^{-4} per cent, whereas in the slowly growing ones $(4.06—4.33) \times 10^{-4}$ per cent [53]. On the other hand the rate of growth of corals is affected by such factors as transparency and temperature of water, irradiation and the presence of zooxanthelles [17]. The enhanced levels of uranium in slow-growing corals can be explained as being due to fractional precipitation of uranium and calcium by polysaccharides in those layers of the corals where calcification takes place [53]. The $^{234}U/^{238}U$ activity ratio in contemporary corals is the same as in the oceanic water and amounts to 1.15 ± 0.02 [14, 18, 26, 40, 56, 58, 60, 61].

The ratio of the U/Ca concentrations in several species of corals from islands and atolls, taken in the central Pacific Ocean amounts to 5.9×10^{-6}, i.e. this is slightly lower than in oceanic water (8×10^{-6}) [63]. In the Atlantic corals the ratio is higher, reaching $(9—14) \times 10^{-6}$ [53].

2.3. On the levels of uranium and thorium in skeletons of fossil corals

There are many reports on the levels of uranium in coral skeleton material from Quaternary and older periods [6, 13, 14, 18, 33, 40, 45, 50, 52, 54, 58, 62, 63]. In the skeletons of marine corals with an aragonite structure, the level of uranium remains constant in contrast to deposits of older corals prone to recrystallization, in which the level of uranium attains a minimum $[(1—2) \times 10^{-4}$ per cent]. A similar phenomenon of leaching the element was observed in older corals exposed to the action of postsedimental tectonic movements on land and in contact with less saline ground and surface waters [25]. Accordingly, diagenetic processes affect the levels of uranium to some extent.

As far as the level of thorium is concerned, skeletons of fossil corals have been insufficiently examined. The levels of thorium in wastes are considerably lower than those of uranium and do not attain 0.1 ppm [29, 58, 61]. Osmond and associates [40] however, found a level of thorium of the order of several ppm in skeletons of old corals.

2.4. Microdistribution of uranium in living corals

In living corals uranium is concentrated principally in three layers: in the organic matter, the aragonite skeleton and on the surface of the skeleton. Most of the uranium from sea water (40—70 ppm) is bound by the organic matter into complexes. The skeleton layer contains only about 3 ppm of U and in the outer parts of the skeleton 0.04—0.06 ppm of U. Due to a low percentage of the uranium-rich organic matter relative to the whole mass of coral (0.1%), this fraction has no significant influence on the level of uranium in these organisms. The fact that only a small quantity of uranium (2 per cent of the total quantity present in the aragonite skeleton) is adsorbed on the surface of coral is explained in terms of a small active surface area of corals, amounting to 1.5—1.8 m^2/g [1].

Examination of other corals showed that the level of uranium decreased on passing from the surface into the inner portions of the skeleton, being frequently 1.6—1.9-times lower than that of the surface parts. In the Oculina diffusa coral, the distribution of uranium is different. There are alternating concentric bands of relatively high and low concentrations of the element. There is also a third type of heterogeneity of microdistribution of uranium in the coral skeletons which manifests itself in disseminated patches of increased uranium contents, the nature of which is independent of the morphological features of the corals [46]. Gvritzman and associates [23] studied the microdistribution of uranium in the Red Sea corals during diagenesis.

3. URANIUM AND THORIUM IN MOLLUSCS

3.1. General remarks

Molluscs inhabit the so-called supralittoral and littoral zones (tidal zone) and even the sublittoral zone (down to a depth of 40 m). The supralittoral zone constitutes an area situated immediately above the so-called high water level, flooded during storms only. The littoral zone is closer to the sea and remains under its constant influence. The epifauna of these zones includes molluscs, insects, crabs etc.

The molluscs include the snails (Gastropoda) and molluscs (Pelecypoda). The size of these invertebrates varies considerably. The giant Tridacna dereasa mollusc attains a length of one meter and a weight of 100 kg, whereas other molluscs are many times smaller and lighter. One species of molluscs lives on sand or coral reefs, others, e.g. Donax, dig themselves into sand periodically overflooded by waves. The tropical snail Oliva belongs to those which dig themselves in most rapidly. Among snails predatory species are also known, e.g. Purpura Thais etc. At present molluscs constitute 75 per cent of all invertebrates caught in an organised manner. In many regions mollusc catches exceed those of fish. So far molluscs have been extracted from their natural environment. The prospects of farming them under artificial conditions seem to be more promising than those of crustaceans. Some molluscs, such as abalons and oysters, were found to contain the narcotic paoline II exhibiting antiviral activity [55].

