

CONTINENTAL RADIOECOLOGY

N.V. Kulikov, I.V. Molchanova

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N. V. Kulikov
I. V. Molchanova

CONTINENTAL RADIOECOLOGY

Soil and Freshwater
Ecosystems

This book deals with problems of continental radioecology, a new field of biological science that investigates the mechanisms of the migration, distribution, and biological effects of radionuclides in ecosystems of dry land and inland waters. The behavior of certain radiologically important radionuclides (^{59}Fe , ^{60}Co , ^{90}Sr , ^{91}Y , ^{137}Cs , ^{144}Ce) in the soil/solution and soil/plant systems is considered, as well as the distribution of these radionuclides within the soil/plant cover under natural conditions (in particular, the characteristics of the migration of radionuclides in tundra biogeocenoses) and the behavior of radionuclides in the system of water/freshwater organisms.

The book also discusses questions relating to the radiosensitivity of terrestrial plants and hydrobionts to sources of external and internal radiation, and a number of interesting data are presented on the effects of ionizing radiation on biocenoses.

This publication is intended for specialists in a wide range of fields: ecology, radiobiology, pedology, hydrobiology, and environmental protection. It includes 78 tables, 53 figures, and 480 bibliographical citations.

INSTITUTE OF PLANT AND ANIMAL ECOLOGY
THE URALS SCIENTIFIC CENTER
USSR ACADEMY OF SCIENCES

N. V. KULIKOV
I. V. MOLCHANOVA

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*Soil and Freshwater
Ecosystems*

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In fond memory
of Yelena Aleksandrovna
TIMOFEYEVA-RESOVSKAYA

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INTRODUCTION

Since the fifties of the current century advances in the atomic industry and extensive use of its products in various areas of human activity have caused a steady increase in the radioactive background of the Earth's biosphere as the result of introduction of artificial radionuclides. The emergence of ionizing radiation as an environmental factor of ever-growing intensity acting on all living matter has contributed to the shaping and development of radioecology, a new branch of radiobiology investigating radiobiological phenomena at the level of intricate natural complexes (biogeocenoses) just as other branches of biology investigate these phenomena at the molecular, cellular, and organismic levels. The principal objectives of radioecology are the study of mechanisms of migration, distribution and biological effects of natural and artificial radionuclides in different biogeocenoses (Kuzin and Peredelsky, 1956; Peredelsky, 1957, 1964; Odum, 1957, 1959; Kuzin, 1964, 1967; Polikarpov, 1964; Aleksakhin *et al.*, 1970; Verkhovskaya, 1971; Kulikov, 1971a, b).

The theoretical basis for this research was laid by V. I. Vernadsky in his concept of the biosphere (Vernadsky, 1926), by works in biogeochemistry (Vernadsky, 1938, 1940; Vinogradov, 1933, 1938, 1946, 1952), by V. N. Sukachov in his concept of biogeocenoses (Sukachov, 1945, 1947, 1960, 1966), and by the most recent developments in radiobiology and ecology. A great contribution to the advances in radiobiological research has been made by experimental biogeocenology (Timofeyev-Resovsky, 1957, 1962; Timofeyev-Resovsky and Tyuryukanov, 1967).

G. G. Polikarpov (1964) was the first ever to have summarized the data of radioecological research concerned primarily with marine biogeocenoses. This was the first to deal with the fundamental problems of marine radioecology, the problems that are ultimately associated with the prediction of long-range effects of the radioactive contamination of seas and oceans and with the development of recommendations for the control of contamination and the attenuation of its biological effects.

Along with marine radioecology, a branch of continental radioecology can be distinguished, encompassing research into radioecological processes occurring in biogeocenoses of the land and inland water. Compared to marine and oceanic biogeocenoses, continental biogeocenoses are distinguished by their greater variety, and the habitats of the organisms populating the continent are subject to

considerably more drastic changes. In addition, during the biosphere radioactive contamination, the concentration of radioactive substances in the continental biogeocenoses may rise more rapidly than in the ocean since in the continent these substances are diluted to a much lower degree.

The waters of reservoirs and soils, as constituents of their respective biogeocenoses, differ greatly in their physical and chemical properties. At the same time they are the only natural media that support processes of biological production (Zenkevich, 1948, 1970; Bogorov and Zenkevich, 1966). This is why soils and bodies of water are the most prolific parts of the biosphere in which the most intensive and most varied bioenergetic and biochemical transformations occur. It follows that soils, bodies of water, and their biogeocenoses should be given priority in comprehensive studies aimed at providing a sound basis for the utilization of the biologically productive potential of the Earth and at devising rational relationships between that utilization and the development of industry. This task is of particular importance today because of the high rate of industrialization and urbanization of our planet.

Radioactive nuclides that find their way to the Earth's surface through radioactive fallout or by other pathways contaminate primarily the soil, its vegetative cover, and waterbodies, whence they enter different food chains and migrate together with soil and ground water. Therefore knowledge of regularities governing the behavior of radionuclides in such primary biogeocenotic links as soil/solution, soil/vegetation, and soil/hydrobiont is especially important. In these studies ample possibilities for application of the radioactive indicators method considerably simplify the problems relating to the behavior of nuclides in systems of varying complexity, and make it possible to use the trace amounts of chemical elements whose concentrations frequently do not exceed those in the environment. Apart from their direct contribution to radioecology the results of such studies provide valuable material for successful development of the pertinent branches of biogeocenology, ecology, biogeochemistry, pedology, and agrochemistry. It thus follows that radioecology, which is developing in conjunction with allied sciences, enriches them with both its factual material and novel methods of research.

The issue of the behavior of radionuclides in terrestrial and freshwater ecosystems is discussed in many reports. The most complete summaries of these reports are presented in a number of collections of articles and in monographs (Gulyakin and Yudintseva, 1962; Radioactive Contamination of the Biosphere and Measures for Its Control, 1962; Aleksakhin, 1963; Timofeyeva-Resovskaya, 1963; Radioecology, 1963; Radioactive Fallout, Soil, Plant, Food, Man, 1965; Problems of Radiation Biogeocenology, 1965; Radioecological Concentration Processes, 1967; Behavior of Radionuclides in Model Systems of Terrestrial and Freshwater Biogeocenoses, 1968; Symposium on Radioecology, 1969; Radionuclides in Soils and Plants, 1969; Pavlotskaya *et al.*, 1970; Present-Day Problems of Radiobiology, 1971).

It should be noted, however, that the majority of these studies have so far been conducted with the use of long-lived isotopes of strontium and cesium as the most hazardous from the radioecological standpoint. Less well studied in this respect are ^{59}Fe , ^{60}Co , ^{91}Y , and ^{144}Ce . However, ^{59}Fe and ^{60}Co , being induced nuclides, and ^{91}Y and ^{144}Ce , being uranium fission products, may also enter the biosphere in appreciable amounts as a result of nuclear explosions and the peaceful use of nuclear power plants. Until their complete disintegration these nuclides, along with ^{90}Sr and ^{137}Cs , will continually contaminate the natural waters and soils, thus entering trophic chains and penetrating into the human organism (Molchanova and Kulikov, 1972).

Yttrium and cerium are rare-earth elements whose geochemistry has not yet been adequately studied. This is why the data found in the literature pertaining to the fate of yttrium and cerium in various parts of the biosphere are few and contradictory. It is known that these elements can be present in a solution in the ionic, colloidal, or pseudocolloidal forms. The ionic forms are the ones most firmly retained in soils and cannot be washed out by water or neutral salt solutions. The sorptive properties of the colloidal forms are quite different (Nishita *et al.*, 1956; Rhoads, 1957; Kokotov *et al.*, 1962; Molchanova and Titlyanova, 1965). As to the mechanisms of yttrium and cerium absorption by soils, the data found in the literature demonstrate the participation of these particular elements in the ion-exchange reactions (Robinson *et al.*, 1958; Chuveleva *et al.*, 1962a, b). On the other hand, a number of authors note the participation of these elements in complexing reactions. This is supported by concepts currently accepted in geochemistry, according to which the chelate compounds play the leading role in translocating and concentrating rare elements during hypergenous processes (Shcherbina, 1956; Beus, 1958; Vinogradov, 1961, 1962; Balashov, 1963; Timofeyev-Resovsky *et al.*, 1966).

Cobalt is assigned to a group of chemical elements firmly fixed by soil particles. This accounts for its low mobility in soils and poor accessibility to plants (Kulikov, 1960a, 1965a; Titlyanova and Timofeyeva, 1962; Yuditseva and Gulyakin, 1968). Cobalt deficit in some soils results in a drastic reduction of its content in the vegetative cover and this, in turn, causes severe endemic diseases of plants and animals (Shkolnik, 1950; Peive, 1952; Kedrov-Zikhman, 1955; Kovda *et al.*, 1959). Consequently, studies of cobalt behavior in soil, besides being of universal biogeochemical and radioecological interest, are of considerable importance in connection with the trace elements problem. Solution of this problem calls for a thorough investigation not only of the biological role of trace elements, but also of the regularities governing their migration in differing biogeocenoses.

Besides its radioecological importance, as mentioned above, the fate of iron in soils and waterbodies draws the attention of research workers for a number of other reasons. Iron is widespread in the biosphere and the fairly high mobility of some of its compounds determines the typomorphism of the element in a series of geochemical landscapes (Perelman, 1961). The part that iron plays in forming podzolic, boggy,

meadow, lateritic, and other types of soils is well known. Hardpans, bog ores, illuvial horizons of podzols and solonetz,* ferrous hydroxide pellicles, and gley soil horizons are a result and manifestations of the high migration ability of iron (Rode, 1937; Verigina, 1950, 1953; Yarkov, 1950, 1954; Ponomareva, 1964; Kaurichev and Titova, 1966). Much attention is given to the close relationship between iron migration and the acid-base and redox conditions in a soil medium, as well as to the association of iron with the soil organic matter (Kryukov and Avseyevich, 1933; Vishnyakov and Rabinovich, 1935; Serdobolsky, 1950; Krumbein and Carrels, 1952; Aleksandrova, 1954a; Makhonina and Molchanova, 1961). Numerous works by domestic and foreign researchers have demonstrated that a particularly important part in the migration of iron in soils is played by water-soluble organic substances of a specific nature that can undergo complexing reactions (Bloomfield, 1953, 1955; Kaurichev and Nozdrunova, 1952; De Long and Schnitzer, 1955; Kawaguchi and Kyuma, 1959; Kaurichev *et al.*, 1960; Kononova and Titova, 1961; Muir *et al.*, 1964; Chebotina and Kulikov, 1970; Molchanova and Kulikov, 1970a, 1972). Recently there arose a particular interest in studies on the behavior of iron in soils and natural waters paralleling studies on the behavior of cobalt, yttrium, and cerium. This comes from the fact that in the presence of trace amounts of the said elements iron may play the role of the nonisotopic carrier, as is the case for calcium with respect to ^{90}Sr and for potassium with respect to ^{137}Cs .

It should be stressed that the biogeochemical aspects of radioecological studies noted by the present authors are closely interrelated with the purely radiobiological aspects of such studies which comprise issues of external and internal irradiation effects on organisms and communities of organisms. As will be demonstrated, this relationship manifests itself in that the radiobiological effect of ionizing radiation depends above all on the radiation dose, while the dose is determined by the content, the migration rate, and the type of distribution of radionuclides in a contaminated biogeocenosis. The latter factor, in its turn, depends to a large extent on the species composition, structure and biomass of the community exposed to radiation.

The book is a summary of radioecological investigations conducted for several years by the authors and their collaborators in the Laboratory of Radioecology (Institute of Plant and Animal Ecology, the Urals Branch of the USSR Acad. Sci.). Attention is focused on comparative studies with the use of relatively similar technique of the migration and distribution of the six earlier mentioned radionuclides (^{59}Fe , ^{60}Co , ^{90}Sr , ^{91}Y , ^{137}Cs , and ^{144}Ce) in biogeocenotic links: soil/solution, soil/plant, and water/hydrobiont. Also discussed are some aspects of the effect on organisms and their communities of ionizing radiation from external sources and from the radioactive contamination of the habitat (soil and water). Despite the far from

* Alkaline soil with salt accumulated at a definite depth.

complete state of research on this subject, we find that a summary of the results of the completed work is quite justified, since it will provide a deeper insight into specific methods of the research and give an idea of achievements in the field of continental radioecology.

The authors are greatly indebted to the late Professor N. V. Timofeyev-Resovsky, D.Sc. (Biology), for his valuable advice and scientific guidance of the research at its initial stage. They also wish to thank their collaborators at the Laboratory of Radioecology who participated in carrying out individual studies or contributed to their accomplishment.

Chapter 1

MIGRATION AND DISTRIBUTION OF RADIONUCLIDES IN THE SOIL/SOLUTION SYSTEM

The entrance of chemical elements from the soil into plants, and the rate of inclusion of these elements in biogeochemical cycles and food chains, depend to a large measure on the pattern of interaction of these elements with the soil and their capacity to migrate in it. The migration of chemical elements in the soil may be considered in terms of a continual repetition of a series of processes of absorption of these elements from the soil (through the soil solution) and of their return to the solution (desorption) under the action of various factors, resulting in the dispersal of chemical elements, their directional migration, or their concentration. The factors responsible for such a migration are the chemical properties of the elements, their physicochemical state and concentration, properties of the soil, the pH of the soil medium, the presence in the soil solution of various ions, mobile colloids, organic matter, and a number of other factors.

The chapter deals with a series of experiments designed to investigate the influence of some of the factors listed above on the behavior of ^{59}Fe , ^{60}Co , ^{90}Sr , ^{91}Y , ^{137}Cs , and ^{144}Ce in the soil/solution system (Table 1).

In carrying out the experiments the following soils were utilized: soddy-meadow (the South Urals, Ilmen Reservation), chernozem (Kursk

Table 1

Some characteristic features of radionuclides in initial working solutions

Nuclide	Half-life period	Radiation type	Chemical compound	Concentration of carrier (M) at radioactivity of solution 10^{-5} Ci/liter
^{59}Fe	45.1 days	beta, gamma	FeCl_3	10^{-6}
^{60}Co	5.27 years	beta, gamma	CoCl_2	10^{-6}
^{90}Sr	24.4 years	beta	SrCl_2	No carrier
^{91}Y	61 days	beta	YCl_3	10^{-9}
^{137}Cs	33 years	beta, gamma	CsCl	No carrier
^{144}Ce	282 days	beta, gamma	CeCl_3	10^{-9}

Table 2

Chemical characteristics of soils

Soil	Horizon and depth (cm)	Hygroscopic moisture	Loss on calcination	Humus	Exchangeable bases (mEq/100 gm)			Gross chemical composition (% per calcinated sample)				pH	
		% per absolutely dry sample			Ca	Mg	Si	Al	Fe	Ca	Mg	Aqueous solution	Salt solution
Soddy-meadow	A ₁ (0-10)	4.48	12.6	13.10	40.0	12.0	26.9	9.30	5.28	5.82	3.66	6.8	5.5
Chernozem	A ₁ (0-10)	3.69	12.3	10.60	55.0	3.5	36.4	6.85	2.40	0.85	0.63	6.7	6.0
Soddy-podzolic	A ₁ (0-10)	2.01	8.2	6.80	not determined		32.6	5.72	1.76	1.20	0.28	4.5	3.9
	A ₂ (10-27)	1.49	4.9	0.14			35.8	8.00	1.95	0.80	0.08	5.3	4.8
	B ₁ (27-60)	3.26	2.8	0.22			36.2	7.67	4.56	0.96	0.42	4.9	4.0
Red soil	A ₁ (0-10)	3.71	13.4	5.30	12.0	0.2	33.5	8.95	4.28	0.74	0.11	6.2	5.8

Table 3

Mechanical composition of soils (percent per dry sample)

Soil	Horizon and depth (cm)	Size of granulometric fractions (mm)						
		1-0.25	0.25-0.05	0.05-0.01	0.01-0.005	0.005-0.001	<0.001	<0.01
Soddy-meadow	A ₁ (0-10)	18.41	22.55	15.79	6.09	11.31	22.37	39.77
Chernozem	A ₁ (0-10)	2.74	2.58	38.07	9.65	13.63	28.89	52.17
Soddy-podzolic	A ₁ (0-10)	11.02	12.26	46.82	8.77	9.59	10.13	28.49
	A ₂ (10-27)	7.43	9.36	46.31	11.26	11.04	13.32	35.62
	B ₁ (27-60)	4.64	10.14	10.92	7.50	7.22	28.00	42.72
Red soil	A ₁ (0-10)	4.05	2.04	18.42	13.34	24.91	37.24	75.49

Table 4

Sorption of radionuclides by soddy-meadow soil as dependent upon the length of time during which soil sample interacted with the radioactive solution at pH 8 (percent of content in the initial solution)

Nuclide	Time (hr)					
	0.5	1	2	4	6	12
⁵⁹ Fe	54	50	52	49	48	50
⁶⁰ Co	90	92	91	89	96	95
⁹⁰ Sr	98	98	98	98	—	—
⁹¹ Y	93	93	96	95	97	96
¹³⁷ Cs	99	98	98	98	—	—
¹⁴⁴ Ce	68	80	81	85	76	87

Region, Central Chernozem Reservation), soddy-podzolic* (Moscow Region), and red soil (Georgian SSR).

Chemical characteristics of these soils and the data from mechanical analyses indicate that their physicochemical properties are substantially different (Tables 2 and 3).

In most experiments soil suspensions were used. The experimental technique was as follows: a 1 gm sample of the air-dried soil (fraction of soil particles with diameter < 1 mm) and 20 ml of the respective radionuclide solution were put into a 50 ml plexiglass test-tube and thoroughly agitated in a rotary mixer. The mixing time was chosen following a preliminary study of the kinetics of radionuclide sorption by the soddy-meadow soils from solutions with a pH of 8 (Table 4). The equilibrium distribution of radionuclides in the soil/solution system appears to stabilize within the first hour. For this reason in all subsequent experiments the mixing time for radionuclide solutions with the soil was set at 2 hr. Following mixing the solution was separated from the soil by centrifugation, and samples were then taken to measure radioactivity. The amount of a radionuclide absorbed by the soil was determined by finding the difference in radioactivity between the initial solution and the centrifuged deposit.

In experiments on desorption, a soil sample (following its interaction with the radionuclide solution) was washed with distilled water (20 ml); the desorbing solution was then added and agitated for 2 hr. The amount of a radionuclide desorbed from the soil was determined by the level of radioactivity of the desorbent separated from the soil sample by centrifugation. The experiments were carried out in three to five replications. The procedures for a series of special experiments will be described in the relevant sections of this chapter along with the presentation of the experimental material.

* In experiments with the soddy-podzolic soil samples of three different genetic horizons (A₁, A₂, and B₁) were used.

Absorption of Radionuclides in Diverse Soils

Many researchers note in their papers a high sorptive capacity of soils and soil minerals with regard to the main products of uranium fission, ^{90}Sr and ^{137}Cs . It was found, among other things, that sorption of these nuclides depends on the absorption capacity and mineral composition of soils. Soils with a high content of organic matter and clay minerals have as a rule a greater sorption capacity than do light-textured soils with a low content of humus (Klechkovsky and Gulyakin, 1958; Klechkovsky *et al.*, 1959; Kawase *et al.*, 1959; Jacobs, 1960; Schulz *et al.*, 1960; Kokotov and Popova, 1962; Tyuryukanova *et al.*, 1966; Yudintseva and Gulyakin, 1968).

As to radionuclides ^{59}Fe , ^{60}Co , ^{91}Y , and ^{144}Ce , the data reported in the literature concerning their absorption by different soils are rather contradictory. A number of papers stress that the degree of absorption of trace amounts of these radionuclides is not related to soil properties, since the absorption capacity of any soil is large enough to immobilize radionuclides (Titlyanova and Timofeyeva, 1962; Kevin *et al.*, 1963; Molchanova, 1965b; Chebotina and Titlyanova, 1965; Timofeyev-Resovsky *et al.*, 1966). At the same time there are papers in which dependence of radionuclide absorption on soil properties is nevertheless noted (Chuveleva *et al.*, 1962a, b).

The results of experiments conducted in our laboratory in accordance with the aforementioned technique involving the use of six soil types with marked physicochemical differences are summarized in Table 5. It shows that ^{59}Fe is absorbed by all of these soils to

Table 5

**Sorption of radionuclides from aqueous solution (pH 6)
by different soils (percent of content in the initial solution)**

Nuclide	Soil					
	Soddy-meadow	Cherno-zem	Red soil	Soddy-podzolic, horizons:		
				A ₁	A ₂	B ₁
^{59}Fe	40 ± 15	29 ± 7	29 ± 10	23 ± 5	35 ± 8	22 ± 5
^{60}Co	93 ± 5	98 ± 2	88 ± 5	—	92 ± 3	—
^{90}Sr	98 ± 2	97 ± 1	98 ± 3	98 ± 2	—	—
^{91}Y	93 ± 4	95 ± 3	91 ± 2	90 ± 4	89 ± 6	97 ± 2
^{137}Cs	98 ± 1	98 ± 3	96 ± 2	98 ± 1	—	—
^{144}Ce	94 ± 3	83 ± 10	92 ± 4	90 ± 5	92 ± 5	90 ± 4

a considerably lesser degree than are the other radionuclides studied. Differences in the degree of absorption of the same radionuclide by various soil types proved to be statistically nonsignificant. It is therefore concluded that the extent of a radionuclide absorption from an aqueous solution does not depend on soil properties under the experimental conditions used. As stated above, it may be explained by the fact that the absorption capacity of the soils under consideration is sufficiently large to bind completely such trace amounts of radionuclides present in the initial solution that are apt to be sorbed.

Influence of Solution pH and Concentration of Isotopic Carriers on the Absorption of Radionuclides by the Soil

The migratory form of any chemical element is the form that remains stable in solution. For the slightly hydrolyzable or nonhydrolyzable elements (cobalt, strontium, and cesium) it may be the ionic form, whereas for the readily hydrolyzable elements (iron, yttrium, and cerium) it may be both the ionic and the fine-dispersion colloidal forms.

The state of chemical elements in a solution and their change from one physicochemical form to another is determined by a number of factors, the most important of which are the pH of the solution and the concentration of the elements in it. When the solubility product is reached, as a result of changes in the pH and the concentration of a chemical element in the solution, the element will change its ionic form to the colloidal one, thereby changing its mobility in the soil. It is well known that colloidal particles of metal hydroxides (MgOH , $\text{Al}(\text{OH})_3$, $\text{Fe}(\text{OH})_3$, etc.) are as a rule charged negatively (Starik, 1959), and are therefore ordinarily absorbed by the soil to a lesser degree than are cations. Increasing the concentration of an element in a solution may result in saturation by the element of the absorption capacity of soils, and in bringing the sorption process to a standstill.

The data found in the literature show that the ionic forms of a number of radionuclides are retained by soils rather firmly whereas the colloidal forms have a variable behavior since their sorptive properties are dependable on their dispersity and may vary with time. This circumstance appears to account for a certain disagreement of data on the behavior in soils of the nuclides in question (Nishita *et al.*, 1956; Rhoads, 1957; Starik, 1959, 1960; Makhonina and Molchanova, 1961; Kokotov *et al.*, 1962; Titlyanova and Timofeyeva, 1962; Filipovic *et al.*, 1961; Berdnikov and Oreshko, 1963, 1965; Molchanova and Titlyanova, 1965; Chebotina and Titlyanova, 1965; Kulikov, 1968).

The results of experiments on absorption by various soils of ^{59}Fe , ^{60}Co , ^{90}Sr , ^{91}Y , ^{137}Cs , and ^{144}Ce from solutions with varying pH are shown in Fig. 1. Since in the foregoing section it was indicated that the extent of a nuclide absorption is unrelated to soil properties, Fig. 1 is based on the averaged data obtained in experiments with six soil types.

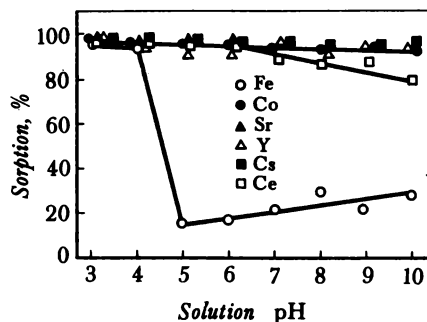


Fig. 1. Sorption of radionuclides by soil as a function of solution pH.

Changes in the acid/base balance of the medium markedly affect absorption of ^{59}Fe in the soil. Sorption of this radionuclide decreases with alteration of the pH from 4 to 5, this being most probably associated with the conversion of its form from the ionic into the colloidal. To the same fact may be attributed the reduced absorption of ^{144}Ce observed in the presence of the alkaline pH. At the same time ^{60}Co and ^{91}Y , hydrolyzable under experimental conditions, are equally well absorbed in the soil regardless of the pH of the initial solution. Such different behavior of iron and cerium, on the one hand, and cobalt and yttrium, on the other, shows that the mobility of these radionuclides in the soil/solution system is determined not only by the form in which they are present in a solution, but also by physicochemical properties of the form (actual dispersion of colloids, sign and magnitude of electrical charge, and so forth). Provided colloidal forms of cobalt and yttrium are positively charged under experimental conditions, the degree to which cobalt and yttrium are absorbed by the soil may be the same as that for ionic forms. The absorption of strontium and cesium radionuclides does not depend on the pH of a solution because with the pH values ranging from 8 to 10 ^{90}Sr and ^{137}Cs are not hydrolyzable.

In other experiments the sorption of radionuclides as a function of the concentration of isotopic carries in a solution was studied. Solutions were prepared from the stable-element salts, with subsequent addition of the required amounts of the respective radionuclides. After addition of hydrochloric acid the solutions were twice evaporated in a platinum bowl with the goal of converting the stable elements and their radioactive isotopes into a single chemical form. Following evaporation the sediment was dissolved in 0.01 N hydrochloric acid and used for preparing solutions of the required concentration.

The experiments showed that the optimum absorption of iron by the soil occurred at an iron concentration in the solution of 10^{-4} M (Fig. 2). In this case the percentage of sorption within the pH range from 4 to 10 remained constant, being on the average 60%. If the concentration value is reduced by one or two orders of magnitude the sorption of iron decreases when the slightly acid pH becomes neutral or alkaline. Such a course of the sorption curve may be attributed to the formation of a ferrous hydroxide solubility product when the

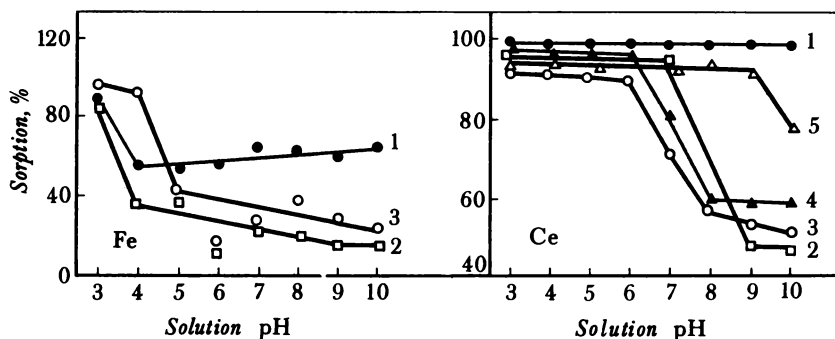


Fig. 2. Sorption of ^{59}Fe and ^{144}Ce by soddy-meadow soil as a function of solution pH and concentration of isotopic carriers. (1) 10^{-4} M; (2) 10^{-5} M; (3) 10^{-6} M; (4) 10^{-7} M; (5) 10^{-8} M.

concentration is 10^{-5} to 10^{-6} with the pH 4 to 5. This is attended by the formation in the solution of the colloidal forms of $\text{Fe}^{3+}\text{-Fe}(\text{OH})_3$, which are more mobile than the cation forms. A relatively higher percentage of sorption and a low dependence of iron absorption on the acid-base balance of the medium at an iron concentration of 10^{-4} are probably due to the formation of large colloid aggregates and to their mechanical adsorption onto soil particles.

Given the molar concentration of cerium in the solution 10^{-4} M the element is virtually completely sorbed by the soil over the entire range of pH values. Reduction of the cerium concentration by one order of magnitude is accompanied by a sharp decrease in absorption within the neutral and acid-base ranges. The course of cerium sorption curves at lower concentrations (10^{-6} , 10^{-7} , and 10^{-8} M) is the same as that of the sorption curve at a concentration of 10^{-5} M, but is characterized by

Table 6

Sorption of radionuclides by soddy-podzolic soil in relation to concentration of isotopic carriers in initial solution at pH 6 (percent of content in the initial solution)

Nuclide	Concentration of stable carrier (mole)						
	10^{-8}	10^{-7}	10^{-6}	10^{-5}	10^{-4}	10^{-3}	10^{-2}
^{59}Fe	—	—	38.4	28.1	56.2	—	—
^{60}Co	—	—	97.0	95.0	97.0	95.5	—
^{90}Sr	—	96.7	97.6	98.0	95.0	93.2	72.0
^{91}Y	95.0	95.0	95.0	97.0	99.0	—	—
^{137}Cs	98.5	98.8	98.5	92.0	87.0	80.0	30.2
^{144}Ce	93.0	95.3	90.0	95.4	98.0	—	—

a less marked decline of absorption in the pH range from 7 to 10. This characteristic feature of cerium absorption by the soil, dependent on the concentration of cerium in the solution and on the pH, is most probably due to the fact that under the given experimental conditions this nuclide is present in solution in varying chemical form. In the pH range of from 3 to 6 cerium may be present as a cation that is almost entirely absorbed by the soil; at higher pH values cerium changes to the hydrolyzable form which results in a reduction of its sorption by the soil.

