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ON THE OCCURRENCE OF URANIUM AND THORIUM IN THE BIOSPHERE OF NATURAL WATERS. PART I. URANIUM AND THORIUM IN PLANKTON AND INHERENT PLANTS

1. INTRODUCTION

Limiting uranium and thorium levels in natural waters usually vary between several hundredths to several micrograms per dm^3 and in the marine and oceanic sediments from several to a dozen or so ppm. Lower levels of the two elements were found in other components of the marine environment, among others in the algae and plankton.

The objective of the present study is to complement an earlier review of literature on the occurrence of uranium and thorium in natural waters [43] and in marine sediments [41].

The uranium levels in the above-ground organs of terrestrial plants, in freshwater organisms and in human tissue have been found to be relatively low, varying between 10^{-9} and 10^{-5} per cent [8, 10, 11, 20, 21, 29, 32, 47, 48, 51].

Some terrestrial plants adsorb uranium and thorium to comparable levels, accumulation coefficients of the two elements being smallest in young parts (sprouts, leaves) and attain maximum values in old parts (branches, bark, wood) [15]. The variations in the uranium and thorium levels may depend, however, not only on the age of the organs of a plant but also on the site of its growth, nature of the soil and the species [5, 28, 49]. Some species of microorganisms and lower plants (fungi, lichens) are exceptionally active in sorbing uranium [9, 17, 33]. On the other hand, increased levels of uranium in other organisms usually occur when they grow in an environment (water, soil) enriched in the element [30, 44, 52].

2. URANIUM AND THORIUM IN PLANKTON OF NATURAL WATERS

2.1. General remarks

There is an enormous amount of suspended living and dead matter in natural waters. It floats or swims in the water, the dead components being referred to as detritus. The matter can be divided into two components, the organic (dead fragments of organisms) and inorganic (clays, erosion slime, slime from melting glaciers, desert dust, volcanic dust, calcareous and siliceous skeletons of microscopic algae).

Plankton is a representative of the living biomass. This term defines all living organisms drifting with the current. They may make slight movements or move vertically, but are helpless in respect of horizontal water currents. Plankton consists of phytoplankton, including the drifting plant matter such as Diatomae, Dinoflagellatae, Coccolithinae, and zooplankton, including animals, the size of which ranges from 0.01 mm to several metres, which have some ability to move by themselves, e.g. Crustacea, Pteropoda, Aurelia and free-swimming larvae of benthic animals. Despite this classification, it is frequently difficult to draw the line between phytoplankton and zooplankton, as some of the species bear features of both plants and animals. This group includes Globigerina, Radiolaria and other species, referred to as the transitional plankton [45].

2.2. The uranium level in inland waters plankton

The ^{232}Th isotope was found [1] to be more adsorbed by small algae, Chlorella ellipsoides and Scenedesmus accuminatus than the ^{238}U isotope. Further, differences have been detected in the accumulation of uranium and thorium in bottom sediment. detritus and lake plants [12, 13]. The ^{232}Th isotope was sorbed to the greatest extent by the freshwater detritus, whereas aquatic plants displayed the highest affinity to the ^{238}U isotope. The ^{226}Ra isotope was found to be uniformly distributed in the two components of the environment.

Plankton of the Issyk-Kul Lake was marked for its maximum uranium level amounting to 2.2×10^{-4} per cent based on dry matter. The water of the lake was found to be enriched with this element (30 μg U/dm^3). The Chara genus alga is worth mentioning here, the accumulation factor for this being as high as 700—1000 [16, 47].

2.3. Extent of accumulation of uranium in freshwater algae

The biological accumulation of uranium by the Scenedesmus quadricanda alga was studied under laboratory conditions [31]. The interpretation of the results was difficult owing to diversity of physico-chemical forms of occurrence of the uranyl ion and physiological processes taking place in the living mass of the alga. The accumulation of uranium was found to occur in two phases. In the first phase, already one minute after the alga had been in contact with the uranium-enriched water, about 60 per cent of the total amount of the element was bound. In the second phase, equilibrium between the uranium level in water and that of the algal biomass was noted after 6 hours only. The extent of accumulation depended on the uranium level in solution and in exceptional cases on the content of the algal biomass.