Owing to the present and future importance of some molluscs for man, the purpose of examining these organisms for toxic elements, including uranium and thorium, need not be substantiated.

3.2. The levels of uranium and thorium in live molluscs

The concentration of uranium in the soft tissue of molluscs was found to be generally lower than in shells. The body of the Rapana thomasiana mollusc taken from the Black Sea contains 5×10^{-6} per cent of U and its shell $(18—25)\times10^{-6}$ per cent [43].

The levels of uranium in the shells of modern marine molluscs fall within the range $(0.001—0.6)\times10^{-4}$ per cent. Similar to corals, the levels vary depending on the locality. In the Pacific waters around California, mollusc shells are particularly enriched in uranium unlike the coastal waters of Japan where the shells contain 5 times lower levels of the element [10]. .

Of the Pacific molluscs (Pelecypoda), the highest concentration of uranium was found in the Protothaca staminea (~ 0.43 ppm of U) and Shizothaerus nuttalli (~0.58 ppm of U) species found off the California coast, the lowest levels being found in Glycimeris aspera (about 0.10 ppm of U) from the coast of Japan and in Gafarium pectinatum (about 0.019 ppm of U) from the Raroia atoll [10].

Among the Pacific snails (Gastropoda) the highest levels of uranium were found in Buccinum baeri morchianum (about 0.39 ppm of U) from the region of the Aleutian Islands and the lowest in Turbo setosus Gmelin (about 0.008 ppm of U) from the Raroia atoll in Cerithium aluco (about 0.006 ppm of U) from Fiji. In the group of Atlantic snails, Littorina littorea found off the coasts of the U.S.A. is worth mentioning, this containing about 0.11 ppm of U. The lowest concentration of this element (~ 0.002 ppm) was found in Tectarius muricatus from the Virgin Islands. Snails from the Pacific Ocean contain from 0.02 ppm (Oliva bulbosa from Zanzibar and Cyprea tigris from Tanganyika) to 0.08 ppm of U (Chicoreus brunneus from India) [10].

As in corals, the enrichment factor of uranium in molluscs depends on the species. In the shells of Marax brevifrons, the concentration of uranium is about 20 times higher than that in Tectarius muricatus taken from the same basin [10], and in the shells of Mya arenaria the concentration is several tens times higher than in Mytilus edulis and Ostrea virginica [57]. No relationship was found between the level of uranium in living molluscs, salinity and temperature [39].

The $^{234}U/^{238}U$ activity ratio in molluscs is comparable with that in corals and amounts to approx. 1.13 — 1.16 [8, 18].

The U/Ca concentration ratio in shells is considerably lower than that in sea water [9].

3.3. The levels of uranium and thorium in the deposits of shells of fossil molluscs

Higher levels of uranium were found in the shells of fossil molluscs than in those of living molluscs, and amounting to about 1 ppm of U [18]. The enhanced concentration of the element is accompanied by the change in the $^{234}U/^{238}U$ activity ratio amounting to 1.50 — 1.89 for shells from the Pleistocene and earlier [18, 47, 57].

3.4. Microdistribution of uranium in the shells of marine molluscs

The distribution of uranium in different structural layers of the mollusc shell was studied. In the outer layer of the shell the uranium content was found to be higher than in the middle layers. This difference is probably due to the migration of uranium towards outer layers of the shell during the growth of the molluscs [9]. Schroeder and associates [46] analysed individual layers of shells of selected molluscs for uranium in detail. In the shell of Mya arenaria the outer crystalline layer contains some uranium, while in the lamellar layer — with the exception of the internal surface — uranium is virtually absent. In the shell of Mytilus edulis, uranium was found only in the innermost layer, which comprises up to 20 per cent of the total thickness of the shell. On the other hand, in the shell of Dendropoma irregulare, the middle portion is poorer in uranium than the inner portions.