Comparative data on the sorption by soddy-meadow soil of all six radionuclides under study, as dependent upon the concentration of their isotopic carriers in a solution at pH 6, have demonstrated that the percentage of sorption of the majority of radionuclides remains unchanged within a rather broad range of concentrations. It is only at relatively high concentrations of some elements (strontium and cesium) that the sorption saturation is observed (Table 6).

Effect of Stable Nuclides of Iron, Aluminum, Potassium, and Calcium on the Absorption of Radionuclides in the Soil

It is evident from the foregoing discussion on the effect of the acid-base condition of the medium and the concentration of elements on the mobility of these elements in the soil/solution system that iron forms in a solution, within a wide pH range, colloids that are poorly sorbed by the soil. The presence in natural waters and soil solutions of such colloids of iron may affect the migration ability of other chemical elements, particularly those that are present in the solution in trace amounts (Chukhrov, 1955; Polynov, 1956). Such an influence can be manifested, for instance, by co-sedimentation or adsorption of the trace component, in the course of the colloid formation, onto colloidal particles of macrocomponents having similar chemical properties. In this case the behavior of a trace component fixed on the colloidal particle will be determined not only by its own chemical properties but also by properties of the macrocomponent (Glagoleva, 1961; Molchanova, 1965a). Mobility of the trace component in the soil/solution system may also be controlled by competitive relations with the respective chemically analogous macroelements (Timofeyev-Resovsky *et al.*, 1966).

Such macrocomponents for cobalt, yttrium, and cerium are iron and aluminum, and for strontium and cesium the macrocomponents are calcium and potassium, respectively. This is based first on a certain affinity in the chemical properties of the respective elements, and, second, on the fact that the concentrations of migratory iron, aluminum, calcium, and potassium in soils and natural waters are always several orders of magnitude greater than those of cobalt, strontium, yttrium, cesium, and cerium (Verigina, 1950; Vinogradov, 1950; Perelman, 1955,

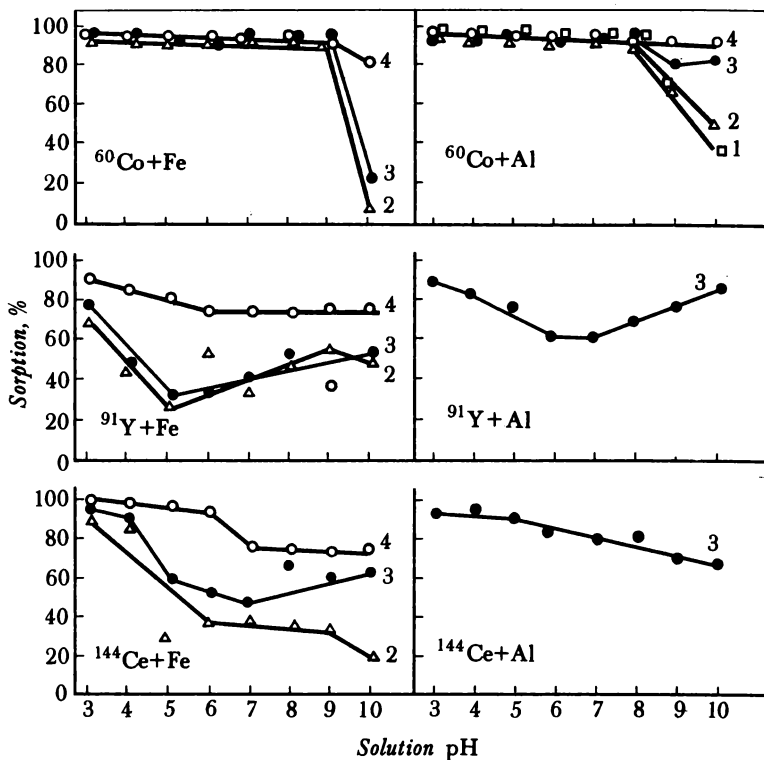


Fig. 3. Sorption of radionuclides by soddy-meadow soil as a function of pH and concentration of stable iron and aluminum nuclides in solution. (1) 10^{-3} M; (2) 10^{-4} M; (3) 10^{-5} M; (4) 10^{-6} M.

1961; Robinson *et al.*, 1958; Skrynnikova, 1959; Kudrin, 1966; Kaurichev *et al.*, 1968).

Tests in our laboratory demonstrated that stable isotopes of iron and aluminum present in a solution noticeably reduce sorption of the cobalt, yttrium, and cerium radionuclides in the soddy-meadow soil (Fig. 3). In experiments with ^{60}Co this reduction occurs at pH 9 to 10, i.e., under such acid-base conditions when cobalt, as well as iron and aluminum, are present in the solution in the colloidal form. It is also noteworthy that the lowest sorption of a radionuclide in the soil was recorded when the macrocomponent concentration was high.

Reduction of soil absorption of the yttrium and cerium radionuclides is also more marked when iron concentrations are higher. Thus, a maximal reduction of yttrium absorption occurs in slightly acid or neutral solutions, while the maximal reduction of cerium absorption occurs in neutral or slightly alkaline solutions. In the presence of aluminum the pattern of yttrium and cerium sorption is retained, although

the reduction of their absorption by the soil is not so distinct as in tests with iron. Absorption of ^{90}Sr by the soil depends inversely on the concentration in the solution of its chemical analog, calcium, while absorption of ^{137}Cs follows the same pattern for its analog, potassium (Timofeyev-Resovsky *et al.*, 1966).

Thus, under certain acid-base conditions of the medium the presence of iron and aluminum in the solution alters appreciably the mobility of the cobalt, yttrium, and cerium nuclides in the soil/solution system. The mechanism of this phenomenon appears to be related to processes of co-sedimentation and adsorption of trace amounts of radionuclides onto iron and aluminum colloids formed under these conditions. A decrease in sorption of ^{90}Sr and ^{137}Cs , in the presence of calcium and potassium cations, respectively, is most probably associated with competitive relations between the elements-analogs.

Influence of Artificial Chelating Agents on the Absorption of Radionuclides in the Soil

Artificial chelating agents (substances that, together with cations of some chemical elements, form readily soluble chelate compounds) are finding ever increasing use in the control of endemic diseases of plants (Antonini, 1954; Bould, 1955; Wallace *et al.*, 1955a, b, 1957; Kroll, 1957; Stewart and Leonard, 1957; Churbanov and Alekseyeva, 1967). Some researchers consider these substances from the standpoint of their potential as agents for effectively influencing the fate of radionuclides that contaminate the soil and water reservoirs (Kulikov, 1961a; Timofeyeva-Resovskaya, 1963; Essington and Nishita, 1966; Nishita and Essington, 1967; Todorovic and Filip, 1967).

Application of chelating agents for the aforementioned purpose is based on their ability to lower sorption of some chemical elements by the soil; this results in a higher uptake of these elements by plants and enhances their capacity for migration with soil solutions.

According to our data, in the presence of the EDTA chelating agent (ethylenediaminetetraacetic acid) sorption of iron, cobalt, yttrium, and cerium radionuclides is drastically reduced (Fig. 4); sorption of ^{90}Sr is reduced to a much lesser extent, and there is no effect whatsoever on sorption of ^{137}Cs . As is generally known, the former four elements, whose sorption is greatly reduced even when the chelating agent concentration in the solution is low, form very stable chelate compounds with EDTA. Values of the stability constants for these compounds incorporating EDTA are as follows: 14.2 for iron; 16.3 for cobalt; 18.5 for yttrium; and 16.0 for cerium. The stability constant value for chelated strontium is considerably lower (8.6), while that for chelated cesium is still lower (Bjerrum *et al.*, 1957).

It thus follows that sorption of radionuclides by the soil from chelating agent aqueous solutions is primarily determined by the stability constant for the compounds they form with this chelating agent.

This may account for the varying effectiveness of chemically different chelating agents that we have used in studying their effect on the soil sorption of some radionuclides (Fig. 5). Four chelating agents were tested: DTPA (diethylenetriaminepentaacetic acid)— $C_{14}H_{23}O_{10}N_3$; EDTA (ethylenediaminetetraacetic acid)— $C_{10}H_{16}O_8N_2$; EADA (ethanolaminediacetic acid)— $C_6H_{11}O_5N$; and EDBIPA (ethylenediaminebis(isopropyl)phosphinic acid)— $C_8H_{20}O_6P_2N_2$.

All chelating agents under study markedly reduced the soil sorption of cobalt and cerium, i.e., the soil sorption of those elements that usually form stable chelate compounds with organic addenda. As stated above, strontium forms with chelating agents compounds of lower stability; moreover, calcium, a close chemical analog of strontium, competes with the latter. Therefore, with respect to ^{90}Sr the chelating agents were less effective. Furthermore, all chelating agents produced no effect whatsoever on sorption of cesium since cesium does not practically form chelate compounds.

Thus, the tests described in this section once again demonstrate that the capacity of radionuclides for migration in the soil/solution system largely depends on the form in which they are present in the solution. Radionuclides forming stable chelate compounds are absorbed by the soil in smaller quantities and become more capable of migration. Thus, the effectiveness of chelating agents with respect to various radionuclides depends on the chelate stability constant.

As indicated below, radionuclides forming stable compounds with chelating agents also retain their high mobility (in the presence of a chelating agent) in a more sophisticated soil/plant system. In this context short-term experiments on sorption, in the simple soil/solution system, may suffice for tentatively assessing the effects of newly developed chelating agents on the mobility of chemical elements in the soil/plant system. The data obtained in such experiments may help to expedite the choice of the most promising chelating agents for further employment in work involving long-term and more labor-consuming greenhouse and field trials.

Effects of Extracts from Vegetative Litter on the Absorption of Radionuclides in the Soil

As a rule, the presence of organic matter in natural waters and soil solutions enhances the mobility of chemical elements in soils. The mobilizing action of organic matter dissolved in water is most diversified. It may manifest itself as a neutralizing effect of the negatively charged ash of organic matter on the positively charged ash of mineral colloids (Aarnio, 1915; Simakov, 1929). This mobilizing action may also be a peptizing effect of the water-soluble organic substances, an increase in acidity of the medium, or formation of simple and complex salts or chelates (Kravkov, 1934; Antipov-Karatayev, 1937; Rode, 1937, 1964;

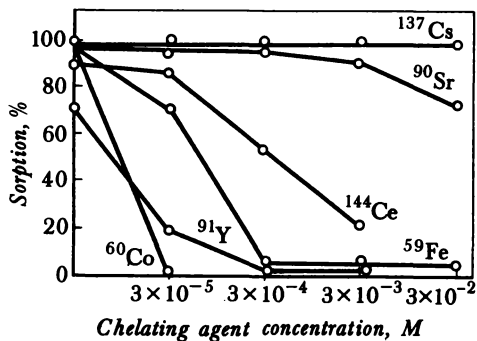


Fig. 4. Sorption of radionuclides by soil from aqueous solution with varying concentration of EDTA chelating agent.

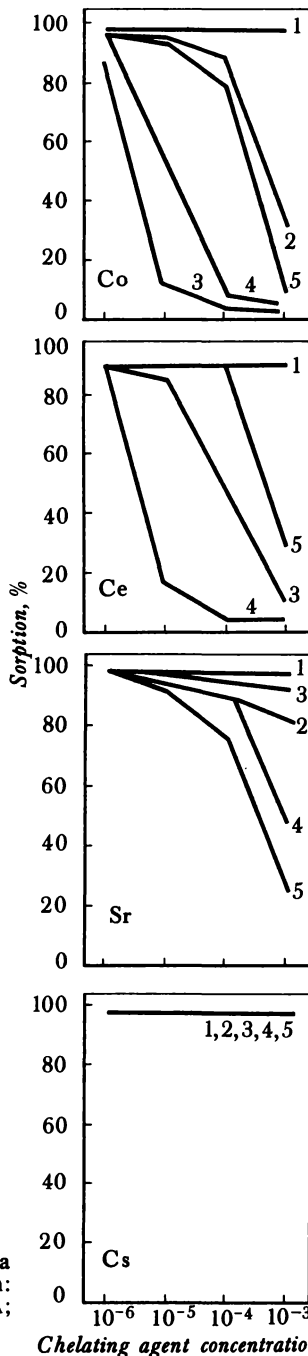


Fig. 5. Sorption of ⁶⁰Co, ⁹⁰Sr, ¹³⁷Cs, and ¹⁴⁴Ce as a function of concentration of chelating agents in solution: (1) control; (2) EADA; (3) EDTA; (4) DTPA; (5) EDBIPA.

Ponomareva, 1949, 1964; Aleksandrova, 1954a, b; Manskaya and Drozdova, 1964; Schnitzer and De Long, 1955; Aleksandrova and Nad, 1958; Bond and Jones, 1959; Nightingale and Smith, 1967).

Recently there has been developed a concept in geochemistry and pedology of the leading role of natural chelating agents, formed as a result of decay of vegetative litter, in the migration of a number of chemical elements in the hypergenesis zone. The main body of data pertaining to this issue concerns iron. It has been found that when aqueous extracts from various plants act on the soil an appreciable amount of iron goes into solution, and after reduction to the bivalent state becomes a component of the stable ferroorganic anion of a complex nature (Schnitzer and De Long, 1955; Bloomfield, 1953; Kaurichev, 1957; Kaurichev and Nozdrunova, 1961; Kaurichev *et al.*, 1960, 1964; Schnitzer, 1959). A number of authors noted the enhancement of mobility of cobalt, strontium, yttrium, and cerium in the soil under the influence of water-soluble products of plant decomposition (Shcherbina, 1956; Kee and Bloomfield, 1961; Volkova and Makhonina, 1962; Chuveleva *et al.*, 1962a; Volkova *et al.*, 1964; Timofeyev-Resovsky *et al.*, 1966; Chebotina, 1968; Molchanova and Kulikov, 1972; Chebotina and Kulikov, 1973). It has also been demonstrated that various groups of organic substances are not equally able to form chelate compounds (Lees, 1950; Mills, 1954; Schlichting, 1955; Wolff *et al.*, 1955; Beres and Kuraly, 1959; Kawaguchi and Kyuma, 1959; Aleksandrova, 1962; Drozdova and Yemelyanova, 1960; Kononova and Titova, 1961; Rydalevskaya and Tereshenkova, 1961; Dyakonova, 1962; Ponomareva, 1964; Randhawa and Broadbent, 1965; Tatsukawa and Franklin, 1966).

A particularly important role in complexing reactions is played by low-molecular organic acids, phenols, polyphenols, amino acids, aliphatic acids, and by a number of other metabolites of microorganisms, plants, and animals (Coulson *et al.*, 1960; Kaurichev and Nozdrunova, 1961; De Datta *et al.*, 1967). These substances are extracted by water from dying organisms or are formed anew in the process of decay of plant and animal remains. On entering natural waters and soil solution they are able to dissolve chemical elements present in the solid phase of the soil, and thereby to enhance the migration of these elements (Lutwich *et al.*, 1952; Atkinson and Wright, 1957; Himes and Barber, 1957; Himes and Shufeldt, 1969; Manskaya and Drozdova, 1959; Titlyanova *et al.*, 1959; Antipov-Karatayev and Tsurupa, 1961).

We carried out experiments designed to study the influence of aqueous extracts from the freshly fallen leaves of several ligneous plants on the sorption of radionuclides by soddy-meadow soil. The extracts were prepared by soaking 100 gm of fallen leaves in 5 liters of filtered lake water for 10 days at room temperature. To the extracts prepared in this manner there were added radionuclides of iron, cobalt, strontium, yttrium, cesium, and cerium; thereupon, in accordance with the technique described above, sorption of the radionuclides by the soil sample was followed up. For comparison parallel experiments were conducted to study the direct sorption by soil of radionuclides from the lake water and from the 0.01 M EDTA chelating agent solution.

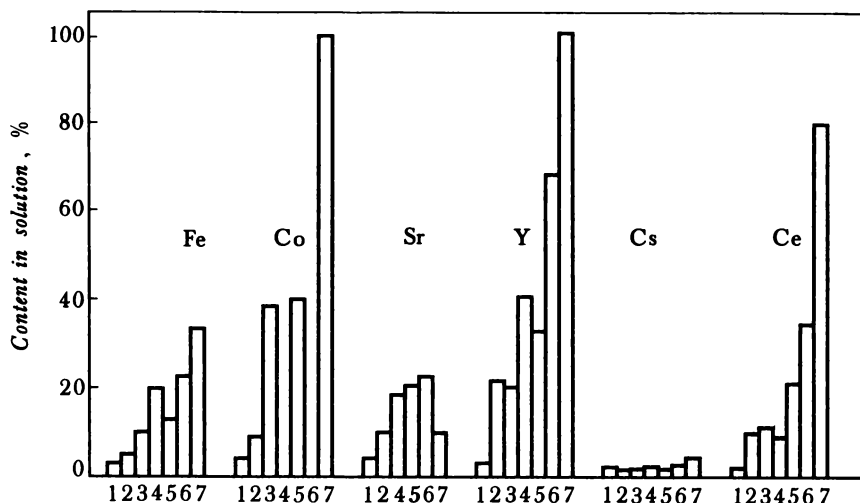


Fig. 6. Effect of plant extracts and EDTA solution on sorption of radionuclides by soddy-meadow soil: (1) lake water; (2) pine needles extract; (3) lime tree leaves extract; (4) birch tree leaves extract; (5) birdcherry leaves extract; (6) aspen leaves extract; (7) EDTA solution.

In the presence of plant extracts the sorption of radionuclides by the soil, except for ^{137}Cs , noticeably decreased (Fig. 6): 20 to 70% of the initial amount of the nuclide was retained as compared to 2 to 4% in experiments with lake water. In the presence of the EDTA chelating agent the sorption of radionuclides of iron, cobalt, yttrium, and cerium was maximally reduced; strontium was less affected and no effect whatsoever was observed with respect to cesium sorption. Of five plant extracts the pine needle extract had, on the average, the least desorbing effect, whereas the greatest effect was exhibited by aspen leaves, which is supposedly associated with a varying content of the desorbing agents in the extracts used (Table 7).

Table 7
Chemical characteristics of plant extracts

Extracts from leaves of:	pH	Dry residue	Content (mg/liter)		
			C	Ca ⁺⁺	Mg ⁺⁺
Aspen	4.9	9.75	3970	940	248
Birdcherry	4.9	8.75	3310	746	83
Birch	4.1	6.00	2660	196	224
Lime tree	4.2	7.90	3130	510	177
Pine	4.0	3.25	730	27	37

In order to find out in what measure the desorbing effect of extracts is linked with their contents of free cations and organic substances, a series of specific experiments was carried out (Chebotina, 1968). The experiments demonstrated that removal of organic substances from extracts almost completely eliminated the desorbing effect of the extracts on iron and, in a lesser degree, on cobalt. In tests with ^{90}Sr it was noted that the desorbing effect of extracts was principally due to the presence of calcium and magnesium cations in the extracts.

It thus follows that the desorbing effect of extracts is possibly related to their containing both organic matter capable of transforming individual chemical elements into soluble compounds and a mineral component.

The data presented in this section therefore show that water extracts prepared from vegetative litter reduce the soil sorption of iron, cobalt, strontium, yttrium, and cerium radionuclides, thereby enhancing the mobility of these isotopes in the soil/solution system. The desorbing effect of vegetative extracts for the group of radionuclides listed is due to both their organic and mineral components.

Stability of Nuclide Retention by the Soil

In the foregoing paragraphs we discussed factors influencing the extent of radionuclide absorption in soils. The mobility of radionuclides in the soil/solution system was assessed according to their distribution between the solid and liquid phases; the presumption was that the greater the amount of radionuclides retained in the solution, the greater its capacity for migration. However, the migration of chemical elements is defined not only by the level of their absorption, but also by the degree of their retention by soils. The firmer an element is retained by the soil and the less is the amount dissolved under the effect of various factors, the smaller is the area of the element dispersal. Thus, in order to more accurately estimate the capacity for migration of the radionuclides in question, one should have a knowledge of both the absorption rate of radionuclides and the stability of their fixation in soils.

While the absorption of radionuclides in soils, as was demonstrated in the foregoing section, can be estimated from the data on the extent of their transfer from the liquid to the solid phase (solution/soil), their retention by the soil can be evaluated by the reverse process, the transfer from the solid to the liquid phase (soil/solution), i.e., according to the extent of their desorption from the soil under the effect of various desorbing agents.

In this connection, our laboratory carried out an extensive series of experiments for investigating the desorption from soils of ^{59}Fe , ^{60}Co , ^{90}Sr , ^{91}Y , ^{137}Cs , and ^{144}Ce . We studied the dependence of radionuclide desorption on the pH of the desorbing solution, on the presence in the solution of different cations, artificial chelating agents, and water-soluble organic matter derived from the vegetative litter, and on soil moisture. In studying this dependence each factor was considered separately.

Influence of pH of the Desorbing Solution

Apart from the direct dissolution of chemical compounds, pH-dependent changes in the desorbing effect of a solution are also produced by such phenomena as the leaching of particular chemical elements from the soil into a solution as a result of absorption of hydrogen ions by the soil. Moreover, changes in the acid-base balance of the medium result in conversion of the ionic form of an element to the colloidal form, which, in turn, may affect the retention of the element by the soil.

Table 8 shows data on the desorption of radionuclides from soddy-meadow soil by lake water with pH ranging from 3 to 10. Radionuclides were sorbed from a solution at pH 4. The desorbing effect of water was found to be similar with regard to all radionuclides, negligible, and not depending on the pH value. This indicated a fairly strong retention by the soil of the radionuclides under study. The retention, in fact, did not depend on whether the nuclides had the ionic or the colloidal form in the initial solution (Table 9). Certain reduction of desorption of cobalt was noted in the range of solution pH in which this element must be present in the colloidal form.

Thus, changes in pH of the solution have no appreciable effect on the retention by the soil of the radionuclides under study.

The Desorbing Effect of Different Cations

In the intricate complex of physicochemical processes in the soil ion-exchange reactions play an important role. It has been found that the ability of cations, participating in these reactions, to replace other cations increases with their atomic weight and valency (Gedroits, 1935; Ginzburg and Ponomareva, 1940). It is also known that K^+ , Na^+ , and Ca^{++} , representing the basic macroelements, participate predominantly in ion-exchange reactions in the soil, whereas complexing reactions are more characteristic of such elements as zinc, copper, aluminum, and iron. In view of this it was of interest to study the desorbing effect of the cations listed for the purpose of detecting some mechanisms involved in the fixation in soil of the radionuclides under study.

Most firmly retained by the soil were ^{59}Fe and ^{137}Cs , since the desorbing effect produced on these nuclides by all cations was negligible (Table 10). ^{60}Co was desorbed relatively quickly by bi- and trivalent cations. In accordance with their capacity to replace this radionuclide in the soil, these cations may be arranged in the following sequence: $Na^+ < K^+ < Ca^{++} < Zn^{++} < Al^{+++} < Fe^{++} < Cu^{++}$. One can note that the replacing capacity of these cations agrees fairly closely with that observed in ion-exchange reactions. Consequently, the binding of cobalt in the soil follows to a great extent the ion-exchange pattern, which was also suggested by other investigators (Spencer and Giesekeing, 1954; Titlyanova and Timofeyeva, 1962; Chebotina and Titlyanova, 1965). The behavior of ^{90}Sr was similar, though it was not so tightly bound in the soil as cobalt. ^{91}Y and ^{144}Ce , though more tightly bound, were nevertheless desorbed to a rather significant degree by some

Table 8**Desorption of radionuclides from soddy-meadow soil by lake water with**

Nuclide	pH		
	3	4	5
⁵⁹ Fe	3.2±0.7	2.5±0.5	2.9±0.4
⁶⁰ Co	3.5	1.7	2.0
⁹⁰ Sr	2.0±0.1	1.8±0.3	2.5±0.3
⁹¹ Y	1.2±0.1	1.5±0.2	1.7±0.4
¹³⁷ Cs	0.5±0.05	0.7±0.02	0.4±0.01
¹⁴⁴ Ce	2.3±0.4	2.7±0.8	1.9±0.8

Table 9**Desorption of radionuclides from soddy-podzolic soil by aluminum and zinc cations (percent of amount sorbed)**

pH of initial solution	⁵⁹ Fe	⁶⁰ Co	⁹¹ Y	¹⁴⁴ Ce
	Desorbing cation			
	Al ⁺⁺⁺	Zn ⁺⁺	Al ⁺⁺⁺	Al ⁺⁺⁺
3	5.0	42.0	9.0	3.0
4	2.0	42.0	8.0	4.0
5	8.0	24.0	9.0	3.0
6	7.0	36.0	7.0	5.0
7	4.0	30.0	7.0	5.0
8	5.0	42.0	8.0	4.0
9	5.0	16.0	8.0	4.0
10	4.0	18.0	7.0	4.0

Table 10**Desorption of radionuclides from soddy-meadow soil by 0.1 N salt solutions (percent of amount sorbed in soil)**

Nuclide desorbed	Desorbing cation						
	Na ⁺	K ⁺	Ca ⁺⁺	Cu ⁺⁺	Zn ⁺⁺	Al ⁺⁺⁺	Fe ⁺⁺⁺
⁵⁹ Fe	1.2	2.0	2.0	4.6	1.8	5.0	—
⁶⁰ Co	3.7	6.0	18.1	86.0	74.2	76.0	79.2
⁹⁰ Sr	18.9	25.0	70.3	—	—	90.0	—
⁹¹ Y	1.0	1.3	4.4	41.6	6.3	8.0	45.0
¹³⁷ Cs	1.2	7.0	1.5	—	—	1.5	—
¹⁴⁴ Ce	2.6	2.6	4.8	7.7	3.2	4.8	22.6

differing pH values (percent of amount sorbed)

of the desorbing solution				
6	7	8	9	10
3.1 ± 0.8	2.8 ± 0.7	2.5 ± 0.8	2.7 ± 0.5	2.7 ± 0.5
2.3	1.5	1.5	1.7	1.9
2.0 ± 0.3	1.7 ± 0.4	1.9 ± 0.2	2.2 ± 0.4	2.5 ± 0.3
2.0 ± 0.3	2.0 ± 0.5	1.8 ± 0.6	1.6 ± 0.4	2.0 ± 0.3
0.5 ± 0.05	0.4 ± 0.02	0.3 ± 0.05	0.3 ± 0.04	0.5 ± 0.09
2.4 ± 0.9	1.9 ± 0.8	1.5 ± 0.6	1.8 ± 0.5	1.9 ± 0.5

cations. Such desorbing cations were iron and copper for ^{91}Y and iron for ^{144}Ce .

The data in Fig. 7 on the desorption by salt solutions of ^{59}Fe , ^{91}Y , and ^{144}Ce derived from differing soils demonstrate that the level of retention of radionuclides by these soils is not uniform. They are particularly firmly retained by the soddy-meadow and chernozem soils, ^{59}Fe being the most firmly fixed of the three.

On the whole, the desorbing effect of bi- and trivalent cations in these experiments was stronger than that of sodium and potassium. Among these desorbing cations there may be distinguished a "specific replacer," i.e., a cation that replaces a particular radionuclide to a greater extent than it does other nuclides. In almost all soils the specific replacer for ^{59}Fe was aluminum, for ^{91}Y the replacers were copper and iron, and for ^{144}Ce the replacer was iron. The existence of "specific replacers" may indicate selectivity of soil absorption of the given radionuclides; it shows that ^{59}Fe forms in the soil compounds that are more characteristic of aluminum than of other cations, whereas ^{91}Y and ^{144}Ce form compounds characteristic of copper and iron, respectively.

Thus, the low level of desorption of the iron, yttrium, cesium, and cerium radionuclides by 0.1 N solutions of NaCl , KHO_3 , and $\text{Ca}(\text{NO}_3)_2$ salts, as well as lack of any dependence of the desorbing effect of the cations on their atomic weight and valency, indicate that trace amounts of these radionuclides are virtually uninvolved in ion-exchange reactions. They participate in reactions and form compounds of some other type from which they can be displaced only by some particular cations. Formation of such compounds is also characteristic of cobalt, although to a lesser extent (Kevin *et al.*, 1963).

We have carried out auxiliary experiments that have demonstrated that the removal of organic matter from the soil through soil treatment with hydrogen peroxide by the technique of Zhuravleva (1965) results in a markedly weaker fixation of the iron, yttrium, and cerium radionuclides (Fig. 8). Tests with iron in soil samples treated with H_2O_2 have shown that the desorbing effect of aluminum increases greatly.

The role of aluminum, as the specific replacer of iron in native soils and in soils free of organic matter consists in the isomorphic replacement of iron in crystalline structures of clay minerals. In tests with ^{91}Y and ^{144}Ce the desorbing effect of cations in humus-free soil increases in the order $\text{Na}^+ > \text{K}^+ > \text{Ca}^{++} > \text{Zn}^{++} > \text{Cu}^{++} > \text{Al}^{+++} > \text{Fe}^{+++}$. This sequence, excluding the shift between zinc and copper, corresponds to the enhancement of the replacing capacity of cations in ion-exchange reactions. Consequently, the absorption of a considerable amount of ^{91}Y and ^{144}Ce by the mineral fraction of the soil follows the pattern of ion-exchange reactions (the exchange is speedy and in equal cation quantities).