2.4. The uranium and thorium levels in plankton of seas and oceans

The levels of uranium in the marine plankton range within $(0.01—1.8) \times 10^{-4}$ per cent based on dry matter [18, 23, 40]. The lowest levels of the element were found in organisms of the transitional plankton (Globigerina) ($n \times 10^{-6}$ per cent U). On the other hand, in the phytoplankton organisms, mainly diatoms, inhabiting tropical waters, the level of uranium was 1×10^{-4} per cent [17].

According to Risik and associates [36] the level of uranium in the Adriatic plankton amounts to $(0.83—6.03) \times 10^{-6}$ per cent based on moist weight and $(0.92—2.44) \times 10^{-5}$ per cent based on ash. The respective concentration factors are 3.5 — 23.2. In Adriatic waters, plankton binds $(1.8—3.3) \times 10^{-9}$ g U/dm^3, which corresponds to 0.1 per cent of the total uranium in sea water. In oceanic water, only 0.08 — 0.003 per cent of the total uranium occurs in suspension [34].

Concentration factors determined for plankton taken during the night were 2—3 times higher than those for plankton taken during the day. This difference is due to the varying content of isopodes and copepodes in marine plankton. During the day copepodes predominate (up to 90 per cent), whereas during the night — isopodes (up to 70 per cent). The latter enrich themselves in uranium, as for the greater period of their lifetime they reside in the bottom mud which contains a higher uranium level [35, 36].

Miyake [22, 23] studied the bioaccumulation of uranium by marine plankton. The author found the extent of sorption of uranium to be 7—15 times higher than that of the ^{230}Th and ^{232}Th isotopes and 3000 times higher than that of radium. At a mean level of uranium in Pacific water

of 3.34×10^{-6} g/dm^3, the mean concentration of the element in plankton was $(1.7 — 7.8) \times 10^{-7}$ g/g and the concentration factors ranged from 48 to 260 [24].

Kharkar and associates [46] analysed zooplankton (predominantly calanoids and cyclopoids) from the Caribbean for content of ^{238}U, ^{234}U, ^{232}Th, ^{228}Th, ^{228}Ra, ^{226}Ra, ^{210}Pb and ^{210}Po. They found the concentration factors in the zooplankton relative to sea water to be the highest for ^{210}Po and slightly lower for ^{228}Th and ^{210}Pb.

The levels of the ^{228}Th isotope in the phyto- and zooplankton found in the waters around South Africa amounted respectively to $(9 — 65) \times 10^{-18}$ and $(2 — 27) \times 10^{-18}$g per g of moist mass suspended in oceanic water, the level of the dissolved form of this isotope being 2.7×10^{-15} g/dm^3 [3]. Shannon [37] supplemented these data and the corresponding values obtained by him (in g per g of dry matter) were 4.7×10^{-6} g of ^{228}Th, 1.7×10^{-12} g of ^{230}Th and 2.7×10^{-7} g of ^{232}Th. The ratio of the mean activity of ^{228}Th/^{232}Th in the plankton is comparable with that of sea water and amounts to about 15.

For the Black Sea phytoplankton, the accumulation factor of ^{234}Th amounts to about 20,000, being lower for zooplankton [50].

2.5. EXTENT OF ACCUMULATION OF URANIUM IN MARINE ALGAE

Heide and associates [2, 19] attempted to isolate uranium from sea water by using single-cell Chlorella algae grown, selected and examined in the laboratory. A particular mutant, subjected to the action of uranium-enriched sea water (0.006 ppm U), is characterized by a high selectivity of sorption, optimal dispersion degree enabling rapid isolation, facile and cheap production of living biomass and a relatively high concentration factor exceeding 1000. This algal biomass accumulates as much as 95 per cent of the total uranium present in sea water [7].