3.5. The mechanism of accumulation of uranium in the shells of molluscs

The mechanism of accumulation of uranium in the shells of living molluscs has not been completely elucidated as yet. It is suggested that the crystalline nature of the aragonite skeleton is the most important factor affecting the increase of concentration of uranium. For this reason shells of aragonite structure contain more uranium than the calcite ones. According to another hypothesis, which cannot be ruled out, the increased level of uranium in the outer layer of the shell is maintained owing to the presence of an organic membrane which protects the shell against leaching by sea water [30]. On the other hand, enhanced levels of uranium in fossil shells of molluscs are due to a continued concentration of the element from the environment (sediments, water, etc.).

Results of other investigations [39] suggest that diagenesis is responsible for enhanced levels of uranium in the shells of living molluscs. The most important role, however, is played by organic matter (conchioline) and the possibility of the formation by uranium (U^{4+}) of isomorphic crystals with calcium.

4. URANIUM IN FISH

4.1. General remarks

Fish are reckoned among the nectonic organisms, i.e. those moving at a speed which permits them to be independent of sea and oceanic currents. Fish provide the major component of necton and play an important role in the food system of the sea, and particularly as a source of food for man. Some fish species, such as Trachinus draco and synacea contain toxic substances which reduce the heart beat rate and act hypotensively [55].

Unlike the sedentary organisms of natural waters, fish generally provide an unrewarding object for studies on the contents of various elements, as their nomadic mode of life makes the determination of the concentration and selectivity factors (among others for uranium and thorium) difficult.

4.2. The levels of uranium in live freshwater fish

Freshwater fish taken in the Bihar area contain about 0.2 ppm of uranium, [34], whereas fish from the Issyk-Kul Lake, which is rich in uranium, contain $(0.25—0.34)\times10^{-4}$ per cent of uranium based on dry matter. A markedly increased concentration of uranium was also found in the Cyprinus specularis species living in some springs in Germany, whose waters have increased levels of uranium [16]. Generally, however, mean concentrations of this element in freshwater fish (e.g. in carp) are lower, amounting to about 0.28×10^{-5} per cent [27, 59]. The concentration factor of uranium in the freshwater fish depends largely on the nature of food these vertebrates consume. Predatory fish have a higher concentration of uranium than species feeding on benthic invertebrates [20]. Moreover, the concentration factor of ^{238}U and ^{232}Th was investigated in some fish [51].

Mizumi and Shinichi [35] examined the accumulation factors of uranium in freshwater fish under laboratory conditions and found the highest quantities in the gills and scales. On the other hand, under natural conditions the greatest accumulation of the element was in the gills and bones.

4.3. The levels of uranium in live sea fish

The concentration of uranium in live sea fish ranges from 10^{-7} to 10^{-5} per cent [5, 41, 42].

Similar to other organisms, uranium is distributed non-uniformly in the body of fish. The highest content (0.05×10^{-4} per cent of U based. on dry matter or 0.01×10^{-4} per cent based on living matter [42, 44]) was found in the outer layer, i.e. in scales, fins, skin and bones. Of the internal organs, enhanced uranium levels were found in the liver and gills (0.02×10^{-4} per cent), as compared with the brain, heart, transversely striated muscles, bile and gonads which contain up to 0.005×10^{-4} per cent uranium only [42].

A similar high content of uranium (more than 90 per cent of its total content in the body) was found in the outer layers of freshwater fish. The accumulation of uranium in the gonads was shown to be identical irrespective of its concentration in the ambient water. This may suggest that uranium plays a role in the reproduction of fish [28].

4.4. The levels of uranium and thorium in the waste of dead fish

In the residue deposited of fish bones on the bottom of seas and oceans, the concentration of uranium attains very high values (up to 700×10^{-4} per cent [7, 11]).

Still higher contents of this element (0.3—0.5 per cent) were found in an apatite fraction of Devonian fossils in sediments rich in fish remains. The material also contained considerable quantities of thorium. So far as uranium was concentrated mostly in the apatite fraction, thorium enriched a hydrocarbon fraction formed during the decay of fish and other organisms [12, 32].