According to some investigators (Schulz *et al.*, 1960; Timofeyev-Resovsky *et al.*, 1966), retention of individual chemical elements depends on their concentration in the soil. The tests carried out with iron, yttrium, and cerium demonstrated that desorption of these elements, expressed in absolute values, changed proportionally to

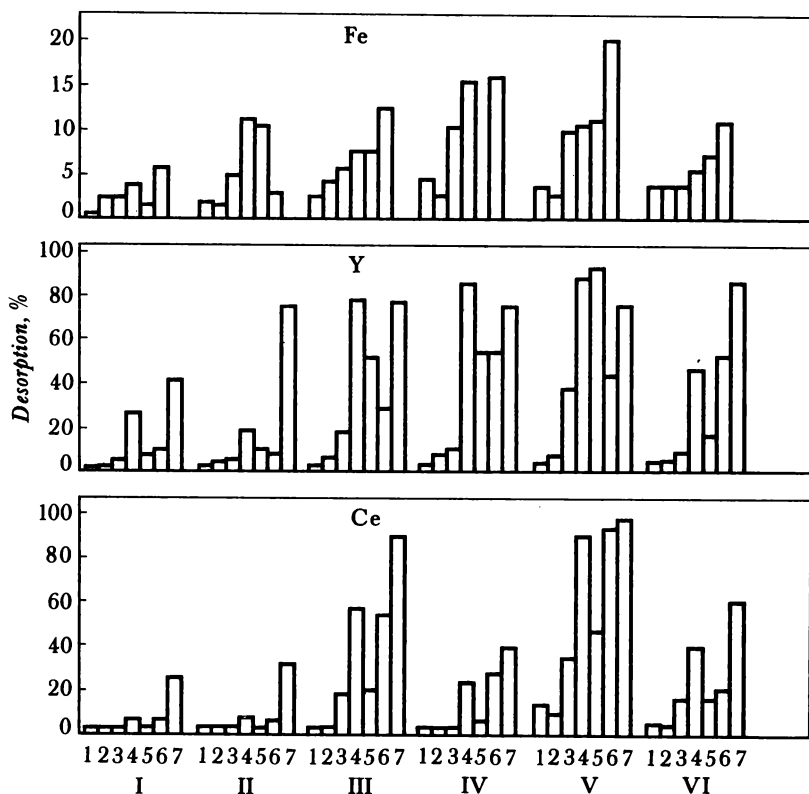


Fig. 7. Desorption of ^{59}Fe , ^{91}Y , and ^{144}Ce by 0.1 N solutions of salts. Soil: (I) soddy-meadow; (II) chernozem; (III) soddy-podzolic, horizon A₁; (IV) soddy-podzolic, horizon A₂; (V) soddy-podzolic, horizon B₁; (VI) red soil. Desorbent: (1) NaCl; (2) KNO₃; (3) Ca(NO₃); (4) CuSO₄; (5) ZnSO₄; (6) Al₂(SO₄)₃; (7) FeCl₂.

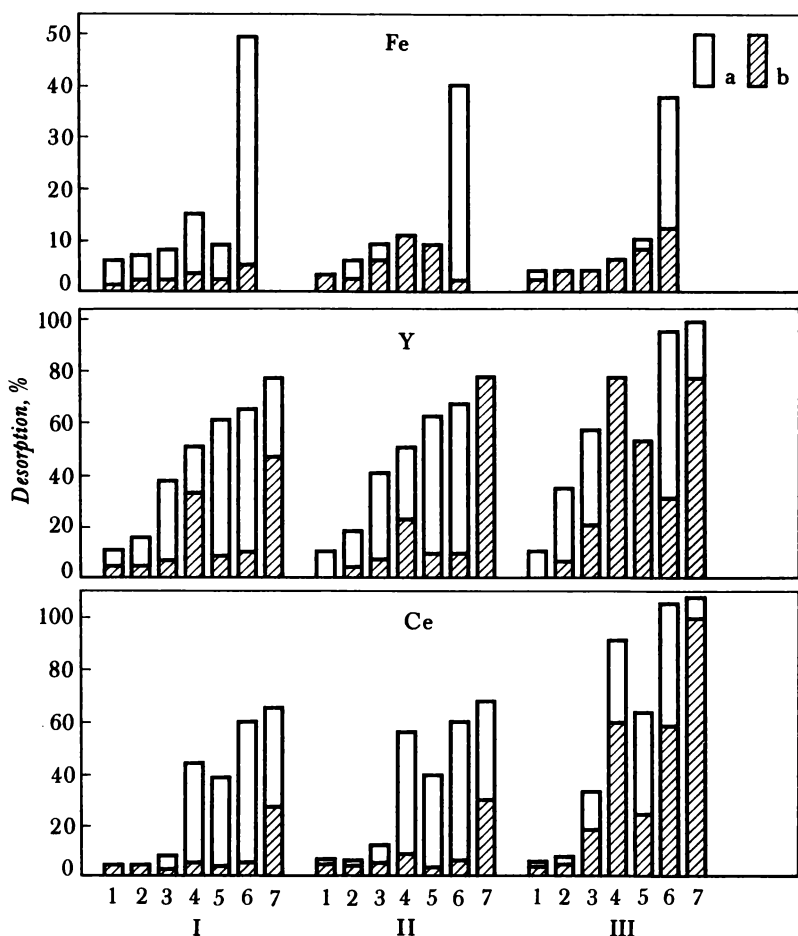


Fig. 8. Desorption of ^{59}Fe , ^{91}Y , and ^{144}Ce from native soils (a) and soils free of organic matter (b) by 0.1 N salt solutions. Soil: (I) soddy-meadow; (II) chernozem; (III) soddy-podzolic, horizon A₁. Desorbent: (1) NaCl; (2) KNO₃; (3) Ca(NO₃)₂; (4) CuSO₄; (5) ZnSO₄; (6) Al₂(SO₄)₃; (7) FeCl₃.

their concentration in the soil. This shows that the degree of retention of these elements by soddy-meadow soil (as judged by the relative value of desorption in percent) remains constant over a broad range of microconcentrations shown in Table 11. The observed decline in the percentage of cerium and yttrium desorption at a concentration of 10^{-6} moles per gram of soil, and that of iron at a concentration of 10^{-7} moles per gram of soil, may be due to the formation at these concentrations of large colloidal aggregates in which cerium and yttrium are replaced by other cations with greater difficulty than in small colloids.

Basically similar results were also obtained in experiments with ^{60}Co

Table 11

Desorption of iron, yttrium, and cerium by 0.1 N $\text{Al}_2(\text{SO}_4)_3$ solution at differing concentrations of elements in the soil (percent of amount sorbed)

Element	Concentration (moles/gm soil)					
	10^{-11}	10^{-10}	10^{-9}	10^{-8}	10^{-7}	10^{-6}
Iron	—	—	5.2 ± 0.7	6.3 ± 0.5	2.1 ± 0.4	—
Yttrium	7.8 ± 0.2	8.7 ± 0.3	8.5 ± 0.5	8.3 ± 0.4	10.3 ± 0.8	6.5 ± 0.2
Cerium	—	10.2 ± 0.4	9.3 ± 0.6	11.6 ± 0.5	9.8 ± 0.4	4.8 ± 0.5

Table 12

Desorption of cobalt by 0.05 N ZnSO_4 solution at differing cobalt concentrations in the soil (percent of amount sorbed)

pH of initial solution	Concentration (moles/gm soil)			
	2×10^{-8}	2×10^{-7}	2×10^{-6}	2×10^{-5}
2	42.5	41.5	42.3	43.0
3	—	38.0	35.2	43.2
4	42.5	31.2	34.4	41.0
5	24.0	32.0	32.2	36.4
6	36.2	33.5	32.9	39.6
7	30.6	28.4	34.2	37.2
8	42.7	29.8	32.3	40.0
9	16.8	38.4	34.2	—
10	18.5	47.3	27.8	—

(Table 12). Regardless of the pH of the initial solution, the percentage of desorption actually remained invariable when the concentration of cobalt in the soil varied over a range of several orders of magnitude (10^{-8} to 10^{-5} moles per gram). Identical results were also obtained in tests with ^{90}Sr and ^{137}Cs (Timofeyev-Resovsky *et al.*, 1966).

Thus, results of diversified tests of radionuclide desorption by different cations make it possible to consider the soil absorption of iron, yttrium, cesium, and cerium as a low reversibility process, determined by organic matter and the mineral fraction of the soil. For ^{60}Co and ^{90}Sr ion-exchange reactions are more characteristic.

In evaluating the desorbing effect of cations with regard to its importance in the migration of radionuclides in soils, it should be noted that in the natural water and soil solutions the concentrations of the chemical elements we employed as desorbents are several orders of magnitude lower than the concentrations used experimentally. It thus follows that under natural conditions the desorbing effect of cations must be much lower.

The Desorbing Effect of Aqueous Extracts from Vegetative Litter and EDTA Chelating Agent

As demonstrated above, aqueous extracts from plants and artificial chelating agents reduce the absorption of radionuclides by soil and thereby increase the mobility of these nuclides in the soil/solution system. We shall now review the desorbing effect of such extracts and chelating agents, i.e., their ability to transfer radionuclides from the solid to the liquid phase of the soil.

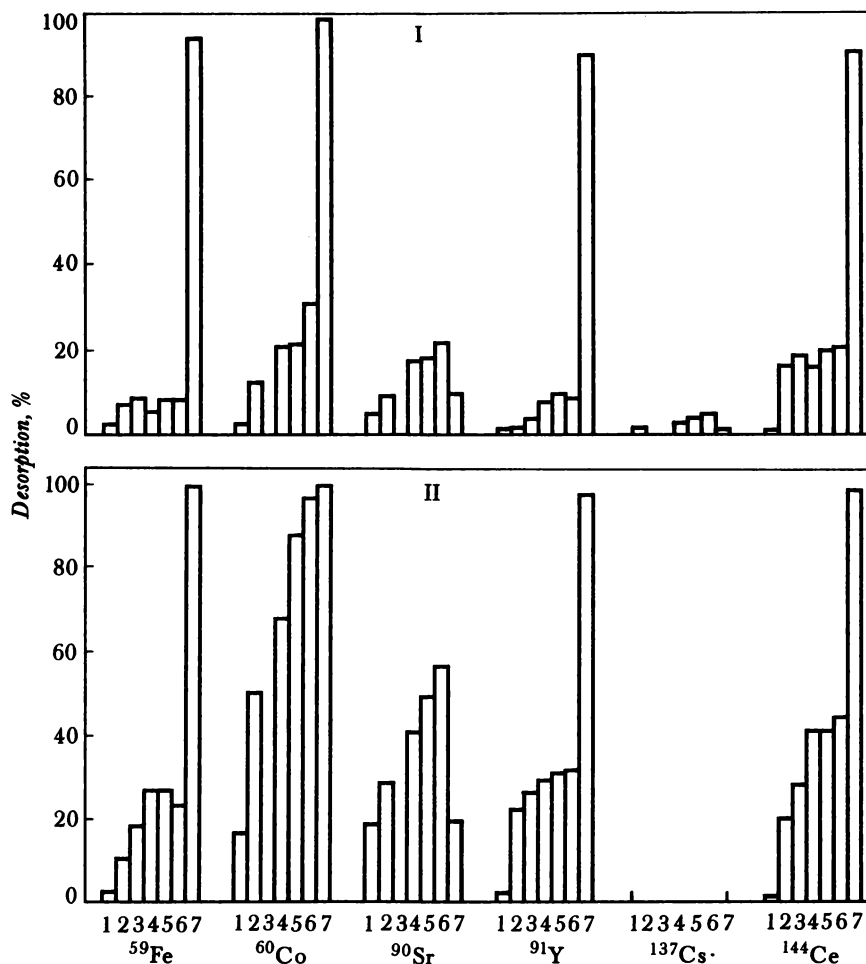


Fig. 9. Desorption of radionuclides from soddy-meadow (I) and soddy-podzolic, horizon A₂ (II) soils by different desorbents. Desorbent: (1) lake water; (2) pine needles extract; (3) lime tree leaves extract; (4) birch tree leaves extract; (5) birdcherry leaves extract; (6) aspen leaves extract; (7) 0.1 M Na/EDTA solution.

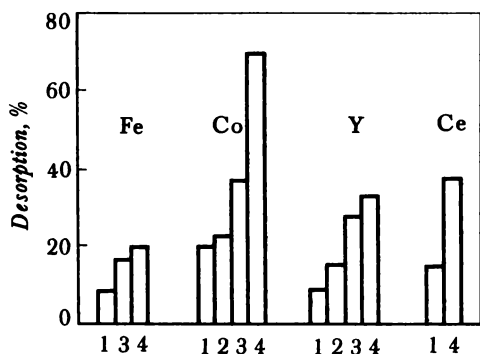


Fig. 10. Desorption of ^{59}Fe , ^{60}Co , ^{91}Y , and ^{144}Ce from various soils by plant extracts (mean values for extracts from pine needles, aspen leaves, lime tree leaves, birch tree leaves, and birdcherry leaves). Soil: (1) soddy-meadow; (2) chernozem; (3) red soil; (4) soddy-podzolic, horizon A_2 .

The strongest desorbent for most of the nuclides studied is EDTA (Fig. 9). This chelating agent carries the absorbed amounts of ^{59}Fe , ^{60}Co , ^{91}Y , and ^{144}Ce into the liquid phase practically fully, irrespective of soil properties. With ^{90}Sr the desorbing effect of EDTA was lower, whereas with ^{137}Cs EDTA proved to be ineffective. One may note that the desorbing action of all plant extracts is more evident in the soddy-podzolic soil. It is particularly vivid in experiments with ^{60}Co , which is desorbed from soddy-podzolic soil nearly as effectively by plant extracts as by EDTA.

The analysis of results concerning desorption from the soil of the four radionuclides belonging to the group of typical complexing substances demonstrates that the desorbing effect of plant extracts depends inversely on the degree of retention of radionuclides in the soil (Fig. 10). Thus, in all the soils under study ^{59}Fe was retained more firmly than ^{60}Co , ^{91}Y and ^{144}Ce , and the desorbing effect of extracts on ^{59}Fe was the weakest. Since the retention is determined not only by the properties of chemical elements but also by the physicochemical properties of the soil, the desorption of the radionuclides by the extracts is the greatest in the soddy-podzolic soil, in which they are retained less firmly.

Influence of the Soil Moistening Regime on the Transfer of Radionuclides from Soil to Solutions

All of the experiments described above, concerned with radionuclide mobility in the soil/solution system, as well as many similar studies by other investigators (Kokotov *et al.*, 1962; Timofeyev-Resovsky *et al.*, 1966; Yudinseva and Gulyakin, 1968) were carried out with the use of soil suspensions, i.e., under conditions in which the amount of the desorbing solution greatly exceeded the amount of the solid phase. The results of such experiments, whose technique is simple and easy to perform, permit to form an opinion about the basic physicochemical mechanisms involved in the migration of radionuclides in soils that contain aqueous solutions. However, during the last few years data have been accumulated indicating that the quantitative parameters of the

passage of some radionuclides into solution in moist soils are different from those in soil suspensions (Prokhorov and Frid, 1969; Molchanova and Karavayeva, 1971; Molchanova and Kulikov, 1972). It has also been shown that under natural conditions and in the appropriate soil/climate zones, the aqueous factor may play an important role in the redistribution of radionuclides in soils (Polyakov, 1966; Molchanova and Kulikov, 1970c).

We have performed a series of experiments to compare the distribution of cobalt, strontium, cesium, and cerium radionuclides in the soil/solution system when the ratio of the solid to the liquid phase of the soil changes over a wide range. For this purpose, specific samples of soddy-meadow soil were mixed with chlorous compounds ($^{60}\text{CoCl}_2$; $^{90}\text{SrCl}_2$; $^{137}\text{CsCl}$; $^{144}\text{CeCl}_3$) of the respective radionuclides, then air-dried, and moistened with distilled water in such a way that the ratio of water volume (milliliters) to the soil weight (grams) in different variations of the experiment would be 0.1, 0.2, 0.3, 0.5, 1.0, 2.0, 5.0, 10.0, and 20.0. These values will be referred to below as moisture coefficients (MC).

The moistened soil samples with MC's less than 1.0 were placed in tightly stoppered sample vessels and allowed to stand in dessicators with water for 20 days, while soil samples with MC's equal to or greater than 1.0 (suspensions) were put in small closed plexiglass cups and stirred in an electric mixer for 2 hrs. As demonstrated by check tests, during this period an equilibrium distribution of all the radionuclides in question was attained between the liquid and solid phases of the soil, regardless of the moistening regime we adopted. On attaining the equilibrium distribution of radionuclides in the soil/solution system, the soil samples employed in all variations of the experiment were centrifuged at 5000 revolutions per minute in a LJC-3 centrifuge. In the soil solution thus derived the content of radionuclides was measured with the aid of a YMΦ-1500 radionuclide counter fitted with a CBT-13 counting tube, the count statistical error being 3 to 5%. All experiments were performed in six replications.

With an increase in soil moisture the total content of all radionuclides in the liquid phase rises (Table 13). At the extremal values of soil moisture the differences in the content of radionuclide solutes reach about one order of magnitude for ^{60}Co , and two and three orders of magnitude for ^{144}Ce and ^{137}Cs , respectively, whereas the content of ^{90}Sr changes only by a factor of three to four. Then, if one would evaluate the mobility of the radionuclides in soil according to their content in the liquid phase, the mobility of ^{90}Sr in the moist soil should be considered as exceeding that of the other radionuclides by several orders of magnitude. With respect to soil suspensions, however, these differences become less marked. As a result, the mobilities of ^{60}Co , ^{90}Sr , and ^{137}Cs appear roughly similar and only ^{144}Ce remains somewhat less mobile under these conditions (Fig. 11).

The mobility of radionuclides in the soil/solution system is sometimes assessed in accordance with the distribution coefficient, which is the ratio of radionuclide concentrations in the soil to those in the

Table 13

Radionuclide content in solution (percent of amount introduced into soil)

Coefficients of soil moisture	⁶⁰ Co		⁹⁰ Sr	
	Content (%)	DC	Content (%)	DC
0.1	0.13	75 ± 5	0.40	26 ± 3
0.2	0.16	125 ± 13	0.50	36 ± 1
0.3	0.12	250 ± 15	0.30	106 ± 9
0.5	0.12	400 ± 15	0.20	229 ± 8
1.0	0.24	416 ± 11	0.40	224 ± 6
2.0	0.44	455 ± 17	0.60	335 ± 5
5.0	0.63	670 ± 35	1.00	517 ± 18
10.0	0.85	1 188 ± 60	1.70	600 ± 69
20.0	1.20	1 635 ± 120	1.40	1 420 ± 33

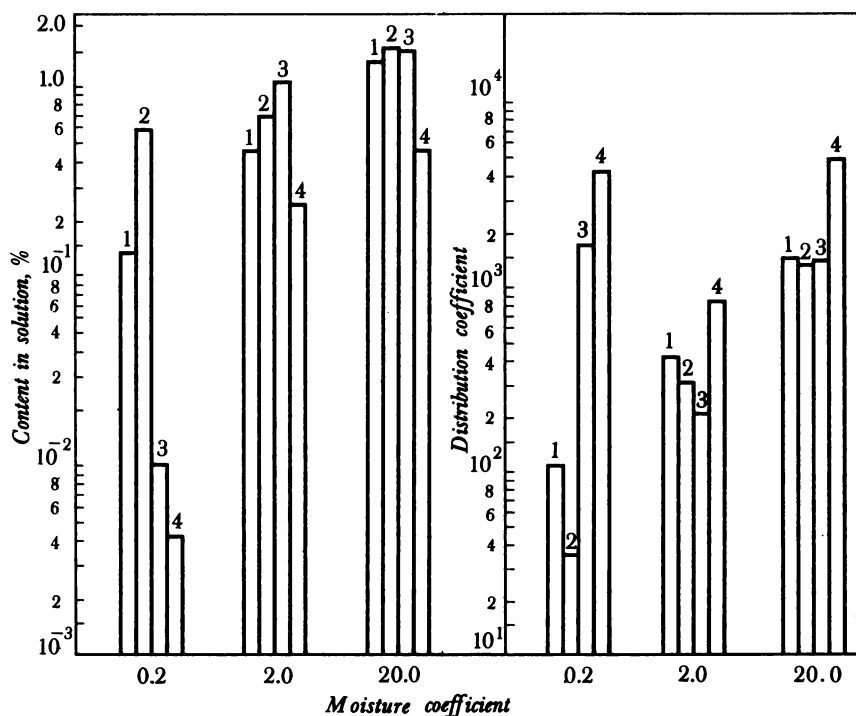
Fig. 11 (left). Content of radionuclides in solution as a function of soil moisture (% of the overall content in the soil/solution system). (1) ⁶⁰Co; (2) ⁹⁰Sr; (3) ¹³⁷Cs; (4) ¹⁴⁴Ce.

Fig. 12. Coefficients of distribution of radionuclides as a function of soil moisture. Same designations as in Fig. 11.

and distribution coefficients (DC)

¹³⁷ Cs		¹⁴⁴ Ce	
Content (%)	DC	Content (%)	DC
0.007	1 450 ± 140	0.003	3 175 ± 500
0.011	1 790 ± 180	0.004	4 535 ± 450
0.020	1 515 ± 120	0.007	4 330 ± 400
0.130	420 ± 30	0.040	1 305 ± 130
0.430	240 ± 10	0.100	980 ± 100
0.910	228 ± 24	0.220	925 ± 90
1.250	417 ± 35	0.280	1 810 ± 180
1.250	830 ± 60	0.450	2 230 ± 200
1.370	1 515 ± 120	0.400	5 195 ± 500

equilibrium solution (Molchanova and Karavayeva, 1971; Chebotina and Kulikov, 1973). The radionuclide mobility is then inversely dependent on the distribution coefficient.

In moist soil the lowest distribution coefficient is characteristic of strontium and cobalt, while the highest coefficient is characteristic of cesium and cerium (Fig. 12, Table 13). In soil suspensions, particularly in diluted ones, all values of the distribution coefficient are within one order of magnitude and the relative radionuclide mobility is, thus, considerably leveled. The data presented also show that with equal values of the distribution coefficient the total content of some radionuclides (cesium) in the liquid phase may vary by several orders of magnitude depending on the extent of soil moisture. Therefore, the distribution coefficient consistently reflects the relative mobility of different radionuclides only under conditions of uniform soil moisture. Under varying conditions of soil moistening it is more expedient to express the radionuclide mobility in the soil/solution system according to the part of the total radionuclide content in the soil that is retained in the liquid phase. In the final analysis, it is this particular fraction of any radionuclide which is most likely to migrate outside the limits of the originally contaminated area of the soil; it is also this fraction that is primarily taken up by the root system of plants.

Considering the relatively high mobility of ⁹⁰Sr in the soil/solution system at a low moistening level, one would expect that in natural soils this radionuclide will be more mobile than the others. However, the available literature data show that for radionuclides compared the vertical migration parameters in the soils of natural biogeocenoses are sometimes very similar (Makhonina *et al.*, 1965; Makhonko and Chumichev, 1969; Kulikov and Piskunov, 1970). The results presented in this section suggest that seasonal variations in soil moisture under natural conditions may serve as a factor for leveling the rate of radionuclide migration in soils.

Chapter 2

MIGRATION AND DISTRIBUTION OF RADIONUCLIDES IN THE SOIL/PLANT SYSTEM

In the preceding chapter ample data were considered characterizing the mobility of radioactive iron, cobalt, strontium, yttrium, cesium, and cerium in the soil/solution system in relation to a number of factors. Comparative analysis of the behavior of these nuclides in the soil/solution system is a more difficult task, since the literature data dealing with the uptake from the soil of nuclides by plants are generally incomparable. In view of this, when presenting the material of this chapter, we shall mainly draw on the studies carried out in our laboratory employing comparable experimental conditions for all radionuclides.

The experiments were conducted in a greenhouse located on soddy-meadow and soddy-podzolic soils. Samples of soil were irrigated with solutions of chlorous compounds of the radionuclides studied, then thoroughly mixed and placed into culture vessels. The radioactivity of radionuclides in the soil in varying experiments ranged from 0.1 to 0.4 mCi/kg; the weight of the air-dried soil in each vessel was 2 kg; each experiment was performed in duplicate; in culture vessels prepared for the experiment, seeds of pea (*Pisum sativum* L.) and barley (*Hordeum vulgare* L.) were planted; at day 30 after sprouting the plants were taken out of the vessels, brought to absolute dry weight, and ashed at 400-500°C. The radioactivity of the preparations was then measured radiometrically. In experiments designed to study the accumulation of radionuclides by plants as dependent on the soil moistening regime, a soil mixture with washed quartz sand was used to enhance soil aeration and moisture permeability. The soddy-meadow soil was mixed with sand in a volumetric ratio of 1:1 and the soddy-podzolic soil in a ratio of 2:1. The moisture of the soil mixture was maintained throughout the experiments within the following limits: (1) from the maximum to the minimum water capacity; (2) from the minimum water capacity to capillary-rupture moistness; and (3) from capillary-rupture moistness to the moistness of withering. The choice of these moisture ranges was imposed by the fact that in transition from one range to the next the water transport in all types of soil changes sharply, as does the accessibility of water to plants (Rode, 1937). In our experiments the ratio of water volume to soil weight (moisture coefficient) was about 0.3, 0.2, and 0.1, respectively, for the mean moisture of each of the ranges. The level of moisture was controlled by weighing.

It should be noted that the amounts of radionuclides used in these particular experiments produced no perceptible effect on the growth and development of plants.

Accumulation of Radionuclides by Plants in Relation to Soil Properties

As pointed out in Chapter 1, the degree of radionuclide retention depends on the physicochemical properties of the soil. Particularly distinct differences in the retention of radionuclides are recorded between the soddy-podzolic and soddy-meadow soils. Based on the data obtained in our experiments and evidence from the literature (Kokotov and Popova, 1962; Graham and Killion, 1962; Kvaratskhelia and Arnautov, 1964; Nishita *et al.*, 1956; Yudintseva *et al.*, 1969), one might expect plants to take up a smaller amount of radionuclides from the soddy-meadow soil than from the soddy-podzolic soil, inasmuch as radionuclides are fixed more firmly in the former than in the latter.

Greenhouse experiments conducted by us with two species of plants fully support this assumption (Table 14). The uptake by plants of all radionuclides from the soddy-podzolic soil did exceed considerably the nuclide uptake from the soddy-meadow soil. One should note, however, that in the superterranean plant mass the accumulation coefficients for ^{90}Sr and ^{59}Fe are higher than those for the other radionuclides. This is explained both by a greater biotropism of the said radionuclides and by their greater mobility in the soil/solution system.

A lower radionuclide uptake by plants from the soddy-meadow soil is possibly associated with a higher content in this soil of organic matter and a greater silt fraction (Gulyakin and Yudintseva, 1962; Yudintseva and Gulyakin, 1968). The validity of this is well supported by the results of greenhouse experiments on ^{60}Co that we carried out using soils with varying amounts of humus (Kulikov, 1961c).

Table 14

**Coefficients of radionuclide accumulation in superterranean plant mass
on soddy-meadow and soddy-podzolic soils***

Plant	^{59}Fe	^{60}Co	^{90}Sr	^{91}Y	^{137}Cs	^{144}Ce
Soddy-meadow soil						
Pea	0.30	0.03	2.35	0.08	—	0.02
Barley	0.30	0.08	0.86	0.08	0.06	0.02
Soddy-podzolic soil						
Pea	0.48	—	6.80	0.80	0.36	0.51
Barley	0.42	0.20	3.67	0.23	0.14	0.20

* Accumulation coefficient equals the ratio of radionuclide concentrations in plants and soil as calculated on the basis of dry mass.

Table 15

Dry weight (gm), radioactivity of dry matter (counts/min) and total radioactivity (percent of amount introduced) of main portions of vetch and oats plants in relation to humus content in soil

Index	Nonradio- active control	Humus content in soil (%)			
		3.4	5.1	13.1	16.9
Oats					
Dry weight:					
Superterranean mass	40.5	48.7	59.1	52.1	170.5
Roots	5.8	5.0	5.0	5.8	17.3
Radioactivity(per gram):					
Superterranean mass	—	20	20	13	8
Roots	—	7250	2480	1930	290
Total radioactivity:					
Superterranean mass	—	0.011	0.012	0.007	0.012
Roots	—	0.400	0.160	0.120	0.056
Vetch					
Dry weight:					
Superterranean mass	43.5	57.8	54.5	60.8	85.3
Roots	5.9	4.4	4.1	5.8	6.7
Radioactivity(per gram):					
Superterranean mass	—	180	160	90	18
Roots	—	4000	3200	1800	450
Total radioactivity:					
Superterranean mass	—	0.110	0.098	0.061	0.017
Roots	—	0.200	0.150	0.110	0.033

In these experiments the soddy-meadow soil was mixed with sand or muck so that its humic content reached 3.4% (variant 1), 5.1% (variant 2), 13.1% (variant 3), or 16.9% (variant 4). To these mixtures CoCl_2 solution was added at the rate calculated to yield the radioactive and stable cobalt concentrations of 50 $\mu\text{Ci/kg}$ mixture and 0.38 mg/kg mixture, respectively. The soil with the radionuclide solution was placed in sowing boxes, each holding 8 kg of the material (in triplicate), in which vetch and oat seeds were planted. Visual observations of plant growth and development were carried out and the radioactivity of plant samples was measured at days 10, 20, and 50 after sprouting and upon completion of the experiment.

The concentration of ^{60}Co in plants, and especially in their roots, decreased consistently as the humus content in the soil increased; the total amount of cobalt accumulated by the above-ground mass of plants (expressed in percentage of the amount of cobalt introduced into the soil) depended not only on the humus content of the soil, but also on the specific biological features of plants (Table 15). Thus, for oats the

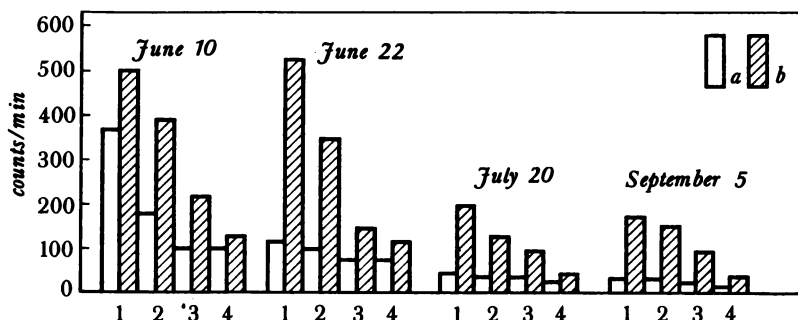


Fig. 13. Concentration of ^{60}Co in superterranean mass of oats (a) and vetch (b) in soils with varying humus content: (1) 3.4%; (2) 5.1%; (3) 13.1%; (4) 16.9%.

amount of cobalt accumulated by the superterranean plant mass under different experimental conditions made up thousandth and hundredth fractions of one percent, whereas for vetch it was hundredth and tenth fractions of one percent.