Degens [6] found small Umbilisosphera organisms — coccolites, to concentrate uranium from sea water as a result of their vital functions. Owing to this feature the biggest uranium deposits of sedimentary origin are to be found in the Black Sea basin. They were formed by the depositing of coccolite plankton rich in uranium over more than 5000 years.

2.6. MECHANISM OF CONCENTRATION OF URANIUM IN MARINE PLANKTON

There are no reports in the literature on the mechanism of concentration of uranium in marine plankton. According to Degens, uranium in

the coccolite plankton is bound with proteins and sugars present in the cells of the plankton, hence its concentration is 10.000 times higher than that in sea water [6].

3. URANIUM AND THORIUM IN AQUATIC PLANTS

3.1. General remarks

Among aquatic plants, the most numerous group constitute marine algae which include more than 20,000 species.

In this section, the contents of the two elements in small marine algae have not been taken into account, as these organisms belong to the previously described (section 2.4) marine plankton. Only the levels of uranium and thorium in large species of marine macroalgae and some freshwater plants have been discussed here. The levels of uranium and thorium in marine and freshwater plants vary and depend on a variety of factors such as the specific features of a plant, its age, region of vegetation, level of the elements in the surrounding environment (water, bottom sediments) and nature of particular organs of the plant (roots, shoots, stem, leaves, etc.).

3.2. THE LEVELS OF URANIUM AND THORIUM IN FRESHWATER PLANTS

The ^{238}U isotope was found to be more readily accumulated by feshwater plants than the ^{232}Th isotope [12, 13]. This phenomenon was also observed, on the basis of analysis for the two elements, in Elodea, Myriophyllum [14] and 13 other plant species, including pilewort (Ranunculus) and Potamogeton [1]. Both plants and fish taken from the Shinkawa River were found to contain 0.002—0.3 ppm U based on wet matter [25].

3.3. ACCUMULATION FACTOR OF URANIUM IN PLANTS SUBJECTED TO THE ACTION OF FRESH WATER CONTAINING THE ELEMENT

In laboratory experiments, concentration factors were determined for rice and bean plants exposed for 5—7 days to the action of water artificially enriched in uranium up to a level of 25 ppm. The accumulation factors were 2—5 and 0.1—0.2 for the roots and leaves, respectively [25].

3.4. THE LEVELS OF URANIUM AND THORIUM IN SEAWEEDS

Miyake and associates [24] found 0.04—2.35 ppm U bassed on dry matter in algae. Similar levels were found in inherent seaweeds harvested in 1961—1968 in several sites off the coast of Puerto Rico. The uranium levels in them ranged from 0.07 to 1.64 ppm. Among Chlorophyta living in this area, the highest levels of uranium were found in Halimeda opuntia (1.46×10^{-4} per cent) and Penicillus capitatus (1.64×10^{-4} per cent), whereas the lowest levels were found in Ulva lactuca (0.16×10^{-4} per cent) and Codium isthmocladium (0.13×10^{-4} per cent). Among Rhodophyta, the highest accumulation factors were found in Galaxaura cylindrica (0.85×10^{-4} per cent) and Acanthophora specifera (0.61×10^{-4} per cent) and the lowest in Bryothamnion triquetrium (0.07×10^{-4} per cent). The Dictyota divaricata phaephyte contains 1.27×10^{-4} per cent U and Sargassum polyceratium 0.44×10^{-4} per cent only.

The thorium levels in these seaweeds ranged from 0.02 to 0.62 ppm [4].

The mean level of uranium in seaweeds of the Laminaria species is 0.101 ppm and in the Porphyra tenera rhodophyte 0.164 ppm [39].