5. URANIUM IN OTHER ORGANISMS

There is little information in the literature about the content of uranium in Echinodermata. In the sea-urchins, Dendraster excentricus and Dendraster laevis, inhabiting the waters off the coasts of California and

Mexico, the concentration of uranium amounts to 0.17—0.18 ppm [50]. Adriatic sea-urchins (Arbata sp.) and Amphiura chiajei contained 0.85 and 0.07 ppm of U, respectively [43]. The concentration factors of uranium for the Arbatia sea-urchin and the Verongia aerophora sponge were 425 and 8 respectively.

6. ON THE MECHANISM OF ACCUMULATION OF URANIUM IN MARINE ORGANISMS

This survey of the literature shows that the concentration of uranium in marine organisms, with the exception of corals, is lower than the mean concentration of the element in the marine and oceanic bottom sediments and the Earth's crust [48]. The U/Ca concentration ratio is always smaller for marine organisms than for sea water. Moreover, the extent of accumulation of uranium in marine organisms was found to be relatively small and comparable with a value characteristic of terrestrial organisms. These findings indicate that uranium is an abiogenic element [21]. In some specific cases, however, it may act as a biostimulant [19].

The concentration of uranium in marine organisms can be due to coprecipitation with calcium carbonate, organic matter and ion exchange. In living organisms, uranium has been shown to be able to form both simple camplexes and chelates with such substances as ATP (adenosine triphosphates), hexose diphosphates and hydroxycarboxylic acids. In plants, uranium can be bound by cellulose derivatives. In the complexing of uranium, both the bicarbonate ion and proteins are important [2—4, 15, 36—38, 41, 49].

Piotr SZEFER, Stanisław OSTROWSKI

Akademia Medyczna w Gdańsku
Zakład Chemii Analitycznej

O WYSTĘPOWANIU URANU I TORU W BIOSFERZE WÓD NATURALNYCH.
CZĘŚĆ II. URAN I TOR W KORALOWCACH, MIĘCZAKACH, SZKARŁUPNIACH I RYBACH

Streszczenie

Dokonano przeglądu literatury na temat występowania uranu i toru w koralowcach, mięczakach, rybach i szkarłupniach. Wykazano, że zawartości uranu w żyjących

koralowcach wynoszą od 2 do 4 ppm i zależą nie tylko od regionu wegetacji i cech osobniczych lecz również od takich czynników, jak przezroczystość, temperatura i naświetlenie wody oraz obecność zooksantelli w tych organizmach. Stosunki aktywności $^{234}U/^{238}U$ oraz stężeń U/Ca w koralowcach są bardzo zbliżone do wartości charakterystycznych dla wody morskiej. Wskazano na duże znaczenie procesów diagenetycznych w kształtowaniu się stężeń uranu w starszych koralowcach.

Zawartości uranu w muszlach współczesnych mięczaków wahają się od 0,001 do 0,6 ppm. Podobnie jak w przypadku koralowców występują tu różnice w zawartości uranu w zależności od regionu życia i cech gatunkowych mięczaków. Stosunek stężeń U/Ca dla muszli jest niższy w porównaniu z tą wartością obliczoną dla wody morskiej a stosunek aktywności $^{234}U/^{238}U$ jest zgodny z analogicznym stosunkiem dla koralowców.

Scharakteryzowano rozmieszczenie uranu w rybach słodkowodnych i morskich oraz wskazano na znaczne zawartości uranu i toru w zwietrzelinach obumarłych ryb. Zwrócono uwagę na nierównomierne rozmieszczenie uranu w niektórych narządach wewnętrznych i w ciele ryby. Wykazano, że zawartość uranu w tych organizmach zależy nie tylko od regionu życia i cech gatunkowych lecz również od charakteru pokarmu,, jakim się żywią te kręgowce.

Uwzględniono mechanizm akumulacji uranu w omawianych organizmach oraz scharakteryzowano charakter rozmieszczenia uranu w koralowcach i mięczakach.