Especially vivid were the differences in accumulation of ^{60}Co between the two species of plant and between the variants of the experimental condition (Fig. 13). In addition, one can see that by the end of the vegetation season cobalt concentration in a plant was markedly reduced.

On the whole, the results of the experiments described bear out that radionuclide uptake by plants depends both on the physicochemical properties of the soil and on the biological features of plants. The radionuclides more firmly retained by the soil are accumulated by plants to a lesser extent than those retained less firmly.

Accumulation of Radionuclides in Relation to Soil Moisture

The data available from the literature concerning the role of moisture in the migration of radionuclides in the soil/plant system are rather conflicting. In a number of works there is noted a rise in the uptake of individual radionuclides by plants as the soil moisture increases (Tsun Tien Chao *et al.*, 1962; Emmert, 1963; Kubota *et al.*, 1963; Sutcliffe, 1962; Pavlotskaya *et al.*, 1966). Other authors point to the nonexistence of such a dependence (Schulz *et al.*, 1959; Nishita and Hawthorne, 1967). The differences in the available data can be attributed mainly to an insufficiency of information on the problem. The difficulties are probably due to the fact that the uptake of chemical elements from the soil by plants varies with the regime of soil moistening. In addition, the influence of moisture on the mobility of radionuclides in the soil/plant system is directly related to the soil properties and to distinctive biological features of plants (Masujima, 1963; Karavayeva *et al.*, 1970).

Table 16

Coefficients of radionuclide accumulation in superterranean mass of pea and barley in relation to moisture of soddy-meadow soil

Soil moisture coefficient	^{59}Fe	^{60}Co	^{90}Sr	^{91}Y	^{137}Cs	^{144}Ce
Pea						
0.3	0.81 ± 0.02	0.28 ± 0.01	9.90 ± 0.10	0.19 ± 0.02	1.06 ± 0.01	0.07 ± 0.01
0.2	0.81 ± 0.06	0.30 ± 0.03	10.80 ± 0.28	0.16 ± 0.02	1.31 ± 0.09	0.08 ± 0.01
0.1	0.86 ± 0.04	0.27 ± 0.01	9.15 ± 0.43	0.16 ± 0.04	1.05 ± 0.03	0.08 ± 0.01
Barley						
0.3	0.55 ± 0.05	0.06 ± 0.01	1.65 ± 0.06	0.03 ± 0.01	0.17 ± 0.02	0.04 ± 0.01
0.2	0.33 ± 0.05	0.13 ± 0.02	1.40 ± 0.04	0.05 ± 0.02	0.24 ± 0.01	0.04 ± 0.01
0.1	0.52 ± 0.03	0.04 ± 0.01	1.65 ± 0.10	—	0.26 ± 0.02	0.05 ± 0.01

Table 17

Weight of and radionuclide accumulation in superterranean mass of plants as a function of moisture of soddy-meadow soil (percent of content in soil)

Soil moisture coefficient	Dry plant mass (gm)	^{59}Fe	^{60}Co	^{90}Sr	^{91}Y	^{137}Cs	^{144}Ce
Pea							
0.3	4.15	0.190	0.060	2.180	0.040	0.230	0.014
0.2	3.51	0.160	0.060	1.930	0.030	0.230	0.012
0.1	2.20	0.140	0.030	0.980	0.010	0.120	0.008
Barley							
0.3	2.09	0.050	0.007	0.180	0.004	0.018	0.005
0.2	1.67	0.030	0.010	0.130	0.003	0.020	0.003
0.1	1.25	0.020	0.003	0.100	—	0.018	0.002

The results of greenhouse experiments directed at the comparative study of plant accumulation of six radionuclides from soddy-meadow soil under different conditions of its moistening are shown in Table 16. As one can see, despite appreciable differences in accumulation of radionuclides by different plant species, the accumulation coefficients and, consequently, the concentration of nuclides in plants, are not actually dependent on the soil moisture. Since in these experiments the soil was premixed with quartz sand, the values of accumulation coefficients for all of the nuclides under comparison proved noticeably higher than in experiments with the native soil (Table 14).

While the accumulation coefficients for each particular nuclide under different regimes of moistening remain more or less invariable, the overall accumulation of the nuclides by plants somewhat rises as moisture increases on account of a greater buildup of the plant biomass (Table 17).

Similar data were obtained by us with other plant species with respect to the accumulation of ^{90}Sr , ^{137}Cs , and ^{144}Ce from the soddy-meadow soil under the same regime of soil moistening (Tables 18, 19).

Thus, the experiments performed have demonstrated that with an increase in soil moisture from the level of withering to that of full water capacity, the release of radionuclides by the superterranean plant mass increases. This increase is attributable to a more prolific growth of the plant biomass on a more moist soil, whereas the concentration and accumulation coefficients of nuclides in plants did not undergo any changes due to different regimes of moistening.

Influence of EDTA Chelating Agent on Plant Uptake of Radionuclides from the Soil

As demonstrated in the preceding chapter, the ability of radionuclides to migrate in the soil/solution system depends to a great extent on the form in which these nuclides are present in solution. In particular, we noted that the chelated forms of radionuclides are poorly absorbed by the soil, and, as a consequence, their accessibility to plants is enhanced.

At present, artificial chelating agents are finding ever-increasing use for the control of plant accumulation of a number of trace elements and their radioactive nuclides (Wallace *et al.*, 1955a, b, 1957; Atkinson and Wright, 1957; De Kock and Mitchell, 1957; Beckwith, 1958; Kulikov, 1960a, b, 1963; Kartashova, 1963; Beringer, 1963; Essington *et al.*, 1962, 1963; Essington and Nishita, 1966; Nishita and Essington, 1967). In addition, there is evidence that many organic chelating agents that are naturally synthesized by living organisms play an important role in mineral metabolism, forming chelate compounds with slow-moving ions of metals, and thereby converting these metals into a transportable form (Semenov and Tregubenko, 1957; Schmid and Gerloff, 1961; Tiffin and Brown, 1962; Tiffin, 1966; Tiffin and Lee, 1967).

In biological studies the EDTA chelating agent has found a most extensive application. As stated earlier, EDTA in combination with a number of nuclides of interest, forms stable complex compounds characterized by considerable mobility in the soil/solution system. For this reason EDTA was included in an expanded series of experiments the results of which are presented in this section.

Tests were conducted in vegetative pots (holding 2 to 2.5 kg of material) with soddy-meadow soil, to which radionuclides of the relevant chemical elements were added in the form of simple salts and chelate compounds with EDTA. In vegetative pots, pea and barley seeds were planted, and at day 30 after sprouting the content of the radionuclides under study was determined.

Table 18

Coefficients of accumulation of ^{90}Sr , ^{137}Cs , and ^{144}Ce in superterranean

Plant	^{90}Sr		
	Soil moisture		
	0.3	0.2	0.1
Meadow foxtail	2.08 ± 0.28	1.90 ± 0.23	1.57 ± 0.10
Haupt spreading meadow-grass	1.46 ± 0.22	1.34 ± 0.26	1.20 ± 0.15
Meadow fescue	1.01 ± 0.03	1.26 ± 0.14	1.26 ± 0.22
Red fescue	1.28 ± 0.15	1.43 ± 0.23	1.38 ± 0.19
Reed-grass fescue	1.09 ± 0.13	1.12 ± 0.12	1.40 ± 0.25
Smooth brome-grass	1.08 ± 0.16	1.61 ± 0.21	1.72 ± 0.18
Red clover	2.21 ± 0.29	2.88 ± 0.29	2.68 ± 0.20
Platystachyous wheat-grass	1.14 ± 0.11	1.18 ± 0.02	1.14 ± 0.03
Sheep fescue	1.96 ± 0.28	2.03 ± 0.34	—
Mean	1.48 ± 0.06	1.64 ± 0.08	1.54 ± 0.08

Iron, cobalt, and yttrium, added to the soil in the chelated form were easily desorbed by water (Table 20). Consequently, their compounds incorporating EDTA were not destroyed. Chelate compounds of strontium, cesium, and cerium proved unstable in the soil; this is shown by their equally firm retention by soil in two variations of the experiment.

The cobalt/EDTA chelate compound was especially stable: it could remain in the soil for several months without decomposing (Table 21).

Table 19

Weight of and radionuclide accumulation in superterranean mass of plants as a function of soil moisture (data averaged for nine plant species)

Soil moisture coeff- icient	Dry plant weight (gm)	^{90}Sr		^{137}Cs		^{144}Ce	
		% of con- tent in soil ($n \times 10^{-3}$)	10^{-9} Ci per gm dry matter	% of content in soil ($n \times 10^{-3}$)	10^{-9} Ci per gm dry matter	% of content in soil ($n \times 10^{-3}$)	10^{-9} Ci per gm dry matter
0.3	0.9	47.0	170.0	7.0	44.0	0.8	6.6
0.2	0.8	35.0	179.0	7.0	50.0	0.6	5.7
0.1	0.3	15.0	161.0	3.0	55.0	0.3	6.6

mass of nine plant species as a function of soil moisture

^{137}Cs			^{144}Ce		
coefficient					
0.3	0.2	0.1	0.3	0.2	0.1
0.11 ± 0.02	0.30 ± 0.02	0.12 ± 0.03	0.03	0.02	0.03
0.12 ± 0.03	0.17 ± 0.02	0.16 ± 0.05	0.01	0.01	0.01
0.11 ± 0.01	0.11 ± 0.01	0.12 ± 0.01	0.01	0.01	0.01
0.07 ± 0.01	0.11 ± 0.01	0.07 ± 0.01	0.01	0.01	0.01
0.11 ± 0.02	0.15 ± 0.01	0.16 ± 0.03	0.01	0.01	0.01
0.24 ± 0.02	0.32 ± 0.02	0.42 ± 0.02	0.01	0.01	0.03
0.62 ± 0.12	0.70 ± 0.11	0.84 ± 0.10	0.01	0.02	0.02
0.07 ± 0.01	0.18 ± 0.02	0.27 ± 0.03	0.04	0.02	0.02
0.02 ± 0.003	0.05 ± 0.01	0.04 ± 0.01	0.01	0.01	0.02
0.16 ± 0.04	0.23 ± 0.03	0.24 ± 0.04	0.02	0.01	0.02

Chelated ^{59}Fe , ^{60}Co , and ^{91}Y were accumulated by plants in appreciably larger amounts than when these nuclides were added in the form of simple salts (Table 22). As might be expected, the migratory capacity of ^{90}Sr , ^{137}Cs , and ^{144}Ce was not enhanced in the presence of EDTA, since chelate compounds of these radionuclides are not sufficiently stable in the soil.

The nuclides whose mobility is appreciably enhanced under the effect of a chelating agent in the soil/solution system are accumulated by plants in larger amounts (Fig. 14).

Table 20

Desorption by distilled water of radionuclides introduced in the form of simple salts and EDTA complexes

Experimental variation	Desorption (% of content in soil)	Experimental variation	Desorption (% of content in soil)
^{59}Fe	2.2	^{91}Y	5.8
$^{59}\text{Fe}/\text{EDTA}$	70.0	$^{91}\text{Y}/\text{EDTA}$	61.0
^{60}Co	5.0	^{137}Cs	1.0
$^{60}\text{Co}/\text{EDTA}$	85.0	$^{137}\text{Cs}/\text{EDTA}$	1.0
^{90}Sr	7.0	^{144}Ce	1.4
$^{90}\text{Sr}/\text{EDTA}$	9.0	$^{144}\text{Ce}/\text{EDTA}$	1.6

Table 21

⁶⁰Co content in plant superterranean mass and roots after its introduction into the soil in the form of chloride or Co/EDTA complex (counts/min per gm dry matter)

Part of plant	Date of cobalt introduction into the soil			
	November 2, 1959		May 20, 1960	
	CoCl ₂	Co/EDTA	CoCl ₂	Co/EDTA
Superterranean mass	210	2 120	340	2 420
Roots	1 680	12 570	2 170	13 750

Of interest were the results of studies of the effect of varying chelating agent dosages on the plant uptake of ⁶⁰Co from the soil. In these studies the initial cobalt/EDTA solution added to the soil had different molarity ratios of chelating agent to cobalt. The latter, when added to the soil in the form of a chelate compound, was invariably accumulated by plants in a larger amount than in the control (Table 23). One may note that as the chelating agent content in the medium increases, the concentration of a radionuclide in plants is progressively reduced.

Table 22

Accumulation of radionuclides by superterranean mass of plants following introduction into soil of radionuclides in the form of simple salts and EDTA complexes

Experimental variation	Content (counts/min per gm dry matter)		Accumulation coefficient	
	Pea	Barley	Pea	Barley
⁵⁹ Fe	2 500	2 580	0.30	0.30
⁵⁹ Fe/EDTA	38 480	9 650	6.30	1.60
⁶⁰ Co	510	90	0.07	0.01
⁶⁰ Co/EDTA	7 950	560	1.09	0.06
⁹⁰ Sr	55 800	—	1.90	—
⁹⁰ Sr/EDTA	5 800	—	2.30	—
⁹¹ Y	530	520	0.08	0.08
⁹¹ Y/EDTA	4 700	1 550	0.76	0.25
¹³⁷ Cs	4 390	—	0.06	—
¹³⁷ Cs/EDTA	3 680	—	0.02	—
¹⁴⁴ Ce	220	180	0.02	0.02
¹⁴⁴ Ce/EDTA	470	190	0.03	0.01

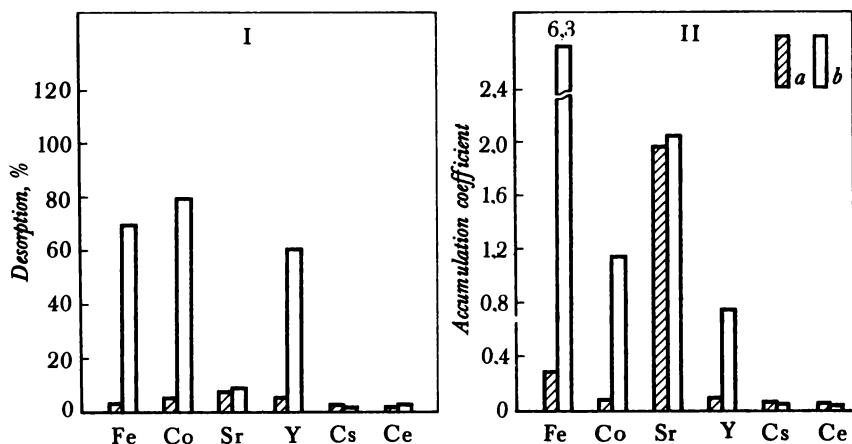


Fig. 14. Desorption of radionuclides from soil by distilled water (I) and accumulation coefficients for superterranean mass of pea (II) when the nuclides were added to the soil in the form of chlorides (a) and chelate compounds with EDTA (b).

Two alternative explanations could be offered with respect to the latter observation: (1) free EDTA molecules taken up by plants form a chelate compound with a portion of cobalt cations, which is then released from the plant into the environment; (2) with excesses of EDTA in the soil, along with free molecules of the chelating agent, readily soluble complexes with other cations are formed which on entering a plant begin to compete with the cobalt/EDTA complex, hampering in this way the uptake of the complex via the root system of plants.

Table 23

Radioactivity of vetch superterranean mass and roots (counts/min/gm dry matter) and the ratio of radioactivity of different portions of plants as a function of molar ratios of a chelating agent and cobalt in the initial solution

Index	^{60}Co control	EDTA/cobalt ratio in solution			
		1 : 1	5 : 1	25 : 1	125 : 1
Superterranean mass	460	7900	6420	6300	5600
Roots	5320	58760	40000	22200	15810
Ratio:					
Roots / Superterranean mass	11.6	7.4	6.2	3.5	2.3
Leaves	2.3	3.8	3.2	3.5	2.5
Stems					

Table 24

⁶⁰Co content in vetch plants by the time of their transfer from radioactive to pure solution and after their growth for 10 days in pure solution (counts/min/plant)

Index	⁶⁰ Co control	EDTA/cobalt ratio in solution			
		1:1	10:1	100:1	1000:1
Prior to transfer to pure solution	2030 ± 10	640 ± 10	300 ± 2	160 ± 30	50 ± 10
After growing in pure solution	1790 ± 270	660 ± 20	420 ± 60	190 ± 10	40 ± 10

In order to test these hypotheses special experiments with aquatic cultures were undertaken in which the uptake of the cobalt chelate compound, as well as its release by plants into the environment, was investigated. In one experimental series, during the first 10 days plants were grown in lake water containing cobalt and EDTA with differing molar ratios. Following this, one portion of plants was examined for the presence of ⁶⁰Co, whereas the other portion, after a thorough washing of the root system, was transplanted into fresh nutrient solution*. At the end of the experiment the content of the radionuclide in water and plants was determined.

In the other experimental series, vetch plants were cultivated under conditions in which one part of the root system of each plant was continually exposed to a solution containing either ⁶⁰Co or the ⁶⁰Co/EDTA compound, whereas the other part was exposed to pure lake water or to nonradioactive EDTA solution without cobalt. At the end of the experiment the radioactivity of different portions of the plants and of the "pure" part of the solution was measured.

In the course of cultivating the plant in pure water its radioactivity did not change as compared to the initial value (Table 24). In the same way as in experiments with soil cultures (Table 23), an increase in EDTA dosage in the solution resulted in a reduction of radiocobalt in the plant.

It should be noted that the uptake by plants of ⁶⁰Co from aqueous solutions was lower when the radionuclide formed a part of EDTA compound than when it was a part of chloride compound. A similar reduction in ⁶⁰Co accumulation, as well as in the accumulation of other radionuclides, from the water medium containing EDTA, was recorded in experiments with freshwater organisms (Timofeyeva-Resovskaya, 1963).

Reduction in the accumulation of cobalt by plants with an excess of EDTA in the medium cannot be explained by its greater release by

* Knopp's nutrient solution: (Ca(NO₃)₂)—1.00 gm/liter; KH₂PO₄—0.25 gm/liter; MgSO₄·7H₂O—0.25 gm/liter; KCl—0.125 gm/liter; Fe₂Cl₆—trace amounts).

Table 25

Accumulation of cobalt by seedlings of cabbage (Slava variety) and tomato (Talalikhinsky variety) as a function of cobalt concentration and the compound form (mg/kg dry matter of the plant superterranean mass)

Amount of cobalt introduced (mg/kg air-dried soil)	Cabbage		Tomato	
	Co(NO ₃) ₂	Co/EDTA	Co(NO ₃) ₂	Co/EDTA
0.15	0.004	0.40	0.004	0.095
15.00	0.800	2.630	1.730	9.940
30.00	2.380	9.300	3.270	21.450
60.00	6.200	20.500	5.300	53.770
120.00	15.250	47.900	9.800	100.500

plants in these conditions (Table 24). This reduction can be attributed to the presence in the medium of free EDTA molecules and EDTA compounds with other cations which compete with the cobalt/EDTA compound for being taken up by a plant. This explanation is supported by the results of the experiments described above involving partitive feeding of plants, which failed to reveal a release by plants of chelated cobalt into the medium. Although this interpretation of the observed effect is perhaps not final, the fact of reduction in cobalt uptake by plants as the EDTA dosage in the medium increases should be taken into consideration when, for instance, chelate compounds of metals are used as microfertilizers.

Cobalt was added to the soil in the form of Co(NO₃)₂ and the Co/EDTA chelate compound. In both variants of the experiment an increase in cobalt concentration in the soil led to an increase in its accumulation by the plants. In addition, the chelated form of cobalt was accumulated by plants to a much greater extent than cobalt added to the soil in the form of nitrate (Table 25).

The results of the experiments designed to study the effect of EDTA chelating agent on the uptake of radionuclides by plants have demonstrated that EDTA enhances the mobility in the soil/plant system of those nuclides with which it forms stable chelates. To this group of radionuclides belong ⁵⁹Fe, ⁶⁰Co, and ⁹¹Y. Chelate compounds of ⁹⁰Sr, ¹³⁷Cs, and ¹⁴⁴Ce are either not formed in the soil or are subject to rapid decomposition.

Chapter 3

MIGRATION AND DISTRIBUTION OF RADIONUCLIDES IN THE SOIL-VEGETATIVE COVER OF NATURAL BIOGEOCENOSSES

The results of the laboratory and greenhouse experiments discussed in the two preceding chapters show the appreciable effect of the factors responsible for the behavior of radionuclides in the soil/solution system (physicochemical properties of the soil, chemical nature of radionuclides in a solution, regime of soil moistening, and so forth) on the behavior of these nuclides in a more complicated soil/plant system. However, the agreement between the laboratory and greenhouse data does not yet make possible a direct extrapolation of the results to natural biogeocenoses. Therefore, we carried out a series of experiments designed to compare the mobility of radionuclides in model systems and under natural conditions.

Comparative Characteristics of the Mobility and Distribution of Radionuclides in the Soil-Vegetative Cover of Biogeocenoses

The experiments described in this section were carried out under natural conditions in two localities of the Ilmen State Reservation: in the vicinity of Lake Bolshoi Tatkul near the Niashevo cordon (Makhonina

Table 26
Chemical characteristics of brown forest soils overlying gneissose granite

Horizon	Loss on calcination (%)	Humus (%)	Exchangeable bases (mEq/100 gm)	
			Ca	Mg
Niashevo				
A	5.04	4.60	9.80	4.40
B	1.98	1.43	7.70	3.10
C	0.83	—	—	—
Tatkul				
A	9.13	4.82	13.80	5.90
B	5.54	2.77	6.50	3.30
C	5.75	—	—	—

Table 27

Mechanical composition of brown forest soil underlain by gneissose granite in the Ilmen Reservation (percent per dry sample)

Horizon	Size of granulometric fractions (mm)						
	1-0.25	0.25-0.05	0.05-0.01	0.01-0.005	0.005-0.001	<0.001	<0.01
Niashevo							
A	31.1	37.6	12.6	3.7	7.1	6.9	17.7
B	37.1	37.2	11.2	4.5	9.1	0.4	14.0
C	45.6	38.1	7.6	0.8	3.2	4.3	8.3
Tatkul							
A	16.8	42.1	13.1	5.3	8.6	13.6	27.6
B	23.2	49.1	8.6	2.3	5.2	8.8	16.3
C	16.5	47.1	19.5	4.9	5.5	5.4	15.8

et al., 1960, 1965; Abaturon *et al.*, 1968; Molchanova *et al.*, 1971; Molchanova and Kulikov, 1972).

The soils in these localities are classified as brown forest sandy soils more or less similar in their chemical composition and mechanical properties (Tables 26 and 27). The site near Lake Bolshoi Tatkul is situated in a birch and aspen forest. The first stratum of the stand is birch aged 60 to 80 years; the second and third strata are aspen. The young growth is abundant, being 50 to 60 cm high, and is represented by aspen grown from sprouts. Its dispersal is uniform and its age is 2 to 4 years. The sparse underwood is 50 to 60 cm high and is represented by broom (*Cytisus rutenicus* Pisch.). The soil cover is 25 to 30 cm high, occupying in total 60% of the area. The herbaceous cover includes goutweed (*Aegopodium podagraria* L.) and gramineous plants in

in the Ilmen Reservation

Gross chemical composition (% per sample calcinated)					pH	
Si	Fe	Al ₂ O ₃ and P ₂ O ₅	Ca	Mg	Aqueous extract	Salt extract
Niashevo						
30.15	4.75	9.48	5.97	1.84	5.6	4.9
27.95	4.74	11.90	5.13	2.44	5.9	5.0
26.51	5.13	16.75	4.84	1.48	6.6	4.5
Tatkul						
36.75	3.14	6.93	1.91	1.07	5.4	4.5
36.72	2.63	6.99	1.96	1.59	5.8	4.8
30.95	4.41	13.15	2.05	2.13	6.0	4.7

Table 28

Distribution of radionuclides in soil-vegetative cover of two natural

Component	⁵⁹ Fe		⁶⁰ Co	
	Deciduous forest	Coniferous forest	Deciduous forest	Coniferous forest
Superterranean plant mass	0.27	0.39	0.04	0.25
Roots	2.85	9.50	32.36*	1.54
Litter	24.50	11.91	11.70	7.20
Soil (total)	72.38	78.20	55.90	91.01
Layerwise (cm):				
0-5	36.92	40.60	50.20	75.01
5-10	18.26	18.80	3.20	8.30
10-15	6.41	13.80	1.40	6.30
15-20	7.50	4.40	1.10	1.40
20-25	3.29	0.60	—	—
25-30	—	—	—	—

* High content of cobalt is explained by a chance aggregation of a great number of roots in this particular plot.

abundance, meadowrue (*Thalictrum aquilegifolium* L.), bedstraw (*Galium boreale* L.), clover (*Trifolium medium* L.), speedwell (*Veronica officinalis* L.), geranium (*Geranium pratense* L.), yarrow (*Achillea millefolium* L.), and a number of other plants.

The site near the Niashevo cordon is located in a ride cut through a pine forest. The pine young growth is abundant, 70 cm high, aged 4 to 10 years. Dispersal is uniform. Birch and larch are rare. The underwood is represented by broom. The herbaceous cover includes gramineous plants and foxberry (*Vaccinium vitis idaea* L.), stoneberry (*Rubus saxatilis* L.), Solomon's seal (*Poligonatum officinale* All.), greater burnet (*Sanguisorba officinalis* L.), bitter peavine (*Lathyrus vernus* Bernh.), monks-hood (*Aconitum excelsum* Rchb.), geranium, bedstraw, and yarrow. The herbaceous cover occupies 50 to 60% of the area, being 20 to 40 cm high.

In the sites described above, 1 × 1 m experimental sample lots were laid out. Early in spring, on these plots prior to plant emergence, radionuclides of iron, cobalt, strontium, yttrium, cesium, and cerium were added. The radionuclides were introduced in the form of chlorous salt solutions with which the plot was superficially irrigated at the rate of about 5 mCi of the respective radionuclide per 15 liters of lake water per plot. The experimental plots with ¹³⁷Cs were studied three years later, those with ⁶⁰Co two years later, and the plots with other radionuclides were studied at the end of the first vegetative season (four months after addition of radionuclide). The following data were recorded: superterranean and root mass of the plants, soil weight in each 5 cm layer down to a depth of 30 cm, and weight of the forest litter. The radioactivity of various portions of plants was determined, after their ashing, by the standard technique, whereas the radioactivity of the soil was estimated following its exsiccation to the air-dried state.

biogeocenoses (percent of total radioactivity)

⁹⁰ Sr		⁹¹ Y	¹³⁷ Cs		¹⁴⁴ Ce
Deciduous forest	Coniferous forest	Desiduous forest	Deciduous forest	Coniferous forest	Deciduous forest
0.34	0.08	0.03	0.03	0.06	0.06
1.70	16.60	1.62	0.30	4.02	1.64
35.91	48.40	22.30	27.17	8.66	20.30
62.05	34.92	76.05	72.50	87.26	78.00
53.89	31.30	61.40	40.00	50.50	51.68
4.27	2.40	6.07	20.30	30.50	17.41
1.14	0.72	4.29	6.20	4.30	5.13
1.13	0.50	4.29	2.90	1.20	3.78
0.38	—	—	3.10	0.58	—
1.24	—	—	—	0.18	—

The superterranean mass of plants carried to the day-time surface only tenth or hundredth fractions of a percent of the total radionuclides of the plot (Table 28). The roots retained approximately one to two orders of magnitude more nuclides than the superterranean mass, whereas the forest litter contained 7.2% to 48.4% of the value. The major amount of radionuclides was concentrated in the top layer of the soil (at a depth of 0 to 5 cm), with all nuclides migrating to a greater or lesser extent to the deeper soil layers.

Of all the radionuclides under study, ⁶⁰Co was, under natural conditions, the least mobile in the soil. Two years after its introduction, nearly 68% of its amount was retained in the upper 5 cm soil layer (Table 29). Low mobility was also characteristic of ⁹⁰Sr and ⁹¹Y, their content in the top layer four months following their addition being 88% and 81%, respectively. The most mobile radionuclides were ⁵⁹Fe and ¹⁴⁴Ce. Their content in the top soil layer was only 51% and 66%, respectively. The content of ¹³⁷Cs in the top layer of the soil was low. It was probably due to a longer duration of experiments with this radionuclide as compared to other nuclides.

Table 29

Distribution of radionuclides throughout the soil profile (percent of the overall radioactivity of the soil)

Depth (cm)	⁵⁹ Fe	⁶⁰ Co	⁹⁰ Sr	⁹¹ Y	¹³⁷ Cs	¹⁴⁴ Ce
0-5	51.2	85.9	88.2	81.4	56.4	65.8
5-10	24.8	7.5	7.2	7.7	31.1	22.5
10-15	13.0	4.8	1.9	5.5	6.8	6.5
15-20	7.8	1.8	1.6	5.4	2.7	5.2
20-25	3.2	—	0.1	—	2.5	—
25-30	—	—	1.0	—	0.5	—

Table 30

General physicochemical characteristics of soils in experimental plots

Topographical element	Biogeocenosis type	Soil	Horizon, depth (cm)	pH of aqueous suspension
Ridge summit	Cowberry pine forest	Primitive accumulative	A ₀ ; 0-6	4.7
			A ₁ ; 6-15	5.3
			AC; 15-25	5.4
Middle of the slope	Baccate pine forest	Brown soil variety	A ₀ ; 0-4	5.2
			A ₁ ; 4-9	5.7
			A ₁ A ₂ ; 9-20	5.5
			BC; 20-55	5.6
Slope foot	Herbaceous pteriodophytic pine forest	Soddy-podzolic	A ₀ ; 0-4	5.1
			A ₁ ; 4-12	5.2
			AC; 12-35	5.3

To check this possibility we carried out additional experiments with ^{137}Cs and ^{90}Sr , in which their migration was studied four years after their addition to the forest litter. Experimental plots were set up in the summer of 1965 on the peak, middle, and foot of the eastern slope of the Ilmen ridge, within the mature pine stands growing on the accumulative, brown-earthy, and soddy-podzolic soils, respectively. The forest types were as follows: cowberry pine on the peak, baccate in the middle part, and herb/fern pine forest at the foot of the slope. The altitude of the ridge was from 450 to 500 m, the angle of dip being within the range of 5° to 10° .