In the Black Sea and Adriatic Chlorophyta levels of uranium ranged from 0.004 to 0.135 ppm, whereas in Phaeophyta and Rhodophyta from 0.154 to 0.71 ppm [35]. These values are by 1—2 orders of magnitude lower than those reported for tropical species. This difference is probably due to the low degree of calcification of the seaweeds taken from the Black Sea and Adriatic. As shown by Strohal and Pinter [38], Adriatic Chlorophyta and Phaeophyta cotain 0.01—0.66 ppm of ^{232}Th, and the Rhodophyta taken from this basin show increased levels of ^{232}Th (1—2.3 ppm). Considerable variations in the accumulation factors of ^{234}Th were observed in the Black Sea Chlorophyta Uva rigida and Cystoseira barbata [26, 50] and in Entermorpha linza [50]. The accumulation factors for these plants were 1200, 60 and 700 respectively. The kinetics of bioaccumulation of the ^{232}Th and ^{234}Th isotopes in various kinds of algae was investigated and the concentration factors of these isotopes were determined for four species [27].

Characteristic mean value of the U/Ca ratio for seaweeds from Puerto Rico is only slightly lower than that calculated for sea water.

The Baltic Fucus vesiculosus phaeophyte contains 0.26—0.41 ppm of U and 0.25—0.33 ppm of Th based on dry matter. A similar level of uranium (approx. 0.25 ppm) was found in Cladophora chlorophyte and in Zostera marina, the levels of thorium in these plants being 0.30 and 0.3—0.6 ppm respectively. The mean concentration and selectivity factors of the two elements were determined for these plants. With Zostera marina, some variations of the level of uranium and thorium

were noted, depending on the location. The age of the plant was also found to affect the levels of both elements, as was demonstrated by analysing young and old parts of Fucus vesiculosus [42].

3.5. Mechanism of concentration of uranium and thorium in seaweeds

Based on statistical evidence [4] one can assume that with strongly calcified seaweeds uranium is either co-precipitated with $CaCO_3$ in the form of carbonate complexes or it is concentrated owing to the ion-exchange process. On the other hand, in weakly calcified algae, the accumulation of uranium is by the formation of organic complexes. In the case of thorium also both concentration means are possible depending on the kind of plant. With Rhodophyta and more calcified Chlorophyta seaweeds, thorium is probably concentrated by ion exchange and co-precipitation. The accumulation of the element in the tissue of Phaeophyta, however, occurs owing to the formation of thorium — protein complexes.

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O WYSTĘPOWANIU URANU I TORU W BIOSFERZE WÓD NATURALNYCH
CZĘŚĆ I. URAN I TOR W PLANKTONIE I WODOROSTACH

Streszczenie

Dokonano przeglądu literatury dotyczącej występowania uranu i toru w planktonie i roślinach osiadłych wód naturalnych. Wykazano, że tor w porównaniu z uranem jest w większym stopniu adsorbowany przez niektóre drobne glony słodkowodne. Zawartości uranu i toru w planktonie morskim wynoszą kolejno 0.01—1,8 ppmU oraz 0,27 ppm ^{232}Th (w przeliczeniu na suchą masę). Zawartości dwóch pozostałych izotopów toru wyrażone w 1 g suchej masy planktonowej wynosiły $4{,}7.10^{-16}$ g ^{228}Th oraz $1{,}7.10^{-12}$ g ^{230}Th, przy czym fitoplankton charakteryzuje się silniejszą akumulacją wymienionych izotopów w porównaniu z zooplanktonem.

Wyznaczony dla planktonu stosunek średnich aktywności $^{228}Th/^{232}Th$ jest zbliżony do wody morskiej, wynosi ok. 15.

Scharakteryzowano występowanie uranu i toru w słodkowodnych i morskich roślinach osiadłych. W wodorostach tropikalnych stężenia obu pierwiastków wynosiły 0,07—1,64 ppm U oraz 0,02—0,62 ppm Th. Stężenia uranu w tych roślinach są o 1—2 rzędy wielkości wyższe niż w czarnomorskich i adriatyckich wodorostach, co można tłumaczyć wyższym stopniem kalcyfikacji gatunków tropikalnych. Średnia wartość stosunku stężeń U/Ca dla roślin osiadłych jest niewiele niższa od wartości obliczonej dla wody morskiej.

Przedstawiono zaproponowany przez niektórych autorów pogląd na mechanizm akumulacji uranu i toru w roślinach osiadłych.

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