REFERENCES

1. Amiel A. J., Miller D. S., Friedman G. M., *Incorporation of uranium in mordern corals*, Sedimentology, 1973, 20(4), p. 523; C. A., 1974, 80, p. 147733b.
2. Andreev P. F., Chumachenko A. P., *O protsessakh vosstanovleniya urana na prirodnykh organicheskikh veshchestvakh*, Geokhimiya, 1964. 1, p. 16.
3. Andreev P. F., Andreeva I. V., Rogozina E. M., *Vzaimodeystviye solei uranila s komponentami rastitel'noy tkani i ikh nekotorymi proizvodnymi*, Geokhimiya, 1962, 4, p. 313.
4. Andreev P. F., Plisko E. A., Rogozina E. M., *Vzaimodeystviye razbavlennykh solei uranila s khitionom i nekotorymi efirami tsellulozy*, Geokhimiya, 1962, 6, p. 536.
5. Aten A. H. W., Dallenberg J. W., Baccum W. C. M., *Concentration of uranium in sea fish*, Health Phys., 1961, 5(3/4), p. 225.
6. Barnes J. W., Lang E. J., Potratz H. A., *Ratio of ionium to uranium, in coral limestone*, Science,1956, 124(3213), p. 175.
7. Baturin G. N., Kochenov A. V., Senin Yu. M., *Kontsentratsiya urana v sovremennykh okeanicheskikh osadkakh v zonakh pod'yema vod*, Geokhimiya, 1971, 4, p. 456.
8. Blanchard R. L., *U^{234}/U^{238} ratios in coastal marine waters and calcium carbonates*, J. Geophys. Res., 1965, 70(16), p. 4055.
9. Blanchard R. L., Oakes D., *Relationships between uranium and radium in coastal marine shells and their environment*, J. Geophys. Res., 1965, 70(12), p. 2911.

10. Blanchard R. L., Cheng M. H., Potratz H. A., *Uranium and thorium series disequilibriums in recent and fossil marine molluscan shells*, J. Geophys. Res., 1967, 72 (18), p. 4745.
11. Blokh A. M., Kochenov A. V., *Elemnty-primesi v kostnom fosfate iskopaemykh ryb*, Geologiya mestorozhdenii redkikh elementov, vyp. 24, Moscow, Nedra, 1964.
12. Bowie S. H. U., Atkin D., *An unusually radioactive fossil fish from Thurso, Scotland*, Nature, 1956, 177 (4506), p. 487.
13. Broecker W. S., *A preliminary evaluation of uranium series inequilibrium as a tool for absolute age masurement on marine carbonates*, J. Geophys. Res. 1963, 68(9), p. 2817.
14. Broecker W. S., Thurber D. L., *Uranium series dating of corals and oolites from Bahaman and Floride Key limestones*, Science, 1965, 149 (3679), p. 58.
15. Hodge H. S., *Mekhanizm otravleniya uranom*, Materialy mezhdunarod. konf. po mirnomu ispolzovaniyu atomnoi energii (Geneva 1965), t. 13, Moscow, Izd. inostr. lit., 1958. p. 279.
16. Clajus P., Ettenhuber E., *Determination of fish/water accumulation factors of some radionuclides important in human radiation exposure*, Abh. Akad. Wss. DDR, 1974, (Pub. 1970); C. A., 1977, 86, 52240y.
17. *Comparative growth rates of some reef corals in the Carribbean*, Lewis J. B., Axelson F., Goodbody I., Page C., Chislett G., Montreal, McGill Univ., Mar. Sci. Rep. No. 10, 1968.
18. Cherdintsev V. V., *Uran-234*, Atomizdat, Moscow 1969.
19. Ellert G., *Khimicheskiye professii urana*, Nauka i zhizn', 1964, 6, p. 66.
20. Ettenhuber E., Clajus P., Roehnsch W., *Determination of accumulation factors fish/water for radionuclides considered to be significant for radiation exposure of man*, Report 1975, SAAS-175, p. 20; C. A., 1976, 84, P86200j.
21. Evseeva L. S., Perelman A. I., Ivanov K E., *Geokhimiya urana v zone gipergeneza*, izdaniye 2-oe, Moscow, Atomizdat, 1974.
22. Friedman G. M., *Geology and geochemistry of reefs, carbonate sediments, and waters of Gulf of Aquaba (Elat.)*, Red. Sea, J. Sediment. Petrol., 1968, 38 (3), p. 895.
23. Gvirtzman G., Friedman G. M., Miller D. S., *Control and distribution of uranium in coral reefs during diagenesis*, J. Sediment. Petrol., 1973. 43 (4), p. 985.
24. Haglund D. S., *The distribution of uranium in recent carbonate sediments and skeletons of organisms and the effect of diagenesis on uranium redistribution*, Ph. D. Dissertation, N. Y. Rensselaer Polytechn. Inst., 1968.
25. Haglund D S., Friedman G. M., Miller D. S., *The effect of freshwater on the redistribution of uranium in carbonate sediments*, J. Sediment. Petrol., 1969, 39 (4), p. 1283.
26. Koide M., Goldberg E. D., *U^{234}/U^{238} ratios in sea water*, Progress in Oceanography, N. Y., Pergamon Press, 1965, vol. 3, p. 173.
27. Koval'skii V. V., Vorotnitskaya I. E., *The biogenic migration of uranium*, Ukr. Biokhim. Zh., 1966, 38 (4), p. 419; C. A., 1966, 65, p. 20779f.
28. Kovalskii V. V., Vorotnitskaya I. E., Lekarev V. S., *Biogeochemical food chains of uranium in aquatic and terraneous organisms*, Radioecol Concent. Processes, Proc. Int. Symp., Stockholm 1966, 329 (Publ. 1967); C. A., 1968, 68, p. 100007a.
29. Ku T. L., *Protactinium-234 method of dating coral from Barbados Island*, J. Geophys. Res., 1968, 73(6), p. 2271.
30. Lahoud J. A., Miller D. S., Friedman G. M., *Relationship between depositional environment and uranium concentrations of molluscan shells*, J.