According to its mechanical composition, the primitive-accumulative soil belongs to sandy loam, while the brown-earthy and soddy-podzolic soils are classed with loam (Table 30). The water regime in the cowberry pine forest is characterized by periodic leaching elutriations and by an acute shortage of soil moisture almost every summer.

In the baccate and the herb/fern pine forests the water regime is also characterized by periodic leaching elutriations. Here the soil moisture deficit is less pronounced (Mironov, 1961).

In the selected sites 10×10 cm lots were cleared of forest litter. In one lot the litter was wetted with an aqueous solution of $^{90}\text{SrCl}_2$, in another it was wetted with a solution of $^{137}\text{CsCl}$, following which the litter was put, in equal weight portions, on the cleared lots.

To each experimental lot $20 \mu\text{Ci}$ of ^{90}Sr or ^{137}Cs , respectively, were applied.

Plots were studied 4 years after the addition of radionuclides; $10 \times 10 \times 5$ cm samples of the soil and litter (unit volumes) were taken upwards and downwards on the slope in relation to the site of nuclide application. Each sample of litter or soil was thoroughly mixed, then a sample of the mixture was air-dried and ground in a mortar. The content of radionuclides in the ground mixture specimens was determined radiometrically in 5 replications using a B-2 radiation

Humus (%)	Content of exchangeable bases (mEq/100 gm)			Mechanical composition of soils (% of each fraction)		
	Ca	Mg	Ca + Mg	1.0-0.01 mm	1.0- -0.001mm	<0.001mm
—	—	—	—	—	—	—
14.7	27.5	4.6	32.1	81.6	9.2	9.2
1.4	3.2	0.3	3.5	85.4	8.1	6.5
—	—	—	—	—	—	—
10.8	16.5	4.7	21.2	62.3	22.8	14.9
1.1	3.4	1.6	5.0	65.4	19.7	14.9
0.5	1.3	1.1	2.4	61.3	22.0	16.7
—	—	—	—	—	—	—
2.4	8.8	2.4	11.2	58.6	24.5	16.9
0.5	8.0	1.9	9.9	68.8	21.1	10.1

counter provided with a БФЛ-Т-25 counting tube. The counting time was chosen so that the statistical error should not exceed 5%. With the experiment performed in duplicate 10 radiometric readings, on average, were taken for each sample of soil and litter.

Figure 15 shows isolines of the concentration of radionuclides in the soil profile of experimental lots by the time of completion of the experiment. For convenience, strontium and cesium concentrations are expressed in terms of percentage of the sum total of their concentrations in all unit volumes examined, the centers of which are shown in the figure by respective dots. The concentration isolines were obtained by the linear interpolation method.

It can be seen that the two nuclides migrate predominantly horizontally, along the litter, from the site of application. However, at almost all experimental points located outside the application sites the relative content of ^{90}Sr was markedly higher than that of ^{137}Cs . Thus, in the soil profile of the plots under investigation, strontium, on the whole, proved more mobile than cesium. To the same conclusion points a graph of the vertical radionuclide distribution in soils as plotted along the axes of maximal migration (Fig. 16). In all experimental plots the depth distribution of nuclides in the soil is sufficiently well approximated by the following exponential relationship:

$$A_h = A_0 \exp(-0.693 \frac{h}{h_0}),$$

where A_0 and A_h are the content of the radionuclide on the surface and at a depth h of the soil, respectively, and h_0 is the depth of half-migration numerically equal to the distance from the surface at which the radionuclide concentration in the soil is reduced by half (Kulikov and Piskunov, 1970, 1971; Molchanova *et al.*, 1973). It can be seen from the parameters of the regression equations of A_h over h (Table 31) that the depth of ^{90}Sr half-migration (h_0) is 1.3- to 2.5-fold greater than that of

Fig. 15. Distribution of ^{90}Sr and ^{137}Cs in the soil profile at the summit (a), middle (b), and foot (c) of the slope 4 years after the introduction of radionuclides into the forest litter: (1) forest litter; (2) site of introduction of ^{90}Sr and ^{137}Cs ; (3) geometric centers of layerwise sampling and the relative concentrations of ^{90}Sr (numerator) and ^{137}Cs (denominator); (4), (5) concentration isolines for ^{90}Sr and ^{137}Cs , respectively; (6), (7) axes of maximal migration of ^{90}Sr and ^{137}Cs , respectively.

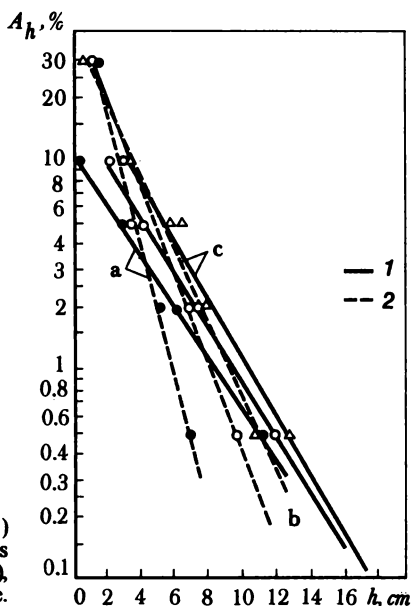


Fig. 16. Vertical distribution of ^{90}Sr (1) and ^{137}Cs (2) in the soil along the axes of maximal migration at the summit (a), middle (b), and foot (c) of the slope.

^{137}Cs (h_0). Differences in h_0 values between the sites located on the peak, middle, and foot of the slope, proved statistically nonsignificant for both cesium and strontium. Consequently, the rate of vertical migration of each nuclide, in soils of sylvatic biogeocenoses of differing types, was practically similar, although at all sites the downward migration of strontium proceeded more rapidly than that of cesium. ^{59}Fe and ^{144}Ce proved to be the most mobile nuclides in these soils of natural biogeocenoses. This was probably due to the physicochemical status of iron and cesium compounds in the soil solution.

From the results of laboratory experiments (see Chapter 1) it follows that at a pH of 6 to 8 (characteristic for all of the soils under examination) trace amounts of iron and cerium radionuclides are partly converted into nonsorbed forms that are relatively highly mobile in the soil/solution system. As compared with the other radionuclides, ^{90}Sr under natural conditions was less mobile in soil than it was expected on the basis of laboratory and greenhouse experiments. This appears to be related to the conversion with time of strontium in the soil into a nonexchangeable state because of isomorphous replacement of calcium in soil minerals (Pogodin, 1968). Moreover, as was noted earlier, leveling of nuclide migration parameters in natural soils may occur under the effects of seasonal change in the regime of soil moistening.

Migration of ^{90}Sr and ^{137}Cs in the Soil-Vegetative Cover of Tundra Biogeocenoses

In the work just described we considered the comparative distribution and migration of the radionuclides under study in the soil-vegetative cover after their artificial introduction into upper layers of the soil or into the forest litter. Under these conditions the nuclide uptake by plants was essentially determined by absorption via roots. Under natural conditions and on a worldwide scale the principal primary source of radioactive substances contaminating biogeocenoses is radioactive fallout. On entering the Earth's surface radioactive substances interact with the vegetative cover, and as a result an appreciable amount of such substances is retained by the superterranean parts of plants. The coefficient of radionuclide retention (the ratio of the amount retained by the superterranean mass of plants to the total amount of radionuclides per unit area) may reach rather high values (Molchanov *et al.*, 1968; Aleksakhin *et al.*, 1969, 1970). With time, through death of plants, falling of leaves, leaching, and other processes, a considerable amount of radioactive substances gets into the soil and the litter layer which, in turn, gradually become a major source of radionuclides to be taken up by plants via the root system.

The behavior of radionuclides falling from the atmosphere on the soil-vegetative cover of differing biogeocenoses depends on a multitude of factors, including climate, type of soils and plants, moisture content in the area, direction of solid and liquid runoff, and so forth. The existence of such a complex relationship dictates to a large measure the necessity for comprehensive studies of radionuclide behavior in landscapes and soils of differing soil-climatic zones. To date specific features of the distribution of some radionuclides in the landscapes and soils of the Central European part of the USSR have been studied to a large extent (Baranov *et al.*, 1965; Tyuryukanova *et al.*, 1964, 1966; Rosyanov *et al.*, 1968; Pavlotskaya *et al.*, 1970). The Far North regions, and in particular the tundra zone, are still inadequately studied in this respect.

Permafrost, excessive moistening, the specific composition of plants with a preponderance of slowly-growing mosses and lichens and a number of other specific features of the tundra landscape determine a small capacity of the biological turnover and its slow rate. All of these

Table 31

Parameters of regression equation (A_h with respect to h)

Location of experimental sites	Line index	For ^{90}Sr		For ^{137}Cs		h_0/h'_0
		$A_0, \%$	h_0, cm	$A_0, \%$	h'_0, cm	
Slope summit	a	10	2.5	61	1.0	2.5
Middle	b	16	2.3	52	1.4	1.6
Slope foot	c	26	2.2	42	1.7	1.3

circumstances should influence the behavior of radionuclides entering northern landscapes with the atmospheric fallouts.

We conducted our investigations in the vicinity of the Harp Station (Yamal-Nenets National District, Tyumen Region, Siberia) affiliated with the Institute of Plant and Animal Ecology, the Urals Branch of the USSR Acad. Sci., where an area was chosen comprising the watershed (summit of the ridge), slope, and foot of the ridge (sphagnum-peat bog). In July 1966 three pits, spaced 2 to 4 m apart, were sunk in the soil of each topographic plot. The soil cover of the site on the ridge summit was represented by pseudogley-clay-loamy soils. In the vegetative cover, moss, lichens, ledum, and foxberry were predominant. On the slope the soils were peaty-pseudogley, humic-infiltrated, clay-loamy (the tundra-gley type of soil). In them the 0-4 cm moss layer is sharply replaced by the thin humic horizon A₁. The plant association of these soils is represented by "yernik" (dwarf arctic birch). Soils of the sledge-sphagnum bog at the slope foot are underlain beneath the sphagnum moss layer by a well-defined peat-forming horizon 15 to 20 cm thick which abruptly changes to horizon G. The soils described are characterized by an acid pH (3.1 to 4.5) of their potassium chloride salt extract; the humus content varies from 6% in the A₁ horizon to one-tenth of this in the Bgl and G horizons. The sum of exchange bases is 10 to 25 mEq per 100 gm of the soil.

The level of ⁹⁰Sr and ¹³⁷Cs was determined in soil samples taken from genetic horizons to a depth of 20 to 25 cm and also in the vegetative cover. ⁹⁰Sr was radiochemically determined from the daughter ⁹⁰Y data (Pavlotskaya *et al.*, 1964). Radioactivity of samples was measured with the aid of a low-background YMΦ-1500 apparatus.

The ¹³⁷Cs content was determined spectrometrically with multi-channel АИ-100-1 gamma-analyzer using a 40 × 40 mm NaI(Tl) crystal as a detector (Molchanova and Kulikov, 1970a, c).

The values of ⁹⁰Sr and ¹³⁷Cs content in the soil-vegetative cover are shown in Table 32. They indicate that the concentration of radionuclides in the vegetative cover and in soils of the eluvial and accumulative landscape areas is practically similar. However, the pattern of strontium and cesium distribution in the soil-vegetative cover of the area under investigation is dissimilar. Thus, on the ridge summit and on the slope the highest concentration of strontium and cesium occurs in plants. In the underlying layers (the forest litter and soil) the concentration of radionuclides does not exceed several percent or decimal fractions of percent of their concentration in plants. On the plot having the sphagnum peat bog at the foot of the slope no predominant accumulation of ⁹⁰Sr in plants was observed. Between the older, peat-forming moss layers and the current year moss increment ⁹⁰Sr was distributed more or less uniformly, whereas cesium concentrated in the growing portions of moss.

Table 33 shows the ⁹⁰Sr and ¹³⁷Cs content in the tundra soil-vegetative cover as calculated per unit area. In comparing the data in Tables 32 and 33 one can see that despite the gross differences in the volume weight of plants and soil genetic horizons, the pattern of

radionuclide distribution remains unaltered regardless of the method of expressing the results. The major portion of ^{90}Sr and ^{137}Cs is retained, independently of the plot, in the plant cover and upper layers of the soil. ^{90}Sr has, however, a greater ability to migrate both through the entire landscape and through profiles of soils of different types. This is indicated by differing ^{90}Sr to ^{137}Cs ratios in eluvial and accumulative parts of the landscape (Table 33). Thus, on the ridge summit, this ratio throughout the entire layer under study is 0.9, whereas in the middle of the slope it is 0.3. This testifies to a more intensive evacuation of strontium from eluvial and transeluvial plots. This inference is well corroborated by a relative enrichment with radioactive strontium of the peat bog located at the slope foot. Here the ratio of ^{90}Sr to ^{137}Cs attains 2.2.

It is of interest to note that the relative enrichment of the peat horizon with strontium increases with depth. Thus, for instance, the ratio of strontium to cesium in the current year increment of the sphagnum moss was 0.6, in the peat-forming layer at a depth of 2 to 7 cm it was 1.6, and at a depth of 7 to 12 cm the ratio was 5.4. Such a pattern of radionuclide distribution in the peat horizon may possibly be explained by the fact that the most intensive uptake of strontium, as compared to that of cesium, takes place in the initial period following radioactive fallout; subsequently, as ^{90}Sr enters biological links and becomes more firmly retained by the soil, the bias in the migration of strontium and cesium in favour of strontium decreases.

Table 32

Concentration of ^{90}Sr and ^{137}Cs in soil-vegetative cover of tundra

Soil	Horizon and depth (cm)	^{90}Sr	^{137}Cs
		$n \times 10^{-12}$ Ci/gm dry matter	$n \times 10^{-12}$ Ci/gm dry matter
Supragleisolic (ridge summit)	Vegetative cover*	26.33 \pm 2.28	38.18 \pm 2.07
	A ₁ 2-7	2.16 \pm 0.34	2.90 \pm 0.63
	Bgl 7-12	0.50 \pm 0.17	0.90 \pm 0.63
	Bgl 12-17	0.38 \pm 0.06	not detected
	G 17-22	0.38 \pm 0.06	not detected
Peaty supragleisolic (middle of the slope)	Vegetative cover*	15.92 \pm 1.76	38.70 \pm 1.04
	A ₀ 2-4	5.48 \pm 0.63	3.81 \pm 0.78
	A ₁ 4-9	0.12 \pm 0.01	0.62 \pm 0.03
	Bgl 9-14	0.05 \pm 0.01	not detected
	Bgl 14-19	0.06 \pm 0.02	not detected
	Bgl/G 19-24	0.03 \pm 0.01	not detected
Sphagnous-peaty-boggy (slope foot)	Vegetative cover	27.00 \pm 6.82	40.68 \pm 2.53
	T ₁ 2-7	41.54 \pm 6.82	17.40 \pm 0.96
	T ₂ 7-12	20.45 \pm 1.31	4.40 \pm 0.52
	T ₂ 12-17	5.34 \pm 2.23	not detected
	G 17-22	0.52 \pm 0.24	not detected

* Vegetative cover also includes the moss layer (0-2 cm) and the higher-plant root system present in this layer.

Table 33

Distribution of ^{90}Sr and ^{137}Cs in soil-vegetative cover of tundra
as calculated per unit area

Soil	Horizon and depth (cm)	^{90}Sr		^{137}Cs		Sr/Cs
		$n \times 10^{-9}$ Ci/m ²	% of total content in soil-vegetative cover	$n \times 10^{-9}$ Ci/m ²	% of total content in soil-vegetative cover	
Supragleisolic (ridge summit)	Vegetative cover	33.9	32.9	56.7	48.6	0.6
	A ₁ 2-7	30.8	29.9	30.0	25.8	1.0
	Bgl 7-12	11.1	13.7	29.9	25.6	0.5
	Bgl 12-17	12.3	12.0	not detected	—	—
	G 17-22	11.8	11.5	not detected	—	—
	Total	102.9		116.6		0.9
Peatysupragleisolic (middle of the slope)	Vegetative cover	27.7	67.4	85.4	69.1	0.3
	A ₀ 2-4	4.7	11.0	11.7	9.4	0.4
	A ₁ 4-9	2.6	6.1	26.5	21.5	0.1
	Bgl 9-14	3.8	8.9	not detected	—	—
	Bgl 14-19	1.8	4.2	not detected	—	—
	Bgl/G 19-24	1.0	2.4		—	—
	Total	42.6		123.6		0.3
Sphagnous-peaty-boggy (slope foot)	Vegetative cover	21.9	12.0	34.8	42.0	0.6
	T ₁ 2-7	58.9	32.5	37.6	45.4	1.6
	T ₂ 7-12	56.2	31.0	10.4	12.6	5.4
	T ₂ 12-17	28.1	15.5	not detected	—	—
	G 17-22	16.3	9.0	not detected	—	—
	Total	181.4		82.8		2.2

Thus, investigation of the ^{90}Sr and ^{137}Cs distribution in the soil-vegetative cover of the three contiguous plots of tundra landscape described here has demonstrated that these radionuclides are taken up by plants to the maximum degree. On the whole, ^{90}Sr has the greater ability for migration and therefore enriches accumulative portions of the landscape.

Effect of the Vegetative Cover on the Vertical Migration of Radionuclides in the Soil

In order to predict the fate of a radioactive substance in a biogeocenosis, quantitative evaluation of the role of plants in vertical migration of individual radionuclides in the soil is needed. We did such an evaluation under experimental conditions with regard to cobalt, strontium, cesium, and cerium radionuclides (Kulikov and Korobitsin, 1968; Kulikov and Piskunov, 1969, 1970, 1971).

The experimental procedure was as follows. On a homogeneous plot of soddy-meadow soil we excised monolithic blocks of earth ($60 \times 75 \times 60$ cm) and placed them in lysimeters* made of stainless steel. The upper sodded soil layer from 0 to 10 cm was removed, sifted, and then returned to the lysimeter. Into the surface soil layer (0 to 2 cm depth) in the lysimeters, after partitive irrigation and thorough mixing, chloride solutions of ^{60}Co , ^{90}Sr , ^{137}Cs , and ^{144}Ce were added in amounts of 1.5, 1.0, 2.4, and 1.0 mCi per lysimeter, respectively. This was equal to 200, 150, 360, and 150 μCi per 1 kg of the air-dried soil in the primary contamination zone.

Following introduction of radionuclides into the soil, lucerne seeds (*Medicago sativa* L.) were planted in some lysimeters, while the remaining blocks were kept free of plants throughout the experiment. Two extra lysimeters into which no radionuclides were added served as controls for checking the possible influence of the introduced nuclides on the development of plants.

The lysimeters were then installed in trenches under the open sky. Moisture of the soil blocks was continually maintained at the level of field moisture capacity. Experiments with cerium lasted for two vegetative seasons, those with cobalt for three seasons, and those with strontium and cesium lasted for four vegetative seasons (39 months). In lysimeters charged with ^{90}Sr and ^{137}Cs , after the third vegetative season (winter 1966/1967), the lucerne was damaged by frost and in its place a community of natural herbs, with a prevalence of greengrass (*Poa pratensis* L.) and common yarrow (*Achillea millefolium* L.), was grown during the last vegetative season. Freezing of the lucerne had no relation to the biological effects of radionuclides since in the control lysimeters the lucerne was also killed by frost.

By the end of each vegetative season the weight of the superterranean mass of plants and the uptake by it of radionuclides from the soil were measured. On completion of the experiment four sections were made in each soil block through which soil samples were taken from each successive layer and at a different distance from the primary contamination zone. From lysimeters containing plants, roots were picked out from all soil layers, and their total biomass and radioactivity were recorded. In this way there was obtained a picture of

* A device for measuring the amount of water seeping through the upper soil layer.

Table 34

Accumulation of radionuclides by superterranean mass and roots of plants in different years of vegetation

Vegetative season of plants after introduction of emitters	Superterranean mass				Roots			
	Dry weight (gm/lysime- ter)	Concentra- tion (counts/ min/gm dry matter)	Accumula- tion coeffi- cient	Content (%) of the amount introduced into soil)	Dry weight (gm/lysime- ter)	Concentra- tion (counts/ min/gm dry matter)	Accumula- tion coeffi- cient	Content (%) of the amount introduced into soil)
				⁶⁰ Co				
First (1964)	38.8	340	0.010	0.009	—	—	—	—
Second (1965)	108.8	30	0.001	0.002	—	—	—	—
Third (1966)	130.3	50	0.002	0.003	98.5	940	0.040	0.02
				⁹⁰ Sr				
First (1964)	55.5	4 170	0.200	0.160	—	—	—	—
Second (1965)	121.6	1 330	0.060	0.120	—	—	—	—
Third (1966)	170.7	2 920	0.140	0.360	—	—	—	—
Fourth (1967)	136.9	5 590	0.270	0.540	181.4	13 680	0.660	0.160
				¹³⁷ Cs				
First (1964)	51.3	700	0.024	0.018	—	—	—	—
Second (1965)	139.1	20	0.001	0.001	—	—	—	—
Third (1966)	164.6	80	0.003	0.006	—	—	—	—
Fourth (1967)	108.2	1 150	0.040	0.064	157.6	3 160	0.100	0.020
				¹⁴⁴ Ce				
First (1964)	43.4	150	0.005	0.003	—	—	—	—
Second (1965)	109.0	110	0.004	0.007	111.9	1 190	0.040	0.012

Table 35

Radioactivity of experimental phytocenosis root system in each layer and radio-

Soil layers (cm)	Root system radioactivity				
	Depth of ^{60}Co				
	0-10	10-30	30-50	50-70	70-90
0-10	2 160	30	0	0	0
10-20	130	2 130	0	0	0
20-30	0	1 980	0	0	0
30-40	0	270	2 190	0	0
40-50	0	0	3 100	0	0
50-60	0	0	250	1 920	0
60-70	0	0	0	1 560	90
70-80	0	0	0	60	3 090
80-90	0	0	0	0	4 140
90-100	0	0	0	0	100

the radionuclide distribution in the plant root system and over layers of the soil with and without vegetative cover. One may note that during the first vegetative season the amount of all radionuclides accumulated by the superterranean mass of the lucerne was somewhat greater than that in the following seasons (Table 34), the concentration of ^{90}Sr being higher than that of other radionuclides. Values of the accumulation coefficient (in this case the ratio of radionuclide concentration in plants to its concentration in the upper, primarily contaminated soil layer) varied in the case of strontium from decimal to hundredth fractions of one percent, and in the case of the other nuclides the values were one to two orders of magnitude lower. Accordingly, the overall uptake of ^{90}Sr by the superterranean mass of plants during one vegetative season reached some decimal parts of one percent of the amount introduced into the soil, while the uptake of the remaining radionuclides did not exceed hundredths and thousandths of one percent.

Decline in the nuclide accumulation by the superterranean mass during the second and third vegetative seasons may be explained by their increased retention by the soil with time (Gulyakin and Yudinseva, 1962; Kulikov, 1965a). In addition, this decline may partly be due to the displacement during the following years of the most active zone of the lucerne root system from the upper, radioactive, layers of the soil to the deeper ("pure") layers. A certain increase in the accumulation of ^{90}Sr and ^{137}Cs during the last (fourth) vegetative season may be attributed to the development in the lysimeters during that period of a rather complex plant community, instead of the lucerne, in which the green-grass and yarrow were prevalent as regards the number of plants and the amount of biomass. These two plant species have a superficial root system and accumulated radionuclides from the upper soil layers more vigorously than did the lucerne.

The coefficients of nuclide accumulation for roots (the zone of

Soil radioactivity				
introduction (cm)				
0-10	10-30	30-50	50-70	70-90
2 750	0	0	0	0
160	2 140	0	0	0
0	3 380	0	0	0
0	100	2 030	0	0
0	0	2 960	0	0
0	0	30	2 240	0
0	0	0	3 300	0
0	0	0	190	2 620
0	0	0	0	3 540
0	0	0	0	70

primary contamination of the soil) were somewhat higher than for the superterranean mass of plants, but even in this case they generally did not exceed some decimal parts of the unity. The peak concentration was recorded in the part of roots that was located within the initially contaminated soil layer. Outside this layer the root radioactivity sharply decreased and at a depth of 8 cm it was practically undetectable. A conclusion can be drawn that the radionuclides under study, after having got into the surface layer of the soil, are rather poorly accumulated in the superterranean portions of plants, have a low migration rate in the root system, and are retained principally in that portion of the roots which is in immediate contact with the radioactive soil.

Distribution of radionuclides over the soil layers in lysimeters with no plants is shown in Figs. 17 and 18, and in those with plants in Figs. 19 and 20. Analysis of these data shows that change in concentration of radionuclides A_h in the soil at depth h is expressed (in the absence of plants) by exponential function, which in the general form can be described by the equation $A_h = A_0 \exp(-kh) = A_0 \exp(-0.693 \frac{h}{h_0})$, where A_0 is the initial amount of the nuclide added to the surface soil layer at the start of the experiment and taken as 100%, k is the logarithmic decrement of the decline in radioactivity with depth, and h_0 is the half-migration depth.

The vertical distribution of radionuclides in lysimeter soils with vegetative cover is functionally different (Figs. 19 and 20). In this case the change in the radionuclide concentration with depth obeys a power

function of the $A_h = A_0 h^{-\frac{\tau}{m}}$, where τ and m are coefficients of dimension and migration, respectively.

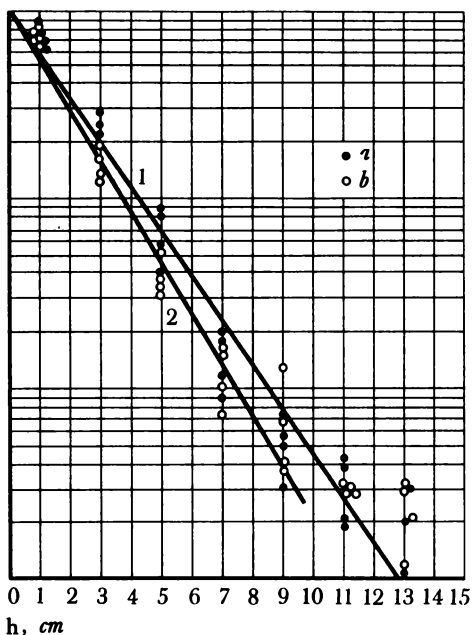
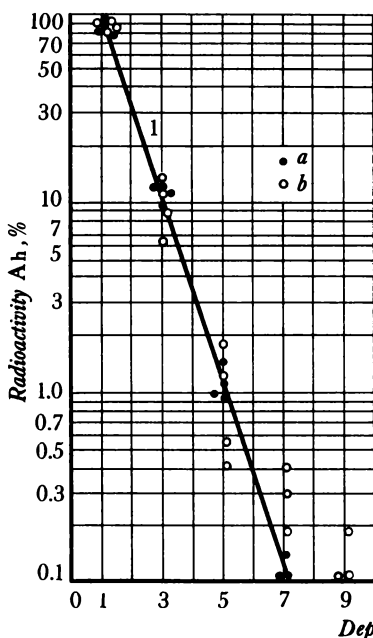


Fig. 17 (left). Vertical distribution of ^{60}Co (a) and ^{144}Ce (b) in lysimeter soil samples with no plants. Theoretical regression line for A_h with respect to h for cobalt and cerium.

Fig. 18 (right). Vertical distribution of ^{90}Sr (a) and ^{137}Cs (b) in lysimeter soil samples with no plants. Theoretical regression lines for A_h with respect to h for strontium (1) and cesium (2).

In comparing the migration of radionuclides in soils with and without vegetation one may note that in the former case the nuclides penetrate to a greater depth. This is due to the influence of the root system of plants, which facilitates a deeper penetration of the nuclides into the soil.

As stated above, the laboratory experimental data indicate that ^{90}Sr is retained by the soil less firmly than the other radionuclides under study. This explains a more intensive uptake of strontium from the soil by plants. On the basis of the data from laboratory and greenhouse experiments one could expect, in our experiments carried out under conditions close to the natural ones, that when the natural structure of the soil blocks was undisturbed, strontium would be more mobile in the soil than cesium. Yet parameters of migration of the four radionuclides proved very similar, particularly in the soils with vegetative cover (Figs. 19 and 20).

Other investigators have also arrived at the same conclusion after analysis of data on ^{90}Sr and ^{137}Cs present in these soils as a result of a radioactive fallout (Makhonko and Chumichev, 1969). It can be assumed that in this instance (as well as in the earlier described

experiments with forest soils) the principal cause of the reduced relative mobility of ^{90}Sr in soils is its change in the course of time to the nonexchangeable state. Since the soil in the lysimeters was continually subjected to excessive moistening, the moisture factor could also be responsible for leveling the rate of radionuclide migration.

The results of experiments with ^{60}Co introduced at different depths from the soil surface also show the negligible role of vegetation in the vertical downward movement of radionuclides in the soil (Kulikov, 1965b).