Sediment. Petrol., 1966, 36(2), p. 541.

31. Livingstone H. D., Thompson G., *Trace element concentration in some modern corals*, Limnol. Oceanogr., 1971, 16(5), p. 786.
32. McKelvey V. E., *Uranium in phosphate rock*, U. S. Geolog. Surv. Profess. Paper, 1956, 300, p. 477.
33. *Milankovitch thypothesis supported by precise dating of coral reefs and deep sea sediments*, Broecker W. S., Thurber D. L., Goddard J., Ku T. L., Matthews R. K., Mesolella K. J., Science, 1968, 159 (3812), p. 297.
34. *Mineral content of aquatic foods*, Viswanathan R., Bhatt Y. M., Sreekumaran C., Doshi G. R., Cogate S. S., Bhagwat A. M., Unni C. K., Proc. Indian Acad. Sci., Sect. B., 1966, 64 (6), p. 301.
35. Mizumi K., Shinichi O., *Uranium concentration in fishes and plants associated with the discharge of low level uranium liquid waste, Tokai Jigyosho*, Dorycku-do, Kakunenryo Kaihatsu Jigyo-dan (Rep.), 1972 PNCT 831-72-0, 1, 180-4; C. A., 1973, 78, p. 6881 7n.
36. Neuman W. F., Main E. R., *The deposition of uranium in bone*. IV. *Adsorption studies in vitro*, J. Biol. Chem., 1949, 179, p. 325.
37. Neuman W. F., Main E. R., *The deposition of uranium in bone*. V. *Ion exchange studies*, J. Biol. Chem., 1949, 179, p. 335.
38. Neuman W. F., Main E. R., *The deposition of uranium in bone*. VI. *Ion competition studies*, J. Biol. Chem., 1949, 179, p. 341.
39. Oglobin K. F., Khalifa - Zade Ch. M., *Uranium distribution in shells of recent and fossil molluscs*, Geokhimiya, 1974, 2, p. 315; C. A., 1974, 81 p. 66849n.
40. Osmond J. K., Carpenter J. R., Windom H. L., *Thorium-230/Uranium-234 age of the pleistocene corals and oolites of Florida*, J. Geophys. Res., 1965, 70 (8), p. 1843.
41. Pertsov L. A., *Prirodnaya radioaktivnost' biosfery*, Moscow, Atomizdat, 1964.
42. Pshenin L. T., *O nakoplenii U — U XI azot-fiksiruyushchimi bakteriyami Chernogo moriya*, Dokl. A. N. S.S.S.R., 1960, 133 (6), p. 1448.
43. Risik N. S., *Nakopleniye i mikroraspredeleniye urana v morskikh organizmakh i prirodnykh usloviyakh. Iskusstvennye i estestvennye radionuklidy v zhizni gidrobiontov*, Naukova Dumka, Kiev, 1973, p. 58.
44. Risik N. S., *Macro — and microdistribution of uranium in aqatic organisms under natural conditions*, Mater. Vses. Simp., Teor. Prakt. Aspekty Deistviya Malykh Doz Ioniz. Radiats., 1973 (Publ. 1973), p. 107, A.N:. S.S.S.R, Komi. Filia, Inst. i Syktyvkar U.S.S.R.; C. A., 1974, 81, p. 74093b.
45. Sackett W. M., Potratz H. A., *Dating of carbonate rocks by ionium-uranium method*, U. S. Geol. Surv. Profess. Paper, 1963, 260-B, p. 1053.
46. Schroeder J. H., Miller D. S., Friedman G. M., *Uranium distribution in recent skeletal carbonates*, J. Sediment. Petrol., 1970, 40(2), p. 672.
47. Stearns C. E., Thurber D. L., *Th^{230}/U^{238} dates of the late Pleistocene marine fossils from the Mediterranean and Moroccan Littorals*, Quaternaria, 1965, 7(1), p. 29.
48. Szefer P., *O występowaniu uranu i toru w morskich osadach dennych*, Studia i Materiały Oceanologiczne, 1977, 17, p. 353.
49. Tannenbaum A., Silverstone H., *Toxicology of uranium*, New York — Toronto — London, Interscience, 1951.
50. Tatsumoto M., Goldberg E. D., *Some aspects of the marine geochemistry of uranium*, Geochim. Cosmochim. Acta, 1959, 17 (3/4), p. 201.
51. Telitchenko M. M., *O nakroplenii i raspredelenii urana, toriya i stro-*

ntsiya v dafniyakh i rybakh Biul. Mosk. Obshchestva Ispytan. Prirody, otd. biol., 1958, 63 (3), p. 57.

52. *The status of U-series method of mollusc dating*, Kaufman A., Broecker W. S., Ku T. L., Thurber D. L., Geochim. Cosmochim. Acta, 1971, 35(11), p 1155.
53. Thompson G., Livingston H. D., *Strontium and uranium concentrations in aragonite precipitated by some modern corals*, Earth Planet. Sci. Letters, 1970, 8(6), p. 439.
54. Thompson J., Walton A., *Redetermination of chronology of Aldabra Atoll by thorium-230/uranium-234 dating*, Nature, 1972, 240 (5377), p. 145.
55. Thorson G., *Życie w morzu*, Wyd. Morskie, Gdańsk 1976.
56. Thurber D. L., *Anomalous uranium-234/uranium-238 ratios in nature*, J. Geophys. Res., 1962, 67 (11)), p. 4518.
57. Thurber D. L., *The dating f molluscs from raised marine terraces by the thorium-230/uranium-234 method*, Symposium on Marine Geochemistry, N. Y. Univ. Rhode Island, Occas. Publ., 1965, 3, p. 1.
58. *Uranium-series ages of Pacific atoll corals*, Thurber D. L., Broecker W. S., Blanchard R. L., Potratz H. A., Science, 1965, 149(3679), p. 53.
59. *Uranovye biogeokhimicheskiye pishchevye tsepi v usloviyakh Issyk-Kulskoi kotloviny*, Koval'skii V. V., Vorotnitskaya I. E., Lekarev I. S., Nikitina E. V., Tr. Biogeokhimicheskoi laboratorii, vol. 9, Moskva, 1968.
60. Veeh H. H., *Thorium-230/uranium-238 and uranium-234/uranium-238 ages of elevated pleistocene coral reefs and their geological implications*, Ph. D. Dissertation San Diego Univ. of California 1965.
61. Veeh H. H., *Thorium-230/uranium-238 and uranium-234/uranium-238 ages of pleistocene high sea level stand*, J. Geophys. Res., 1966, 71(14) p. 3379.
62. Veeh H. H., Chappel J., *Astronomical theory of climatic change support from New Guinea*, Science, 1970, 167 (3919) p. 862.
63. Veeh H. H., Turekian K. K., *Cobalt, silver and uranium concentrations in reef-building corals in the Pacific Ocean*, Limnol. Oceanogr., 1968, 13 (2), p. 304.