The maximal uptake of cobalt by the superterranean mass of the experimental plants (equal to hundredths of one percent of the amount introduced into the soil) was observed following superficial introduction of the element to a depth of 10 cm (Fig. 21). When the nuclide was introduced into deeper layers (10 to 30 cm and 30 to 50 cm) its uptake dropped to thousandths of one percent, and at depths lower than 50 cm the nuclide became practically inaccessible to the plant superterranean mass, since in that particular case ^{60}Co accumulated only in that part of the root system which was developing directly in the radioactive soil layer (Table 35), as was the case in experiments with lysimeters. After 3 years following its introduction, cobalt was practically fully retained

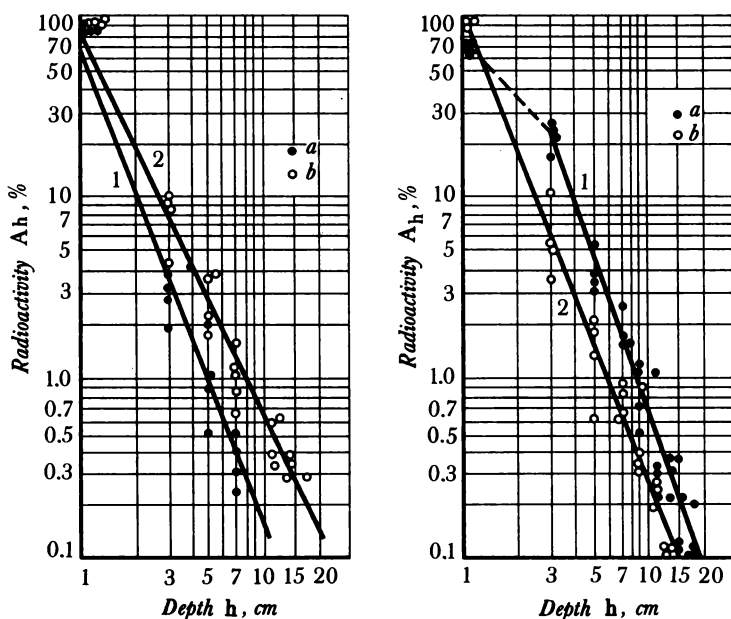


Fig. 19 (left). Vertical distribution of ^{60}Co (a) and ^{144}Ce (b) in lysimeter soil samples with plants. Theoretical regression lines for A_h with respect to h for cobalt (1) and cerium (2).

Fig. 20. Vertical distribution of ^{90}Sr (a) and ^{137}Cs (b) in lysimeter soil samples with plants. Same designations as in Fig. 18.

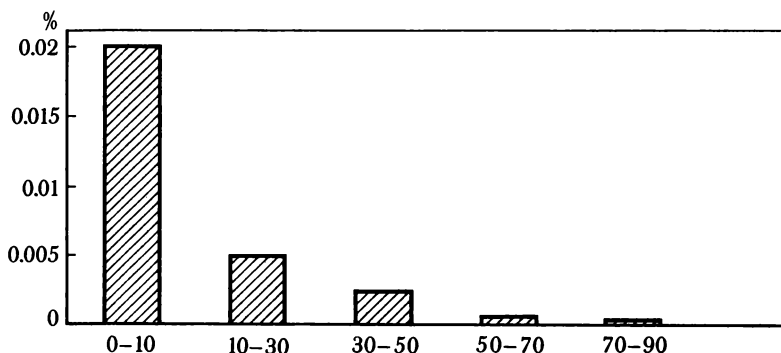


Fig. 21. Accumulation of ^{60}Co by superterranean biomass of experimental phytocenosis. On abscissa depths are plotted to which the radionuclide was introduced into soil (cm); ordinates show its accumulation (percentage of amount introduced per kilogram of dry mass of plant).

by those soil layers in which it had been originally introduced; only a small portion of cobalt migrated into the underlying soil layers to a depth not exceeding 10 cm.

Overall, the data presented in this section suggest that in the soil the vertical migration rate of the radionuclides under study is not high and that the grass cover only insignificantly accelerates the migration of these nuclides deeper into the soil.

Chapter 4

MIGRATION AND DISTRIBUTION OF RADIONUCLIDES IN THE WATER/FRESHWATER ORGANISM SYSTEM

The water/hydrobiont system, like the soil/plant system for the terrestrial biogeocenosis discussed above, is one of the primary links in the migration of radioactive substances in bodies of water. At the same time this is the system that is most sensitive to diverse damaging factors, since it is within this system that the living organisms function.

The role of hydrobionts in processes of concentrating radioactive elements from water was originally noted by V.I. Vernadsky and his collaborators as far back as the 1920's (Vernadsky, 1929, 1930; Brunovsky, 1935; Brunovsky and Kunashova, 1932; Vinogradov, 1935). Later, many investigators corroborated a high concentrating capacity of aquatic organisms with respect to natural radioactive elements (Wiesner, 1938; Agranat, 1958, Tsivoglou *et al.*, 1958; Templeton, 1958; Polikarpov, 1960, 1964; Polikarpov and Ten, 1961; Kovalsky and Vorotnitskaya, 1965; Kovalsky *et al.*, 1968; Popova, 1965; Iskra *et al.*, 1969a, b, 1970a, b, 1971a, b).

In the last few years numerous studies have been carried out concerning the accumulation by hydrobionts of fission fragment radionuclides entering waterbodies from atmospheric fallouts and with overland water runoff. These studies were reviewed in monographs by Timofeyeva-Resovskaya (1963), Polikarpov (1964), and by Kulikov (1971a, b).

Comparative Characteristics of Radionuclide Accumulation by Freshwater Hydrobionts

The most important works on the comparative study of radionuclide accumulation by diverse freshwater flora and fauna have been done by the staff of the Institute of Plant and Animal Ecology, the Urals Branch of the USSR Acad. Sci. (Timofeyev-Resovsky, 1957, 1962; Timofeyeva-Resovskaya, 1963; Gileva, 1965; Ivanov *et al.*, 1965; Timofeyeva and Kulikov, 1967; Kulikov *et al.*, 1968a, 1971e, f; Kulikov, 1971b; Piskunov *et al.*, 1971a,b). These publications contain data on the coefficients of accumulation of radioisotopes of 20 chemical elements for more than 40 species of freshwater plants and 25 species of animals.

On the average, freshwater plants have considerably higher

coefficients of accumulation for radionuclides under study than do aquatic animals (Tables 36 and 37). The highest accumulation coefficients were noted for iron, cobalt, yttrium, and cerium, and the lowest coefficients for strontium and cesium. It is noteworthy that the value of the accumulation coefficient is defined not only by the chemical nature of radionuclides, but also by the species specificity of aquatic organisms. Among freshwater plants and animals included in this study there are some species that have very high accumulation coefficients for

Table 36

Coefficients of radionuclide accumulation for different freshwater plant species calculated per dry mass (Timofeyeva-Resovskaya, 1963)

Hydrobionts	⁵⁹ Fe	⁶⁰ Co	⁹⁰ Sr	⁹¹ Y	¹³⁷ Cs	¹⁴⁴ Ce
Aquatic bacteria			440		100	11 140
Rock lichen						
bacteria	555	370	205		200	740
<i>Scenedesmus quadricauda</i> Bret.	9 000	390	130	22 050	280	20 700
<i>S. acuminatus</i> Chodat.	35 000	250	105	1 530	390	38 500
<i>Cladophora flacta</i>						
Kützing	26 800	8 750	1 910	119 625	1 230	35 600
<i>Gl. glomerata</i> Kützing	31 500	1 985	900	40 000	1 565	31 000
<i>Mougeotia</i> sp.	34 000	238 000	190	15 600		5 400
<i>Spirogyra crassa</i>						
Kützing	1 660	17 000	235	6 860	285	41 200
<i>S. sp.</i>	3 120	5 640	550	13 990	1 920	12 800
<i>Chara</i> sp.			350	3 790	200	4 120
<i>Ch. aspera</i> Willdenow			280	5 825	180	6 800
<i>Ch. fragilis</i> Desw.	15 500	7 425	400	6 335	365	19 510
<i>Ricciocarpus natans</i> L.			590	20 425	715	9 800
<i>Fontinalis</i> sp.			360		1 020	9 650
<i>Ranunculus confertifolius</i> Fries			465		765	7 880
<i>Myriophyllum spicatum</i> L.	530	3 500	445	4 710	295	4 100
<i>Ceratophyllum demersum</i> L.	4 510	4 665	510	920	300	11 250
<i>Lythrum</i> sp.			50		185	
<i>Cicuta virosa</i> L.			130		140	
<i>Utricularia vulgaris</i> L.		11 650	665	6 935	340	6 050
<i>Typha angustifolia</i> L.			25		20	
<i>Calla palustris</i> L.			45		70	
<i>Lemna minor</i> L.	9 050	3 900	400	4 425	2 425	15 000
<i>L. trisulca</i> L.		14 000	315	6 170	940	105 000
<i>L. polyrrhyza</i> L.			590	8 435	1 050	5 520

Table 36—continued

Hydrobionts	⁵⁹ Fe	⁶⁰ Co	⁹⁰ Sr	⁹¹ Y	¹³⁷ Cs	¹⁴⁴ Ce
<i>Potamogeton natans</i>						
L.			670		205	1 370
<i>P. compressus</i> L.			1 020		115	2 435
<i>P. filiformis</i> Persoon			585	11 670	185	3 255
<i>P. perfoliatus</i> L.			685	4 740	195	1 925
<i>Elodea canadensis</i>						
Richard	4 735	3 490	805	2 120	285	5 300
<i>Vallisneria spiralis</i> L.			220		360	7 100
<i>Stratiotes aloides</i> L.		4 900	615		375	
<i>Hydrocharis morsus</i>						
<i>ranae</i> L.		5 430	415	5 000	535	2 485
<i>Carex</i> sp.	6 865	4 595	160	6 535	670	6 730
Mean:	12 775	18 665	470	14 440	545	11 660

Table 37

Coefficients of radionuclide accumulation for different freshwater animals calculated per dry mass (Timofeyeva-Resovskaya, 1963)

Hydrobionts	⁵⁹ Fe	⁶⁰ Co	⁹⁰ Sr	⁹¹ Y	¹³⁷ Cs	¹⁴⁴ Ce
Worms:						
<i>Herpobdella</i> sp.	330	275	8	145	10	125
Mollusks:						
<i>Anadonta cellensis</i>						
Schröter		1 090	85		90	370
<i>Limnaea stagnalis</i> L.	125	325	3 100	2 020	215	2 180
<i>Radix auricularia</i> L.		925	60		290	1 050
<i>R. ovata</i> Draparnaud			320		390	7 130
<i>Galba palustris</i>						
Müller		1 160	585			2 300
<i>Bithynia tentaculata</i>						
L.		560	170		690	3 300
<i>Aplexa hypnorum</i> L.	720	1 380	505		510	7 220
<i>Anisus vortex</i> L.		870	305		60	
<i>Planorbis planorbis</i> L.		800	210		60	
Crustacea:						
<i>Diaptomus graciloides</i>			90		1 270	6 460
<i>Mesocyclops</i> sp.			170		530	
<i>Chydorus sphaericus</i>			370		1 020	
<i>Rivulogammarus lacustris</i> Sars	350	1 100	400		235	1 580

Table 37—continued

Hydrobionts	⁵⁹ Fe	⁶⁰ Co	⁹⁰ Sr	⁹¹ Y	¹³⁷ Cs	¹⁴⁴ Ce
Larvae and insects:						
<i>Culex pipiens pipiens</i> L.		4 010	355		245	9 050
<i>Theobaldia alasensis</i> Ludlow					40	12
<i>Halesus interpunctatus</i> Lett.	410	8 500	12		120	1 100
<i>Leptocerus</i> sp.			20		20	360
<i>Phryganea grandis</i> L.			18		680	2 430
<i>Aeschna</i> sp.		635	7		100	2 600
<i>Lestes</i> sp.			4		315	325
<i>Eristalis</i> sp.		390	60		35	805
<i>Tendipes</i> sp.	680	375	105	123	530	1 020
Vertebrates:						
<i>Cyprinus carpio</i> L. × × <i>Cyprinus carpio</i> <i>haematopterus</i> Tem- minck et Schlegel		60	160		55	820
<i>Rana</i> sp. (tadpoles)			210			70 100
Mean:	435	1 405	305	765	325	5 730

one or another radionuclide, much in excess of the mean value for it (Tables 36 and 37). These species were termed "specific accumulators" or indicator-species signaling the presence of a particular radionuclide in the environment (Timofeyeva-Resovskaya, 1963; Polikarpov, 1964).

Analysis of the accumulating capacity of differing taxonomic groups of hydrobionts with respect to radionuclides of 14 elements confirmed the conclusion (Gileva, 1965) that the accumulation coefficients for plants are higher than those for animals, and that, on the average, unicellular and filamentous algae accumulate these radionuclides to a greater degree than do higher plants. Consequently, these data make it possible to trace a certain dependence of radionuclide accumulation on the position of various hydrobionts in the phylogenetic system. It should be noted that aquatic bacteria exhibit appreciably lower accumulation coefficients with respect to a number of radionuclides than algae and even some higher plants (Zharova, 1961; Morgan, 1961; Timofeyeva-Resovskaya, 1963). In our experiments with a group of natural radioactive elements the accumulation coefficients for ²³⁸U and ²²⁶Ra in higher plants were, on the average, greater than for algae, and only in the case of ²³²Th the relationship was reversed (Iskra *et al.*, 1970b).

The great majority of data reported in the literature on comparative studies of radionuclide accumulation by freshwater hydrobionts were experimentally obtained in laboratory aquariums. In such a simplified model of a waterbody the ecologic regime of hydrobionts is generally very different from the conditions existing in a natural waterbody. Besides, the duration of laboratory experiments is usually short. Allowing for these circumstances it is to be expected that the accumulation coefficients calculated under experimental conditions will not always correspond with those estimated on the basis of radioecologic surveys of natural waterbodies. In an effort to check this presumption the authors, in collaboration with Lyubimova and Fleishman, determined the coefficients of ^{90}Sr and ^{137}Cs accumulation under natural conditions for the principal representatives of the freshwater flora growing in two South Urals lakes (Kulikov *et al.*, 1968a, b, 1971e, f).

The research was conducted in the summer of 1966 at Lakes Bolshoye Miassovo and Bolshoi Tatkul situated on the territory of the Ilmen State Reservation. Sample plants were picked out from the waterbody, rinsed, weighed, exsiccated to the absolutely dry state, and ashed in a muffle furnace at 450 to 500°C. Two 200-liter water samples from each lake were acidified, evaporated, and the dry residue was calcinated at 450 to 500°C. The ^{90}Sr content in the water and plant samples was measured radiometrically against the daughter ^{90}Y . The ^{137}Cs content was determined with a scintillation spectrometer by the procedure described in detail in the publications cited above.

The radionuclide accumulation coefficients calculated in accordance with the data obtained in surveys of natural waterbodies were compared with the coefficients obtained experimentally for the same plant species by Timofeyeva-Resovskaya (1963). In the latter case the water and plants for laboratory experiments were taken from Lake Bolshoye Miassovo.

In the natural waterbodies the accumulation coefficients for ^{90}Sr for all of the plants under study were, on the average, about half those for ^{137}Cs (Table 38). The charophyte group of algae (*Talipellopsis stelligera* in particular) were accumulators of ^{90}Sr , whereas the moss (*Drepanocladus sendtneri*) and the duckweed were ^{137}Cs accumulators.

Accumulation of both radionuclides was found to depend on ecologic peculiarities of plants (Table 39). The highest accumulation coefficients were observed for a group of floating-on-the-water plants, and the lowest coefficients for littoral aquatic and submerged, attached to the bottom plants.

For comparison Fig. 22 shows coefficients of ^{90}Sr accumulation for a number of identical plant species in the experiment and in the natural waterbody. One can note that for charophytes the coefficients of ^{90}Sr accumulation under natural conditions are considerably higher than in the experiment. This can be attributed to the fact that in charophytes ^{90}Sr forms poorly soluble carbonate compounds ($^{90}\text{SrCo}_3$) in quantities directly dependent on the length of time the plants are in contact with ^{90}Sr -containing water. For some plant species a fairly good agreement was noted between the ^{90}Sr accumulation coefficients under

Table 38

Coefficients of ^{90}Sr and ^{137}Cs accumulation by freshwater plants under natural waterbody conditions
(calculated per dry mass)

Plant	Dry weight (% of wet)	Ash (%) content	Accumulation coefficients		Collection site (lake)	Collection date (month)
			⁹⁰ Sr	¹³⁷ Cs		
Algae:						
<i>Nostoc</i> sp.	1	10	1 860	3 310	Bolshoye Miassovo	July
<i>Cladophora fracta</i> Kützing	—	18	1 042	—	Bolshoi Talkul	August
<i>Spirogyra</i> sp.	4	20	286	3 840	B. Miassovo	June
<i>Chara</i> sp.	29	45	1 131	—	"	July
<i>Ch. fragilis</i> Desw.	18	55	1 360	—	"	August
<i>Ch. tomentosa</i> L.	13	30	1 031	804	"	June, August
<i>Nitella hyalina</i> Ag.	4	45	1 014	1 710	"	August
<i>Tolipellopsis stelligera</i> Mig.	—	50	5 555	2 210	"	August
Moss:						
<i>Fontinalis antiperetica</i> L.	—	26	578	2 034	"	July
<i>Drepanocladus sendtneri</i> (Schimp) Warnst	8	10	664	7 730	"	June
Higher plants:						
<i>Lemna minor</i> L.	3	19	511	4 872	B. Talkul	June
<i>Hydrocharis morsus ranae</i> L.	3	20	1 211	2 100	"	July
<i>Stratiotes aloides</i> L.	7	18	589	1 030	B. Miassovo	June-August
<i>Ceratophyllum demersum</i> L.	7	17	301	1 152	"	June, July
<i>Ranunculus circinnatus</i> Sibth.	6	14	345	1 218	"	July
<i>Elodea canadensis</i> L.	11	18	892	472	"	July

<i>Najas flexilis</i> Rosthovius et Schmidt	—	15	268	725	"	August
<i>Myriophyllum spicatum</i> L.	10	11	445	655	"	June, August
<i>Potamogeton lucens</i> L.	17	8	309	257	"	June, July
<i>P. perfoliatus</i> L.	10	10	410	426	"	July
<i>P. compressus</i> L.	15	11	528	1 048	"	July
<i>P. crispus</i> L.	6	15	585	455	"	July
<i>P. natans</i> L.	12	8	333	800	"	June, July
<i>Nymphaea candida</i> Presel.	12	9	150	955	"	July
<i>Nuphar luteum</i> Smith	12	11	100	1 133	"	July
<i>N. pumilum</i> (Hoffman)	12	11	187	348	"	July
<i>Polygonum amphibium</i> L.	—	10	357	930	"	July
<i>Phragmites communis</i> Trinius	40	5	25	285	"	July
<i>Scirpus lacustris</i> L.	23	7	34	—	"	July
<i>Typha latifolia</i> L.	14	7	127	312	"	July
<i>Equisetum heleocharis</i> L.	—	15	273	1 137	"	July
<i>Menyanthes trifoliata</i> L.	11	9	177	675	B. Tatkul	June
<i>Calla palustris</i> L.	9	10	245	715	"	August

Mean:

710

1 770

Table 39

Coefficients of accumulation of ^{90}Sr and ^{137}Cs for plants from various ecological groups (averaged data)

Ecological group	^{90}Sr	^{137}Cs
Plants floating on water surface	860	3 440
Submerged plants not attached to the bottom	670	830
Submerged plants attached to the bottom	420	590
Submerged plants attached to the bottom with leaves floating on water surface	270	790
Littoral aquatic plants	150	600

experimental and natural conditions. On the other hand, some species under experimental conditions had somewhat higher accumulation coefficients than those in the natural waterbody. Such cases are most likely associated with the disturbance of mineral metabolism of plants kept under insufficiently favorable conditions of a laboratory aquarium.

The coefficients of ^{137}Cs accumulation in the natural waterbody for all plant species under study were several times higher than those under experimental conditions (Fig. 23). These differences in the cesium accumulation coefficients may be due to the conversion of a portion of cesium taken up by plant tissues to a nonexchangeable form in a quantity that increases in proportion to the time the plants are exposed to the cesium-containing water (Gileva, 1965; Ivanov *et al.*,

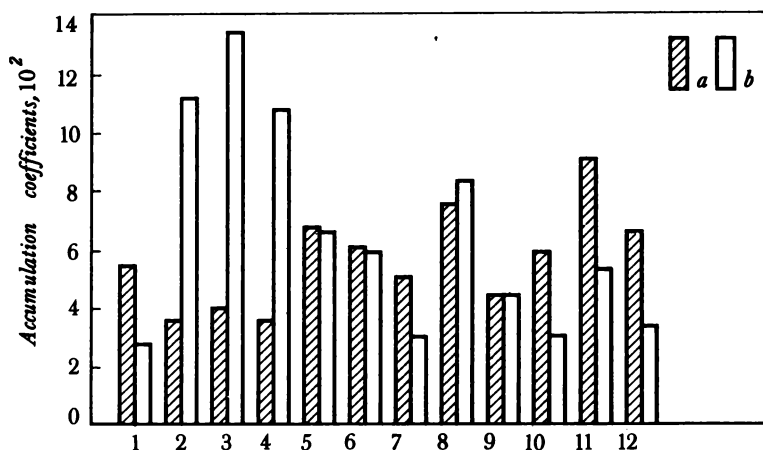


Fig. 22. ^{90}Sr accumulation coefficient for freshwater plants under experimental (a) and natural waterbody (b) conditions. (1) *Spirogyra* sp.; (2) *Chara* sp.; (3) *Chara fragilis*; (4) *Fontinalis* sp.; (5) *Drepanocladus sendtneri*; (6) *Stratiotes aloides*; (7) *Ceratophyllum demersum*; (8) *Elodea canadensis*; (9) *Myriophyllum spicatum*; (10) *Potamogeton perfoliatus*; (11) *Potamogeton compressus*; (12) *Potamogeton natans*.

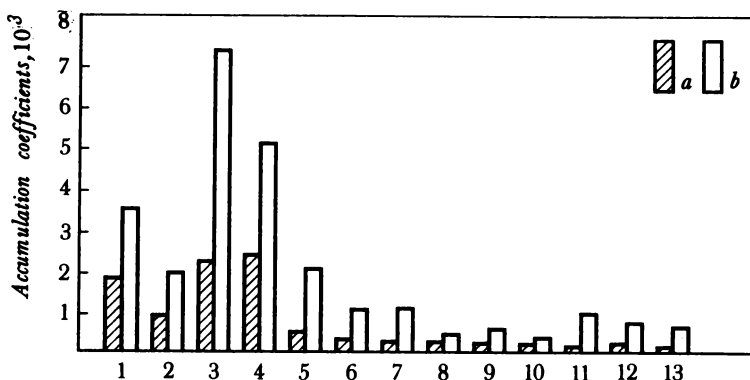


Fig. 23. ^{137}Cs accumulation coefficients for freshwater plants under experimental (a) and natural waterbody (b) conditions. (1) *Spirogyra* sp.; (2) *Fontinalis* sp.; (3) *Drepanocladus sendtneri*; (4) *Lemna minor*; (5) *Hydrocharis morasus ranae*; (6) *Stratiotes aloides*; (7) *Ceratophyllum demersum*; (8) *Elodea canadensis*; (9) *Myriophyllum spicatum*; (10) *Potamogeton perfoliatus*; (11) *Potamogeton compressus*; (12) *Potamogeton natans*; (13) *Calla palustris*.

1965; Kulikov *et al.*, 1968a, 1971e). Under conditions of a short-term laboratory experiment a reduction in the coefficient of cesium accumulation by plants may be expected under the influence of the time factor alone.

Thus, the examples described indicate that the values of the radionuclide accumulation coefficients estimated experimentally under laboratory conditions for hydrobionts may in a number of cases be lower when compared to the same coefficients obtained in a natural waterbody.

Effect of Some Physicochemical Factors of the Medium on the Accumulation of Radionuclides by Hydrobionts

Among a multitude of factors affecting the accumulation of radionuclides by freshwater hydrobionts, the following factors were specially studied: (1) the concentration of isotopic and nonisotopic carriers in the medium; (2) the physicochemical state of radionuclides in water and the pH of the medium; and (3) the water temperature, illumination, and some other factors.

The results of most investigations designed to study the accumulation of radionuclides by hydrobionts as a function of the specific radioactivity of water and the concentration in it of the respective isotopic carriers give ground to believe that in the microconcentrations range (all other things being equal) the concentration of chemical elements in hydrobionts is directly proportional to their concentration in water, and, consequently, the accumulation coefficients remain constant. Within the range of macroconcentrations (beginning with 10^{-5}

and 10^{-4} mole/liter) the reverse relationship is observed between the accumulation coefficients and the concentration of chemical elements in the aqueous medium (Knauss and Porter, 1954; Williams and Swanson, 1958; Timofeyeva-Resovskaya *et al.*, 1959; Titlyanova and Ivanov, 1961; Polikarpov, 1964, 1967; Gileva, 1965; Timofeyeva and Agre, 1965).

Of significant interest for predicting the fate of radionuclides in a waterbody are studies on the accumulation of radionuclides by aquatic organisms as dependent upon the concentration in water of the respective macroelements (the chemical analogs) or, as they are otherwise termed, nonisotopic carriers. It has been demonstrated that the coefficients of ^{90}Sr accumulation are inversely related to the content in water of its chemical analogs calcium and magnesium while the coefficients of ^{137}Cs accumulation are likewise related to the potassium content (Marei *et al.*, 1958; Williams, 1960; Pickering and Lucas, 1962; Kevern *et al.*, 1963; Kevern, 1964; Ivanov *et al.*, 1965; Timofeyeva, 1965; Lebedeva, 1964, 1966; Piskunov *et al.*, 1971a, b). Such pattern of dependence of ^{90}Sr and ^{137}Cs accumulation on the content of their macroanalogs in water indicates a possible cause of the relatively low (as mentioned in the preceding section) accumulation coefficients for these radionuclides in the majority of the hydrobionts under investigation. The cause is apparently a rather high content of calcium and potassium in the water of the reservoirs studied (Table 47).

Inasmuch as calcium and potassium play an important role in the biogeochemical fate of ^{90}Sr and ^{137}Cs , respectively, it is of interest to find out in what quantitative ratios these chemically closely related elements pass from the water medium into plants. In order to evaluate quantitatively the relative uptake by living organisms of ^{90}Sr and calcium, on the one hand, and of ^{137}Cs and potassium, on the other, the so-called discrimination coefficient is used. This coefficient takes account of the difference between quantitative ratios of micro- and macrocomponents in an organism and in the medium from which these components are absorbed.

Discrimination coefficients (C_D) for ^{90}Sr in relation to calcium, and for ^{137}Cs in relation to potassium, vary among hydrobionts within a fairly broad range (Tables 40 and 41). For strontium and calcium the discrimination coefficients fluctuate from 0.11 in spatterdock (*Nuphar luteum*) to 1.9 in frog's-bit (*Hydrocharis morsus ranae*), and for cesium and potassium from 0.05 in naiad (*Najas flexilis*) to 1.2 in moss (*Drepanocladus sendtneri*). Statistical processing of the data obtained yielded evidence of a positive correlation between the concentration in plants of calcium and the coefficients of ^{90}Sr accumulation, as well as between the concentration of potassium and the coefficients of ^{137}Cs accumulation (Levina and Lyubimova, 1971; Lyubimova, 1971a).

Thus, when the two radionuclides are absorbed from the aqueous medium, their discrimination coefficients with respect to their chemical macroanalogs are determined by biological features of plants. Species containing much calcium tend to take up more ^{90}Sr , whereas species accumulating much potassium tend to have higher accumulation

Table 40

Calcium concentration in freshwater plants, accumulation coefficients (C_A) for ^{90}Sr and discrimination coefficients (C_D) for ^{90}Sr with respect to calcium under natural conditions

Plant	Ca (mg/gm dry matter)	C_A for ^{90}Sr	C_D for $^{90}\text{Sr}/\text{Ca}$
<i>Cladophora fracta</i>	19.6	1042	0.90
<i>Spirogyra</i> sp.	15.2	286	0.45
<i>Chara</i> sp.	98.6	1131	0.27
<i>Ch. tomentosa</i>	90.6	1031	0.29
<i>Ch. fragilis</i>	144.6	1360	0.23
<i>Nitella hyalina</i>	96.0	1014	0.25
<i>Fontinalis antipetrica</i>	8.6	578	1.50
<i>Drepanocladus sendneri</i>	12.2	664	1.30
<i>Lemna minor</i>	12.4	511	1.14
<i>Hydrocharis morsus ranae</i>	13.5	1211	1.90
<i>Stratiotes aloides</i>	19.4	589	0.73
<i>Ceratophyllum demersum</i>	10.0	301	0.73
<i>Elodea canadensis</i>	25.3	892	0.88
<i>Ranunculus circinatus</i>	13.0	345	0.64
<i>Najas flexilis</i>	8.2	268	0.78
<i>Myriophyllum spicatum</i>	17.2	445	0.62
<i>Potamogeton lucens</i>	13.9	309	0.53
<i>P. perfoliatus</i>	16.2	410	0.60
<i>P. compressus</i>	17.0	528	0.74
<i>P. crispus</i>	12.8	585	1.08
<i>P. natans</i>	9.5	333	0.84
<i>Nymphae candida</i>	9.6	150	0.36
<i>Nuphar luteum</i>	15.3	100	0.11
<i>N. pumilum</i>	14.0	99	0.19
<i>Polygonum amphibium</i>	17.5	357	0.49
<i>Scirpus lacustris</i>	2.3	34	0.35
<i>Typha latifolia</i>	5.2	127	0.58
<i>Phragmites communis</i>	1.5	25	0.39
<i>Equisetum heleoharis</i>	14.2	273	0.46
<i>Menyanthes trifoliata</i>	6.9	177	0.70
<i>Calla palustris</i>	7.4	245	0.57

coefficients for ^{137}Cs . Our additional experiments have demonstrated that the discrimination coefficient of ^{90}Sr does not depend on the calcium concentration in water over a wide range of values (Table 42).

The data from the literature attest to the fact that the accumulation of radionuclides by freshwater organisms is also defined by the physico-chemical state of nuclides in a solution and by the concentration of hydrogen ions in it. Thus, in the course of studies of ^{91}Y accumulation by filamentous algae (*Cladophora fracta* Kützinger) a twofold reduction in the nuclide accumulation was noted in the range of aqueous solution pH values in which yttrium changes to the colloidal form (Gileva, 1965). It has also been established that radioisotopes of a number of chemical elements present in a solution in the form of chelate compounds are accumulated by hydrobionts in lesser amounts than the same radioisotopes introduced into the medium in the ionic form (Timofeyeva-Resovskaya, 1963).

Table 41

Potassium concentration in freshwater plants, accumulation coefficients (C_A) for ^{137}Cs and discrimination coefficients (C_D) for ^{137}Cs with respect to potassium under natural conditions

Plant	K (mg/gm dry matter)	C_A for ^{137}Cs	C_D for $^{137}\text{Cs/K}$
<i>Spirogira</i> sp.	34.5	3 840	0.50
<i>Chara tomentosa</i>	17.7	804	0.20
<i>Ch. fragilis</i>	108.7	10 150	0.42
<i>Nitella hyalina</i>	47.4	1 710	1.10
<i>Drepanocladus sendtneri</i>	29.4	7 730	1.20
<i>Fontinalis antiperetica</i>	13.7	2 034	0.63
<i>Lemna minor</i>	33.0	2 541	0.26
<i>Hydrocharis morsus ranae</i>	63.8	2 100	0.11
<i>Stratiotes aloides</i>	27.5	430	0.07
<i>Ceratophyllum demersum</i>	51.3	993	0.09
<i>Elodea canadensis</i>	46.5	820	0.08
<i>Ranunculus circinatus</i>	29.1	625	0.10
<i>Najas flexilis</i>	65.5	725	0.05
<i>Myriophyllum spicatum</i>	11.3	655	0.30
<i>Potamogeton lucens</i>	20.6	181	0.04
<i>P. perfoliatus</i>	26.0	426	0.07
<i>P. compressus</i>	21.9	1 048	0.21
<i>P. crispus</i>	38.1	455	0.07
<i>P. natans</i>	12.0	797	0.30
<i>Nuphar luteum</i>	23.1	841	0.16
<i>N. pumilum</i>	23.1	556	0.10
<i>Polygonum amphybium</i>	17.6	930	0.24
<i>Typha latifolia</i>	30.1	312	0.05
<i>Phragmites communis</i>	10.2	260	0.11
<i>Equisetum heleoharis</i>	48.2	3 000	0.21
<i>Menyanthes trifoliata</i>	23.9	675	0.10
<i>Calla palustris</i>	25.9	715	0.10

Table 42

Dependence of ^{90}Sr accumulation coefficient (C_A) and discrimination coefficient (C_D) for water milfoil (*Myriophyllum spicatum* L.) on calcium concentration in water

Calcium concentration in water (mg/liter)	C_A for calcium	C_A for ^{90}Sr	C_D for $^{90}\text{Sr/Ca}$
19.7	1 432	1 012	0.7
26.2	1 292	754	0.6
43.7	631	519	0.8
83.4	327	292	0.9
128.8	264	168	0.6
191.0	205	140	0.7
226.2	165	95	0.6
287.2	134	75	0.5
331.3	106	75	0.7
633.2	75	42	0.5

On the whole it should be noted that the problems of the accumulation of radionuclides by freshwater organisms as related to the physicochemical state and the form of these radionuclides in the aqueous medium are far from being adequately studied, because of the lack of information on the state of radionuclides in water when they are taken up by hydrobionts. Still less well studied until now have been the regularities governing the accumulation of radionuclides by freshwater organisms as dependent on such factors as water temperature and illumination conditions.

In experiments with the developing eggs of a pike and a perch we found that at water temperatures of 10°C and 20°C the accumulation coefficients for ^{90}Sr did not change, although the rate of embryos development (till the stage of hatching) at 20°C was doubled. At the same time the accumulation coefficients for ^{90}Y at the end of incubation period at 20°C were twice as high in the pike and almost three times as high in the perch as those observed at 10°C (Figs. 24 and 25). The accumulation of radionuclides by larvae of the two fish species during the first 4 to 5 days following hatching was approximately the same at both temperatures, but later there was a drastic increase in the radionuclide accumulation at 20°C (Figs. 26 and 27).

Foster and Davis (1956) recorded a reduction in the accumulation of radioactive substances by fish under natural conditions in cold periods on the Columbia River. They attributed this to a lesser consumption by fish of radioactive vegetable food in cold seasons. On the other hand in experiments with freshwater plankton at water temperatures from 13.5 to 28.5°C, a complete lack of dependence of ^{85}Sr accumulation on water temperature was observed, and it was only at 28.9°C that an appreciable increase in the accumulation of this radionuclide was recorded (Patten and Iverson, 1966). Unfortunately, we failed to find in the literature available to us any other information on the influence of temperature on the processes of radionuclide accumulation by freshwater organisms.

As to the effect of light on the absorption of radionuclides by hydrobionts, the literature pertinent to the subject is practically lacking and the available fragmentary information is rather contradictory. For example, Patten and Iverson (1966) noted an increase in the accumulation of ^{85}Sr by freshwater plankton in the presence of light, while in darkness this process was brought to a standstill. At the same time in experiments with freshwater macrophytes no relationship was found between the accumulation of radioactive strontium and illumination conditions (Owens *et al.*, 1961; Agre and Telitchenko, 1963).

Of special interest is a comparative investigation of the accumulation of radionuclides by hydrobionts in running and nonrunning waterbodies. It is regrettable that so far very few data on this subject are to be found in the literature. The available material indicates, however, that in a running waterbody the accumulation coefficients for cobalt, zinc, zirconium, niobium, and ruthenium radionuclides in a number of aquatic plants attained lower levels than in a nonrunning waterbody during the same length of time (Ivanov, 1965a). Ivanov accounts for this by the fact that in a running waterbody no balance is

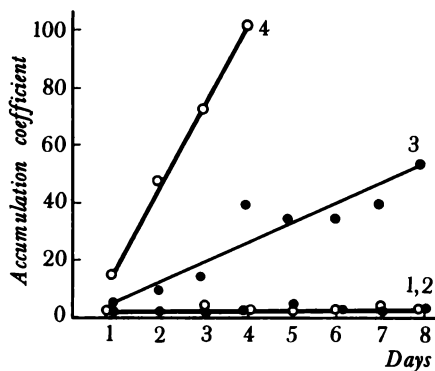


Fig. 24. Dynamics of ^{90}Sr and ^{90}Y accumulation by developing pike eggs at different water temperatures. Strontium accumulation at 10°C (1) and 20°C (2); yttrium accumulation at 10°C (3) and 20°C (4).

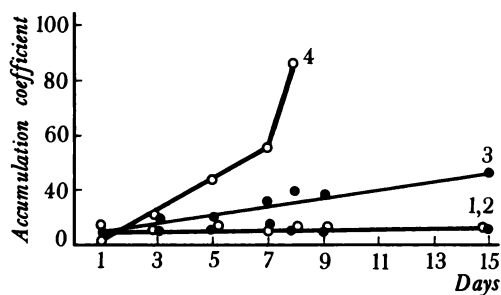


Fig. 25. Dynamics of ^{90}Sr and ^{90}Y accumulation by developing perch eggs at different water temperatures. Strontium accumulation at 10°C (1) and 20°C (2); yttrium accumulation at 10°C (3) and 20°C (4).

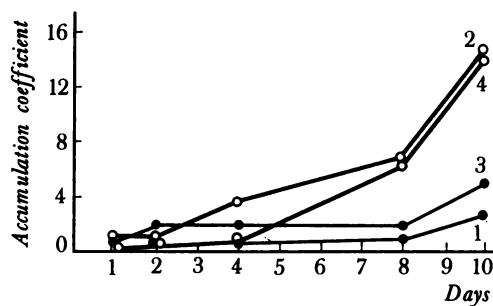


Fig. 26. Dynamics of ^{90}Sr and ^{90}Y accumulation by pike larvae at different water temperatures. Strontium accumulation at 10°C (1) and 20°C (2); yttrium accumulation at 10°C (3) and 20°C (4).

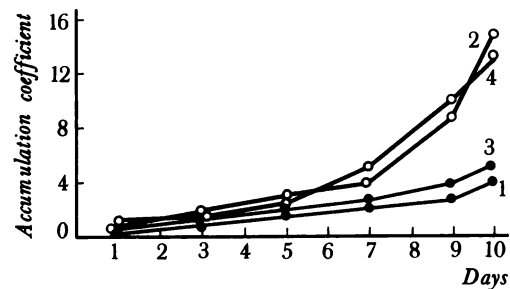


Fig. 27. Dynamics of ^{90}Sr and ^{90}Y accumulation by perch larvae at different water temperatures. Strontium accumulation at 10°C (1) and 20°C (2); yttrium accumulation at 10°C (3) and 20°C (4).

reached in the distribution of radionuclides over the water, ground, and plant biomasses.

Leinerte (1969, 1970) studied the accumulation of fission fragment radionuclides (^{90}Sr , ^{137}Cs , and ^{144}Ce) by three species of freshwater plants following the irradiation of these plants with ^{60}Co gamma-rays, at doses over the range from 2.5 to 32 kilorad. It was found that the accumulation coefficient for ^{90}Sr changed but negligibly even when the highest radiation dose was applied. With an increase in the radiation dose a sharp decline in ^{137}Cs accumulation was observed, whereas for ^{144}Ce the accumulation coefficients increased following high-dose irradiation.

In laboratory experiments with a filamentous alga (*Zygnema sp.*) a reduction in the accumulation of ^{90}Sr and ^{137}Cs was recorded only during the first 24 hr after irradiation (in doses of 25, 50, and 75 kilorad). Subsequently, with recovery of algae from the radiation injury, the coefficients of radionuclide accumulation returned to the control level (Lyubimova and Famelis, 1970).

Migration of ^{90}Sr and ^{137}Cs from Fish in the Spawning Period

In the last few years the issue of the accumulation of radionuclides in fish, as well as their distribution over individual organs and tissues, has drawn the attention of a large number of investigators. They usually consider either the aqueous medium or food, or both together, as sources of radionuclide uptake by fish (Marei, 1955; Krumholz *et al.*, 1957; Saurov, 1957; Foster and Davis, 1956; Agranat, 1958; Rudakov, 1958; Lebedeva, 1961, 1962; Rosenthal, 1960; Ilyin and Moskalev, 1961; Häsänen and Miettinen, 1963; Polikarpov, 1964; Kolehmainen *et al.*, 1966; Ophel and Judd, 1967; Fleishman, 1971a, b). There is another pathway of radioactive contamination of fish: direct transfer of radionuclides from the contaminated female fish to her progeny either directly (in viviparous fish) or via her spawn (in spawning fish).

One can find in the literature quantitative evaluations of the passage of radionuclides from pregnant females to the offspring in mammals (Kulikova, 1964, 1970; Vokken, 1967; Buldakov and Moskalev, 1968), but no similar data have so far been available for fish. At the same time, considering that the spawn mass given off by the organism during spawning in many fish species constitutes a fairly high percentage of the weight of spawning females, the transfer of radionuclides to the progeny may be regarded as a pathway of biological migration of radionuclides in an aquatic biogeocenosis.

The present section covers the results of our investigations into the migration of ^{90}Sr , ^{137}Cs , and their chemical analogs (calcium, magnesium, and potassium) together with fish roe in the spawning period (Kulikov *et al.*, 1971c, d).

Two fish species, studied in 1969, were tench (*Tinca tinca* L.) and pike (*Esox lucius* L.).

Male and female reproductive fish aged 5 to 7 years were caught in spawning areas of Lake Bolshoye Miassovo and transported to the laboratory where carcasses (minus the gastric contents) and spawn were weighed. Thereupon three specimens of each species and roe portions (three 1 kg samples of the wet mass) were ashed in a muffle furnace at a temperature of 450 to 500°C. The spawn was taken from 10 to 15 females and one composite sample was prepared by mixing. The ^{90}Sr content in ashed residues of fish carcasses and in the spawn was determined radiochemically according to the daughter ^{90}Y data; ^{137}Cs was determined using an АИ-100-1 gamma-spectrometer with a 40×40 mm scintillation crystal NaI(Tl) as a detector; calcium and magnesium were measured complexometrically (Arinushkina, 1961), and potassium was measured with the aid of a ППФ-УНИИЗ flame photometer.

Table 43 shows characteristics of female tench and pike studied, including the levels of the radionuclides and their chemical analogs in their bodies. One can see that in the two fish species the amount of ^{137}Cs exceeds that of ^{90}Sr and the content of calcium is higher than that of potassium and magnesium. The total ash weight of the tench carcass is somewhat lower than that of the pike, while the ash weight of fish roe for the two species is approximately the same.

Differences in the accumulation of individual chemical elements by various fish species are particularly evident from analyses of accumulation coefficients (Table 44). It may be noted that almost all chemical elements under investigation are accumulated by the pike more vigorously than by the tench. Potassium is an exception and its accumulation coefficients for the two fish species are similar.

An increased accumulation of a number of chemical elements and their radioactive isotopes in predatory fish, as compared to benthophages and plankton-eaters, has also been recorded by other investigators (Agnedal *et al.*, 1958; Kolehmainen *et al.*, 1966). Presumably these differences may be accounted for by properties of the feed consumed.

One can also see from Table 44 that in the two species of fish on the average higher accumulation coefficients were recorded for the alkaline elements (^{137}Cs and potassium) as compared with the alkaline earth elements (^{90}Sr , potassium, and magnesium). Especially low, when compared to other elements, were the accumulation coefficients for ^{90}Sr , and also for ^{90}Sr and calcium in the spawn. It is known that calcium and strontium are skeletal elements, and that they therefore enter into the spawn in relatively smaller amounts than cesium, potassium, and magnesium.

The relative shift of analogous chemical elements when these are taken up by organisms from the environment is expressed by the discrimination coefficient (C_D) calculated, for instance, for the strontium-calcium pair according to the formula:

$$C_D = \frac{{}^{90}\text{Sr}/\text{Ca (in organism)}}{{}^{90}\text{Sr}/\text{Ca (in water)}}$$

Table 43
Content of ^{90}Sr , ^{137}Cs , and their chemical analogs in female tench and pike at spawning

No. of specimens	Date of catch	Weight of fresh fish with spawn (gm)	Spawn weight (% of total weight)	Ash content (% of total weight)		Content of chemical elements in a single female fish with spawn					
				Carcass	Spawn	⁹⁰ Sr	¹³⁷ Cs	K (mg)	Ca (mg)	Mg (mg)	
						$n \times 10^{-12}$ Ci	$n \times 10^{-12}$ Ci				
Tench											
1	July 2, 1969	1 379.6	14.1	2.8	1.8	179	1 840	3 240	7 113	3 156	
2	"	1 205.2	12.8	2.8		408	1 102	2 806	4 736	2 391	
3	"	1 004.4	14.8	2.9		165	1 012	—	—	—	
Pike											
1	April 24, 1969	2 080	22.1	3.5	1.7	500	7 120	5 360	12 066	5 482	
2	"	2 400	20.7	4.7		683	9 970	7 870	14 554	10 016	
3	"	2 420	11.1	4.1		451	8 050	6 239	17 246	3 395	

Table 44
Accumulation coefficients for ^{90}Sr , ^{137}Cs , and their chemical analogs in tench and pike (calculated per wet mass)

Element	Tench			Pike		
	Males	Females	Spawn	Males	Females	Spawn
^{90}Sr	49 ± 20	39 ± 16	5 ± 0.4	77 ± 7	57 ± 12	7 ± 1
^{137}Cs	290 ± 42	426 ± 90	233 ± 6	1 873 ± 458	1 360 ± 209	1 036 ± 130
Calcium	378 ± 70	240 ± 25	2 ± 0.9	423 ± 42	330 ± 15	7 ± 2
Magnesium	230 ± 21	210 ± 46	114 ± 15	431 ± 153	330 ± 179	100 ± 5
Potassium	385 ± 13	415 ± 75	496 ± 45	380 ± 13	410 ± 76	493 ± 50

Table 45**Discrimination coefficients for chemical elemental analogs entering**

Pairs of elemental analogs	Tench		
	Males	Females	Spawn
$^{90}\text{Sr}/\text{Ca}$	0.13 ± 0.05	0.16 ± 0.02	2.70 ± 0.1
$^{90}\text{Sr}/\text{Mg}$	0.22 ± 0.11	0.21 ± 0.13	0.04 ± 0.0
Mg/Ca	0.62 ± 0.12	0.90 ± 0.16	64.00 ± 23.0
$^{137}\text{Cs}/\text{K}$	0.80 ± 0.10	1.00 ± 0.25	0.46 ± 0.03

The tench and the pike, both male and female, absorb less ^{90}Sr than calcium or magnesium (Table 45). The values of the discrimination coefficients for the pairs $^{90}\text{Sr}/\text{Ca}$ and $^{90}\text{Sr}/\text{Mg}$ are about 0.2. Magnesium and calcium pass into the two fish species in about the same ratio that these two elements have in the aqueous medium and their discrimination coefficients are close to unit. The ratio of the pair $^{137}\text{Cs}/\text{K}$ in the tench is the same as that in water, whereas in the pike the amount of ^{137}Cs is 3 to 5 times greater than that of potassium.

The amount of ^{90}Sr in tench spawn is about 3-fold that of calcium, while in pike spawn the ratio $^{90}\text{Sr}/\text{Ca}$ is the same as that in water. In the spawn of the two fish species a strong discrimination of ^{90}Sr and calcium in relation to magnesium is noted; the amount of ^{137}Cs in relation to potassium as accumulated in tench spawn is half as great, while in pike spawn ^{137}Cs exceeds potassium by a factor of 2.

Since chemical elements enter into the spawn directly from the female fish itself and not from the environment, it will be more appropriate to calculate discrimination coefficients for the female/spawn system (Table 46) than for the water/spawn system, as presented in Table 45. In the spawn of the two fish species the amount of ^{90}Sr , if compared to its content in the carcasses of mature fish, is higher than that of calcium, but approximately 3 times less than that of magnesium. The latter passes into the spawn from the female tench and pike in amounts exceeding that of calcium 75 and 15 times, respectively. The

Table 46**Discrimination coefficients for chemical elemental analogs upon their passage from female fish into spawn**

Pairs of elemental analogs	Tench	Pike
$^{90}\text{Sr}/\text{Ca}$	15.9 ± 10.7	4.0 ± 1.1
$^{90}\text{Sr}/\text{Mg}$	0.3 ± 0.2	0.3 ± 0.1
Mg/Ca	75.4 ± 31.7	14.8 ± 6.8
$^{137}\text{Cs}/\text{K}$	0.3 ± 0.1	0.6 ± 0.02

Pike		
Males	Females	Spawn
0.17 ± 0.04	0.17 ± 0.04	1.00 ± 0.30
0.20 ± 0.08	0.20 ± 0.08	0.06 ± 0.004
1.00 ± 0.46	1.00 ± 0.42	15.40 ± 7.12
4.80 ± 1.26	3.30 ± 0.12	2.10 ± 0.17

discrimination coefficient for ^{137}Cs and potassium passing into tench spawn is 0.3, and into pike spawn 0.6.

The data presented on the discrimination coefficients demonstrate that despite the chemical affinity of elemental analogs, their uptake from the aqueous medium by fish, as well as their transition to the spawn of the female fish, is significantly different. The degree of discrimination of elemental analogs in their passage from one link to another is defined by the properties of chemical elements and also by the specific biological features of fish. This latter circumstance was noted earlier with respect to plants (Timofeyeva and Kulikov, 1968). A strong discrimination of ^{90}Sr in relation to potassium in the water/fish system may be partially attributed to the probability that these elements have not yet attained equilibrium in this particular system.

Knowing the female fish weight at spawning, the weight of spawn, and the content of chemical elements, one can evaluate quantitatively the amount of radionuclides and their chemical analogs passing from female fish into the environment with the spawn. The alkaline elements, which in the animal are accumulated predominantly in the soft and developing tissues are excreted with the spawn in an appreciably greater amount

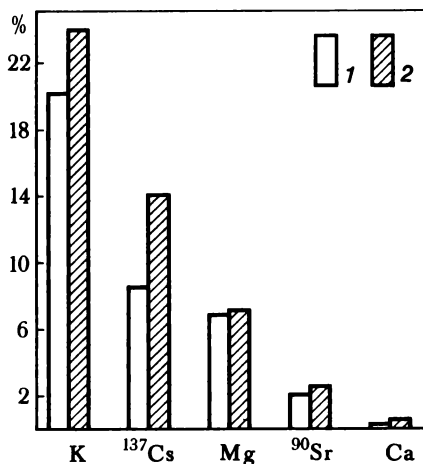


Fig. 28. Excretion of chemical elements from tench (1) and pike (2) females at spawning (% of total body content).

than the alkaline-earth elements that are accumulated chiefly in the skeleton (Fig. 28). The percentage of nearly all elements excreted by the pike is higher than that excreted by the tench. On the average, the female tench and pike studied excreted with the spawn: 20% and 24%, respectively, of potassium, 8% and 14%, respectively, of ^{137}Cs , about 7% of magnesium, 2 to 2.5% of ^{90}Sr , and some decimal fractions of one percent of calcium.

Excretion of chemical elements with the spawn by fish at spawning may be considered on the one hand as a peculiar pathway of the migration of these elements in a waterbody, and on the other, as a means for self-purification of the fish from the incorporated radionuclides. The role of such self-purification may be particularly important when fish from waterbodies containing radioactive substances migrate into "pure" spawning ponds, or in the case of fish species that have several spawning cycles during one season.

Accumulation of ^{90}Sr and ^{137}Cs by Freshwater Plants as Depending on Specific Type of Waterbody (Trophicity)

It was demonstrated in the first section of this chapter that the accumulation of radionuclides by hydrobionts depends on a multitude of factors, which, in different combinations, characterize different types of waterbodies as places of habitation. Lake-type waterbodies have particularly distinct physicochemical and biological differences (Zernov, 1934; Konstantinov, 1967; Kuznetsov, 1970). Therefore under natural conditions the radionuclide accumulation coefficients for freshwater organisms, as distinct from marine organisms, vary over a rather wide range (Ophel, 1963; Fleishman, 1968; Agnedal, 1967; Kalninya, 1970).

We have carried out in our laboratory investigations with the objective of determining the coefficients of accumulation of ^{90}Sr and ^{137}Cs by freshwater plants in lakes of varying trophicity (Lyubimova, 1971a). The objects of investigation were the two lakes mentioned above: Lake Bolshoye Miassovo and Lake Bolshoi Tatkul. Lake Bolshoye Miassovo occupies a large area and is fairly deep. Its shores are predominantly rocky while the vegetation is encountered on isolated sites along the shoreline. According to the accepted typology, the lake can be referred to as being of the oligomesotrophic type (Rossolimo, 1967). The second lake is overgrown with algae and is almost five times smaller than the first lake, its maximum depth not exceeding 3 meters. The shores are boggy and the abundant vegetation extends practically throughout the whole depth of the water. During the summer months the concentration of the major macroelements (potassium, sodium, calcium, and magnesium) in the water of Lake Bolshoye Miassovo is considerably higher than that in Lake Bolshoi Tatkul; the same holds true with regard to the content of strontium and cesium radionuclides (Table 47). A set of indices warrants assignment of Lake Bolshoi Tatkul to the mesodystrophic type.

Table 47

Chemical composition of water and morphometric characteristics of Lakes Bolshoye Miassovo and Bolshoi Tatkul (July, 1968)

Index	Bolshoye Miassovo, oligomesotrophic lake	Bolshoi Tatkul, mesodystrophic lake
Area (km ²)	11.5	2.5
Average depth (m)	10.9	1.9
Maximal depth (m)	22.5	3.0
Clarity (m)	4-5	1-2
Water pH	8.3	7.3
Dry residue (mg/liter)	177.2	159.2
K ⁺ , mg/liter	6.6	3.3
Na ⁺ , mg/liter	15.5	9.0
Ca ⁺⁺ , mg/liter	23.0	19.9
Mg ⁺⁺ , mg/liter	9.6	7.0
SO ₄ ⁻ , mg/liter	19.3	10.4
CO ₃ ⁻ , mg/liter	3.0	2.8
HCO ₃ ⁻ , mg/liter	128.2	125.1
Cl ⁻ , mg/liter	20.8	15.2
Free CO ₂	Absent	Absent
Oxidizability (mg O ₂ per liter water)	3.5	6.6
⁹⁰ Sr ($n \times 10^{-12}$ Ci/liter)	5.1	4.1
¹³⁷ Cs ($n \times 10^{-12}$ Ci/liter)	2.8	1.7

The values of the accumulation coefficients of both strontium and, especially, cesium for the plants of Lake Bolshoi Tatkul are noticeably higher than those for the plants of Lake Bolshoye Miassovo (Figs. 29 and 30). Similar data have been obtained in laboratory experiments with two species of plants and with the water taken from the two lakes (Fig. 31). We assume that one of the main causes of the reduced accumulation of radionuclides by the plants in Lake Bolshoye Miassovo, as compared to the plants of Lake Bolshoi Tatkul, is a higher content of potassium, sodium, calcium, and magnesium salts in the water of this lake (Table 47). As indicated earlier, an increase in the concentration of these elements in water leads to a reduction in the accumulation of radionuclides by hydrobionts.

During the summer months the hydrochemical and ecological regime of Lake Bolshoye Miassovo is more stable than that of the dystrophic Lake Bolshoi Tatkul. This is shown by the data on the change of calcium and magnesium concentrations in the water of the lakes under study, as well as by the change in the coefficients of accumulation of ⁹⁰Sr by plants during the summer months (Figs. 32 and 33). In the water of Lake Bolshoye Miassovo the calcium and magnesium concentrations remain during the summer at almost the same level,

whereas in Lake Bolshoi Tatkul rather substantial concentration shifts occur in July and August when plants extend throughout the lake water depth. Accordingly, the ^{90}Sr accumulation coefficients for the plants growing in Lake Bolshoye Miassovo are more stable with time than in the latter lake. The highest accumulation coefficients are registered at the height of vegetation (July and August). The reduction in the accu-

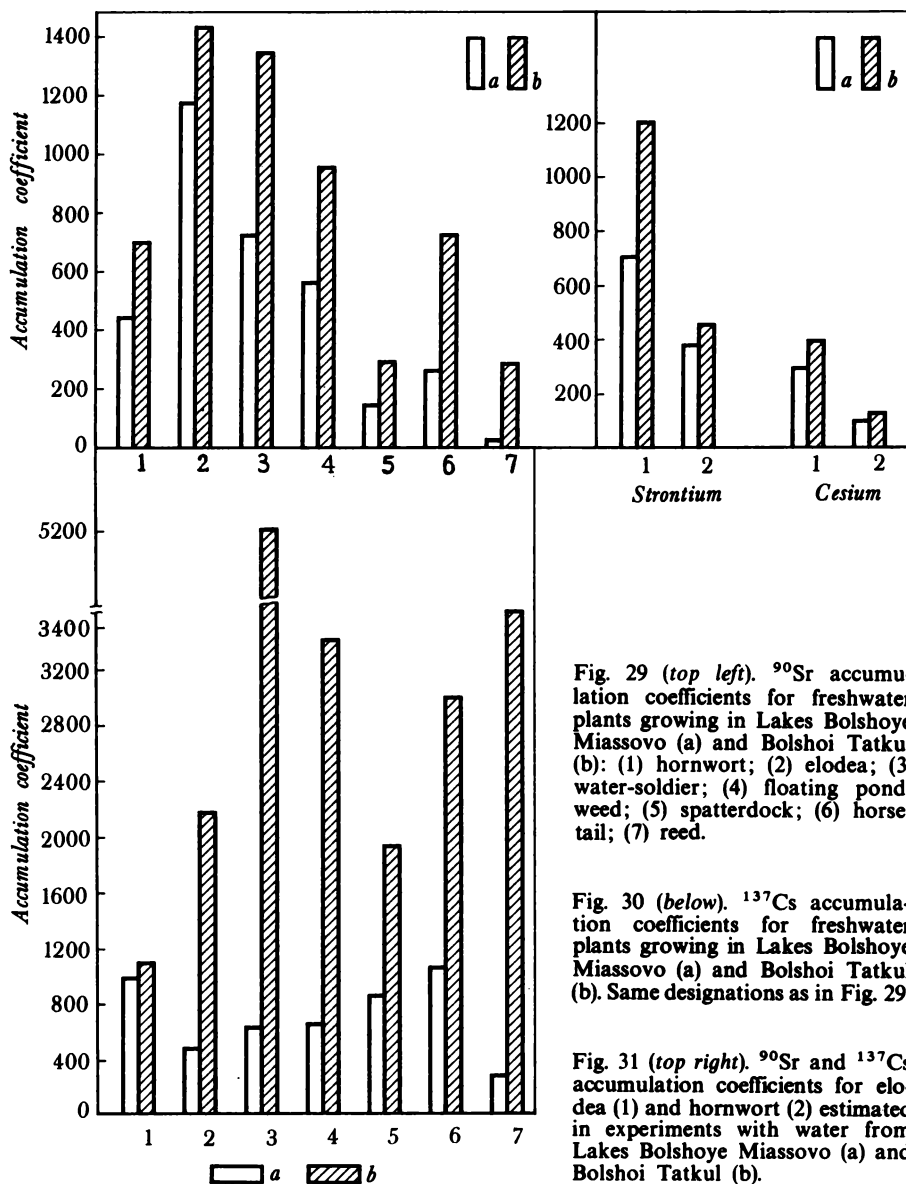


Fig. 29 (top left). ^{90}Sr accumulation coefficients for freshwater plants growing in Lakes Bolshoye Miassovo (a) and Bolshoi Tatkul (b): (1) hornwort; (2) elodea; (3) water-soldier; (4) floating pondweed; (5) spatterdock; (6) horsetail; (7) reed.

Fig. 30 (below). ^{137}Cs accumulation coefficients for freshwater plants growing in Lakes Bolshoye Miassovo (a) and Bolshoi Tatkul (b). Same designations as in Fig. 29.

Fig. 31 (top right). ^{90}Sr and ^{137}Cs accumulation coefficients for elodea (1) and hornwort (2) estimated in experiments with water from Lakes Bolshoye Miassovo (a) and Bolshoi Tatkul (b).

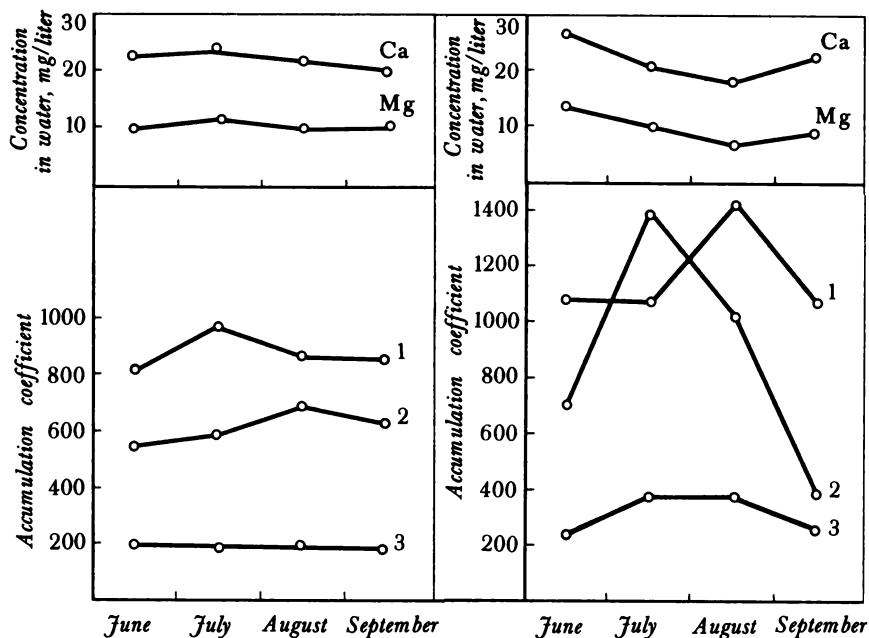


Fig. 32 (left). Dynamics of changes in concentrations of calcium and magnesium in water of Lake Bolshoye Miassovo during summer and ^{90}Sr accumulation coefficients for plants: (1) water-soldier; (2) floating pondweed; (3) spatterdock.

Fig. 33. Dynamics of changes in concentration of calcium and magnesium in water of Lake Bolshoi Tatkul during summer and ^{90}Sr accumulation coefficients for plants. Same designations as in Fig. 32.

mulation coefficients by the end of the season (September) appears to be associated with a partial extinction of plants and the sinking of their older portions, having the highest ^{90}Sr content, to the lake bottom.

Fluctuations in the potassium and sodium content in the water of the two lakes are less pronounced. The ^{137}Cs accumulation coefficients for the plants from Lake Bolshoye Miassovo increase until September, whereas in Lake Bolshoi Tatkul in September some plant species already show a reduction in ^{137}Cs accumulation (Figs. 34 and 35). This observation can be explained by the fact that the plants in the dystrophic Lake Bolshoi Tatkul begin to die off earlier than in Lake Bolshoye Miassovo.

The Role of Freshwater Plants in the Cosedimentation of ^{90}Sr with Calcium Carbonates

Formation of carbonate is one of the most important mechanisms of transfer of calcium from the water to the bottom deposits of a waterbody (Vinogradov, 1932; Isachenko, 1951; Strakhov *et al.*, 1954;

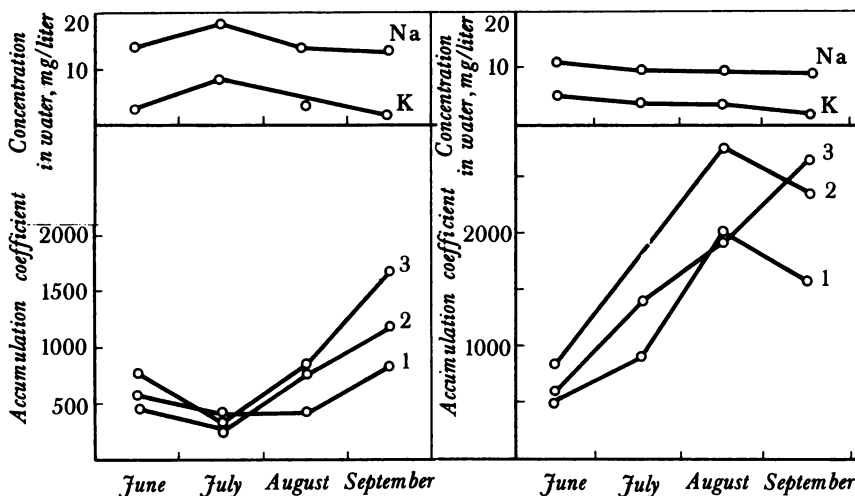


Fig. 34 (left). Dynamics of changes in concentrations of potassium and sodium in water of Lake Bolshoye Miassovo during summer and ^{137}Cs accumulation coefficients for plants. Same designations as in Fig. 32.

Fig. 35. Dynamics of changes in concentration of potassium and sodium in water of Lake Bolshoi Tatkul during summer and ^{137}Cs accumulation coefficients for plants. Same designations as in Fig. 32.

Strakhov, 1960). In freshwater reservoirs the formation of calcium carbonate sediments is closely associated with the vital activity of aquatic plants (Kuznetsov, 1970). Thus, for instance, elodea brakes can excrete in one day as many as 2 kg of lime calculated per 100 kg of elodea dry mass (Voronikhin, 1953). Considering that the biomass of the carbonate-forming plants reaches 3 to 5 tons/hectare (Guseva and Ekzertsev, 1965), it is easy to estimate what a huge amount of calcium can be extracted from the aqueous medium and deposited in the form of slightly soluble sediments on the bottom of a waterbody. Of particular importance in the processes of carbonate formation is the role of charophytes (Abrosova, 1959). The calcium carbonate content in the ooze beneath these charophytes may be as high as 72.4% (Strakhov, 1960).

^{90}Sr , being the chemical analog of calcium, becomes incorporated in the same links of the biochemical migration chain as calcium. For this reason the stable calcium isotope may be regarded as a nonisotopic macrocarrier of ^{90}Sr . The latter is involved with calcium in the carbonate formation process, passing from water-dissolved compounds into a slightly soluble sediment.

We have run in our laboratory (Kulikov *et al.*, 1970a) a series of tests for the quantitative study of co-sedimentation of ^{90}Sr and calcium carbonates formed on surface of algae. Our investigations were conducted both under laboratory conditions in an aquarium and with the use of the material collected in a natural waterbody (Lake Bolshoye Miassovo).

The experimental procedure was described in detail by Timofeyeva

(1965) and also by Timofeyeva and Kulikov (1968). Calcium in water, plants, and carbonate sediments was determined complexometrically (Arinushkina, 1961). ^{90}Sr in the samples collected in a natural waterbody was measured radiochemically according to the daughter ^{90}Y data.

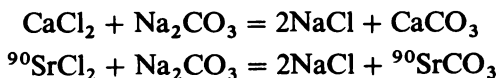
It can be seen from the data in Table 48 that the concentration of ^{90}Sr in the carbonate sediment formed on algae exceeds that in the plant tissues by almost one order of magnitude. The charophytes concentrate strontium much more vigorously than do other plant species, probably owing to a greater ability of these algae to form carbonates in their tissues.

To assess quantitatively the ratios of ^{90}Sr to calcium in which they pass from water into the carbonate sediment resulting from the vital activity of plants, special experiments were performed (Table 49). It was found that the discrimination coefficient (C_D) of ^{90}Sr calculated from the

ratio $C_D = \frac{^{90}\text{Sr}/\text{Ca (in carbonate sediment)}}{^{90}\text{Sr}/\text{Ca (in water)}}$ is the same for all carbonates

collected among plants of various species; it does not depend on the calcium concentration in water and its mean value is about 0.3. Approximately the same C_D was obtained following determination of the ^{90}Sr and calcium content in water and in carbonates collected from plants of two species in a natural waterbody. Consequently, regardless of the water content of calcium and the species of plants in which carbonation occurs, the relative (as compared to water) concentration of ^{90}Sr in carbonate sediment is about one-third that of calcium. At the same time the C_D of ^{90}Sr relative to calcium when these two elements enter directly from the aqueous medium into the plant tissues is highly variable for different plant species (Table 48).

Since the C_D for ^{90}Sr in carbonates does not depend on biological features of plants, one can assume that the process of carbonate formation, being in the final analysis a result of the vital activity of the plant, occurs not within the plant organism but at the plant/water interface. This assumption is supported by data on the chemical co-sedimentation of carbonates from calcium and ^{90}Sr chloride solutions in the following reactions:



It was found that in the carbonate sediment obtained in this manner, the C_D of ^{90}Sr relative to calcium was 0.33, i.e., close to that observed during the carbonate sediment formation due to the vital activity of plants. Approximately the same C_D value was obtained when calculated from the data on the contents of stable strontium and calcium in seawater and in the carbonates of sea silt (Baranova and Polikarpov, 1968).

Although the mechanism of carbonate formation with the participation of aquatic plants has not yet been completely studied, there

Table 48

⁹⁰Sr concentration in aquatic plants and carbonate sediment formed on their surface under natural waterbody conditions

Plant	$n \times 10^{-9}$ Ci/kg dry matter
<i>Ceratophyllum demersum</i> L.	3.3
<i>Elodea canadensis</i> Richard	5.2
<i>Potamogeton lucens</i> L.	3.5
<i>Chara tomentosa</i> L.	9.2
<i>Ch. fragilis</i> Desw.	13.7
Elodea carbonates	43.7
Pondweed carbonates	48.4

Table 49

Coefficients of discrimination of ⁹⁰Sr with respect to calcium when these elements passed into carbonate sediment formed on the surface of different plants under laboratory conditions

Calcium concentration in water (mg/liter)	Cladophora	Elodea	Milfoil	Hornwort
25	0.37	0.32	—	—
57	—	0.32	—	—
162	0.29	0.33	0.37	0.38
380	0.31	0.31	0.36	0.32

are several interpretations of this phenomena (Pia, 1933; Voronikhin, 1953; Berezina, 1963; Kuznetsov, 1970). The most generally recognized hypothesis postulates that the formation of carbonates may be due to the absorption of CO₂ by photosynthesizing plants from bicarbonates. Our data confirm that this process proceeds on the plants' surface, inasmuch as the C_D of ⁹⁰Sr relative to calcium does not depend on the biological peculiarities of plants when the sediment is formed, and is easily reproduced in a simple chemical experiment. The phenomenon of discrimination of ⁹⁰Sr relative to calcium during their transition to carbonates can be associated with the differing solubility in water of the calcium and strontium bicarbonates.

Accumulation of Radionuclides by Dead Tissues of Freshwater Plants

As demonstrated in the foregoing sections, many investigations during the last few years have been concerned with the problem of accumulation of radionuclides by freshwater plants and the role of the latter in the radionuclide migration in a waterbody. At the same time very few data are currently available on the fate of radionuclides after the death of plants and during the formation of detritus. In this respect we can

mention several works dealing with marine hydrobionts (Rice, 1956; Williams, 1960; Polikarpov, 1961, 1964). Yet the specificity of the marine environment does not permit these data to be used directly for the interpretation of radioecological processes occurring in a freshwater reservoir.

We have undertaken a special series of experiments to study the accumulation of major fission fragment radionuclides (^{90}Sr , ^{106}Ru , ^{137}Cs , ^{144}Ce) by the living and dead tissues of freshwater plants. The objects of our investigation were chiefly the moss (*Drepanocladus sendtneri*) and the hornwort (*Ceratophyllum demersum*).

The procedure for these experiments consisted of the following: in some glass vessels holding 4 liters of lake water there were placed living plants while in other vessels we placed plants that had been killed by heating in a thermostat at 105°C for 10 min. Radionuclides were introduced in the form of chlorides at a concentration of about $10\text{ }\mu\text{Ci/liter}$ water. Two, 4, 8, and 16 days after the start of the experiment the content of radionuclides in the plant biomass and in the water was determined. On day 16 all living plants and decaying residues of dead plants that had accumulated radionuclides were transferred into aquariums with the pure lake water. Following this on days 2, 4, 8, and 16 samples were again taken to perform the radiometrical analysis. At the end of 16 days the aquariums were fully cleared of radioactive plants and detritus, and living plants were introduced into the aqueous solution. On the basis of the radioactivity of the plants and water, the behavior pattern of radionuclides released into the solution by living plants and dead residues was appraised. A detailed account of the techniques used in these experiments was given in earlier publications (Kulikov *et al.*, 1967, 1968a, b; Lyubimova *et al.*, 1968).

The difference in the ^{137}Cs accumulation coefficients between the living and dead plants was not statistically significant for the hornwort, whereas for the moss the corresponding value for the living plants was somewhat higher than for the dead (Fig. 36). The data on ^{106}Ru and ^{137}Cs clearly demonstrate that these nuclides are accumulated by the dead organic residues to a larger extent than by the living plants. It was noted that the time course of the accumulation of different radionuclides varies. In the water/plant (living and dead) system, strontium equilibrium became established during the first 2 days, whereas the coefficients of accumulation of ruthenium, cesium, and cerium, both in living plants and in the organic residues of dead plants, increased throughout as many as 16 days.

The radionuclides studied can be arranged in the following sequences in accordance with their capacity for accumulation in plants: cerium $>$ cesium $>$ ruthenium \geq strontium for living plants, and cerium $>$ ruthenium $>$ strontium \geq cesium for dead plants.

The bonding strengths of various radionuclides in plants can be estimated from the results of the experiments on the release of radionuclides from living and dead plants on day 16 after their transfer from the respective radioactive solutions to the pure lake water (Table 50). The release of all radionuclides by living hornwort plants somewhat

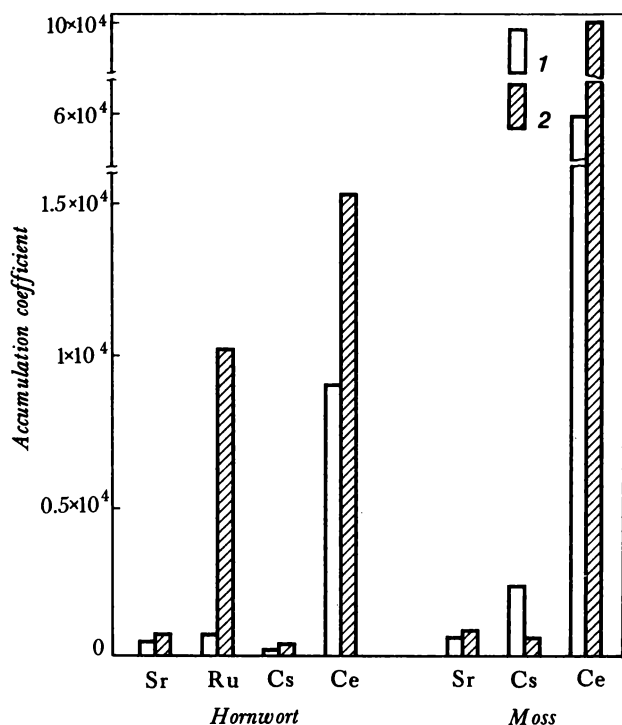


Fig. 36. Coefficients of radionuclide accumulation for living (1) and dead (2) tissues of hornwort and moss.

exceeds that of moss. Strontium is excreted from the dead remains of plants of these two species in about the same amount as from the living plants, while the amount of cesium released from dead plants (particularly from moss) is relatively higher. Ruthenium and cerium pass into solution in appreciably lesser amounts from the dead moss residue than from the living plants, and none of these nuclides passes into solution from dead hornwort. According to their capacity to pass from plants into solution, the radionuclides under study can be arranged in

Table 50

Radionuclide release by living and dead tissues of hornwort and moss (percent of initial plant radioactivity)

Plant	⁹⁰ Sr	¹³⁷ Cs	¹⁰⁶ Ru	¹⁴⁴ Ce
Hornwort:				
Living plants	64.3	37.2	51.2	40.5
Dead plants	54.7	45.2	1.0	1.0
Moss:				
Living plants	45.9	18.4	21.7	37.2
Dead plants	61.6	75.4	17.4	12.6

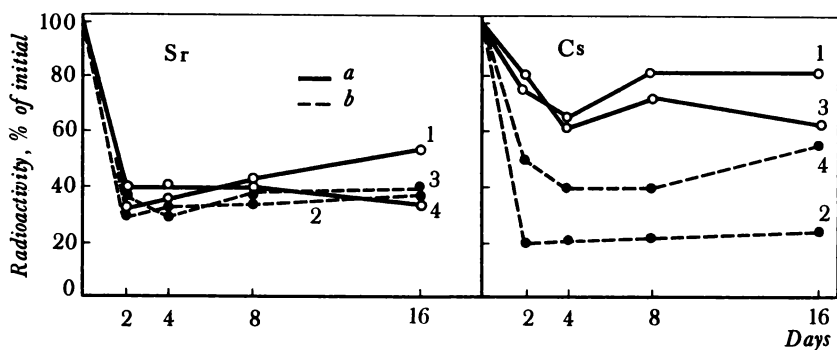


Fig. 37. Reduction of ^{90}Sr and ^{137}Cs content in living (a) and dead (b) plants after single replacement of solutions: (1), (2) moss; (3), (4) hornwort.

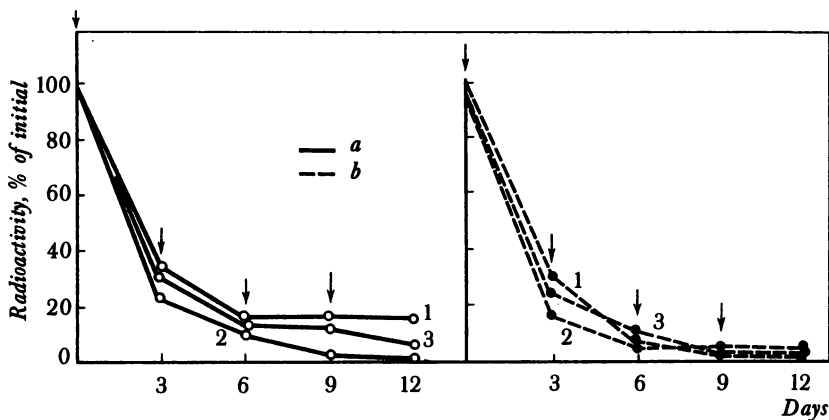


Fig. 38. Reduction of ^{90}Sr content in living (a) and dead (b) plants of cladophora (1), moss (2), and hornwort (3) after four replacements of solutions. Arrows indicate days of solution replacement by pure lake water.

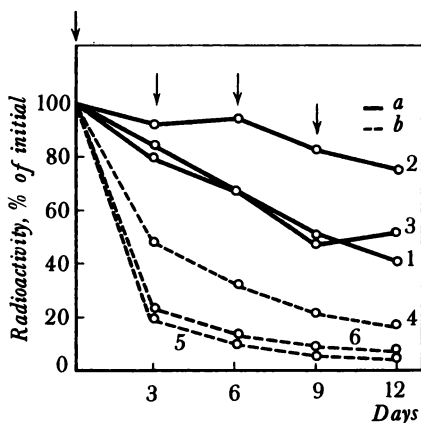


Fig. 39. Reduction of ^{137}Cs content in living (a) and dead (b) plants of cladophora (1), (4), moss (2), (5), and hornwort (3), (6) after four replacements of solutions.

the following sequences: strontium > cerium \geq ruthenium > cesium for living plants and cesium \geq strontium > ruthenium \geq cerium for dead plants.

Considerable output of radionuclides from plants occurs during the first 2 days; thereupon equilibrium becomes established, and the radioactivity remains essentially unchanged (Fig. 37). The output of strontium by the living and dead tissues of the two plant species is practically similar, whereas the output of cesium from dead plants is higher than that from living ones. These differences in the output of strontium and cesium were especially distinct in the experiments in which the solution medium was repeatedly replaced by the pure lake water (Figs. 38 and 39).

By the day 12 (after four replacements of the solutions), ^{90}Sr was almost completely (98 to 99%) given off by the decaying mass of plants of all species, whereas in the living plants the remaining amount of ^{90}Sr was about 1% in moss, 7% in hornwort, and 20% in *Cladophora*. ^{137}Cs , as well as ^{90}Sr was more profusely excreted by the dead plant mass; however, at the end of the experiment, the remaining amount of cesium somewhat exceeded that of strontium. The living plant tissues retain cesium considerably more firmly. In the course of 12 days during which the living plants were desorbed four times, hornwort gave off only 58%, *Cladophora* 48%, and moss only 25% of cesium.

It can thus be put on record that in the three species of freshwater plants we studied, strontium is present in both living and dead tissues chiefly in the mobile, exchangeable form. ^{137}Cs is relatively mobile in the dead plant mass, but is retained by living plants much more firmly than strontium.

As indicated above, the dead plant mass was obtained experimentally by heating of the living plants at 105°C for 10 minutes. It might be assumed that plants dying under natural conditions accumulate and retain radionuclides in a manner different from that in killed plants. However, as some auxiliary experiments have demonstrated, no essential difference could be found in this respect (Tables 51 and 52). The coefficients of strontium and cesium accumulation by moss that died under natural conditions were roughly the same as in killed plants. The rate and pattern of radionuclide output by plants that have died in a natural way were closely similar to those of killed plants. Consequently the methods used in our experiments do not affect the sorptive properties of the naturally dead vegetative substrate.

Of interest were the data on plant accumulation of radionuclides released into water by living and dead plant tissues. The accumulation coefficients for ^{90}Sr in all variants of the experiment exceeded those for ^{137}Cs (Table 53). However, the coefficient values for each individual nuclide in the experimental variants that involved introduction of the nuclides in water, in the form of their chlorides, and in the variants in which the aqueous solution was contaminated by radioactive living and dead plants, were fairly similar. This similarity of behavior of the strontium and cesium radionuclides under the experimental conditions used suggests that living and decaying plants release these nuclides in

Table 51

Coefficients of radionuclide accumulation for plants that died under natural conditions and for killed moss (at day 16 after onset of accumulation)

Vegetative substrate	^{90}Sr	^{137}Cs
Dead plants	579 ± 69	132 ± 44
Killed plants	524 ± 81	147 ± 36

Table 52

Radionuclide release by plants that died under natural conditions and by killed moss after six replacements of solution medium by pure lake water (percent of initial amount)

Vegetative substrate	Days					
	3	6	9	12	15	16
^{90}Sr						
Dead plants	60 ± 4.8	84 ± 4.2	95 ± 1.0	98 ± 0.6	99 ± 0.1	98 ± 0.1
Killed plants	76 ± 3.6	91 ± 0.2	97 ± 0.1	98 ± 0.1	99 ± 0.1	99 ± 0.1
^{137}Cs						
Dead plants	67 ± 1.0	78 ± 4.6	86 ± 3.6	—	91 ± 4.2	93 ± 2.6
Killed plants	78 ± 4.7	89 ± 3.2	94 ± 2.4	—	95 ± 0.9	96 ± 1.0

Table 53

Coefficients of radionuclide accumulation by living moss and hornwort plants (nuclides were introduced in form of chlorides and in form of output by living plants and dead remains)

Nuclide form	^{90}Sr after:			^{137}Cs after:		
	2 days	4 days	8 days	2 days	4 days	8 days
Hornwort						
Chlorides	479 ± 43	556 ± 59	527 ± 28	116 ± 16	117 ± 7	254 ± 14
Output by living plants	583 ± 38	758 ± 18	806 ± 27	107 ± 3	187 ± 24	246 ± 8
Output by dead remains	796 ± 35	765 ± 18	885 ± 23	124 ± 13	203 ± 10	238 ± 9
Moss						
Chlorides	593 ± 58	588 ± 12	605 ± 35	284 ± 16	556 ± 14	1356 ± 360
Output by living plants	645 ± 51	868 ± 52	852 ± 30	258 ± 12	447 ± 37	646 ± 30
Output by dead remains	679 ± 28	711 ± 21	743 ± 13	286 ± 11	707 ± 12	718 ± 64

a mobile form that is easily included in the repetitive cycles of biological substances turnover of a freshwater reservoir.

On the whole, the material presented in this section permits the conclusion that, along with living plants, dying aquatic plants play an important role in the fate of radionuclides in a waterbody. In dead plants the coefficients of ^{90}Sr and ^{137}Cs accumulation proved close to those in living ones, while the coefficients of ^{106}Ru and ^{144}Ce accumulation were severalfold higher than those in living plants. The strength of retention of ^{106}Ru and ^{144}Ce by the organic matter of dead plants was also greater than in living plants, and it was only ^{137}Cs that was more firmly retained by living tissues than by dead organic residues.

One may assume that owing to a stronger retention by the dying plant mass, ^{106}Ru and ^{144}Ce will be transferred with the detritus into bottom deposits, whereas ^{90}Sr and ^{137}Cs , in contrast, are likely to pass back into solution to be included once more in the biological turnover of substances in a waterbody.

Distribution of Radionuclides among the Main Constituents of a Model Waterbody

As noted above, radionuclides, on entering waterbodies, are absorbed not only by living organisms, but also by the dying portions of hydrobionts and by the ground. As a result the major portion of radioactive substances (especially in lake-type waterbodies) becomes, after some time, accumulated in the bottom deposits and in hydrobionts, and their content in water sharply decreases. This phenomenon provided a basis for a number of studies of the role of hydrobionts in deactivation processes in radioactively contaminated waters (Agafonov, 1957; Agre and Korogodin, 1960; Timofeyeva-Resovskaya and Timofeyev-Resovsky, 1960; Agre, 1962; Agre *et al.*, 1962, 1964, 1966; Timofeyeva-Resovskaya, 1963; Agafonov and Ivanov, 1965; Ivanov, 1965a, b; Wlodek, 1967; Iskra *et al.*, 1969b, 1970a, 1971a, b; Meshalkina, 1971).

In the papers listed above, extensive experimental material is presented concerning the distribution of different radionuclides among individual constituents of waterbody in both running water and closed model systems. The works of Timofeyeva-Resovskaya (1963) on comparative studies of the distribution of 18 radioisotopes of different chemical elements between lake water, ground, and freshwater hydrobionts deserve special mentioning. The studies were conducted, using a single experimental procedure, in an aquarium waterbody model. According to this procedure, to a glass aquarium filled with lake water and having a 5 cm thick bottom layer of lake sand and a certain number of aquatic plants, a radionuclide was added to produce a water concentration of about 10^{-5} Ci/liter. Two to 3 months after a near-equilibrium state between the water, sand, and the biomass was attained, the water was drained off, and the plants and sand were removed and weighed. Concentrations of the radionuclide in the water,

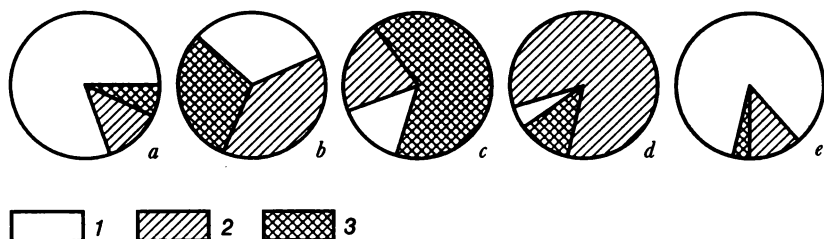


Fig. 40. Patterns of radionuclide distribution: (a) hydrotropes; (b) equitropes; (c) biotropes; (d) pedotropes; (e) relative mass distribution of three components in aquariums. Sectors correspond to radioactivity (in percentage) in water (1), ground (2), and bio-mass (3) (Timofeyeva-Resovskaya, 1963).

sand and in an absolutely dry plant mass were determined radiometrically. The results of these studies have demonstrated that under comparable experimental method and design conditions the distribution of different radionuclides between the waterbody components proceeds in a different fashion. Timofeyeva-Resovskaya subdivided the radionuclides under study into three major groups according to specific type of distribution (Fig. 40).

The "hydrotropic" group includes ^{35}S , ^{51}Cr , and ^{71}Ge ; these are the nuclides which in the course of experimentation remained predominantly in water. The "equitropic" group includes ^{60}Co , ^{86}Rb , ^{90}Sr , ^{106}Ru , and ^{131}I ; these nuclides are more or less uniformly distributed between individual constituents of a waterbody. The "pedotropic" group embraces radionuclides that are accumulated primarily in the ground: ^{59}Fe , ^{65}Zn , ^{91}Y , ^{95}Zr , ^{95}Nb , and ^{137}Cs . To the "biotropic" group are assigned nuclides accumulated predominantly in hydrobionts: ^{32}P , ^{115}Cd , ^{144}Ce , and ^{204}Hg .

A similar scheme was adopted by us for a series of experiments with the group of naturally occurring radioactive elements: ^{226}Ra , ^{232}Th , and ^{238}U . It was found that, in accordance with the type of distribution between different constituents of a waterbody, radium belongs to the equitropic, thorium to the pedotropic, and uranium to the biotropic groups (Iskra *et al.*, 1969b, 1970a).

It should be noted, however, that the classification described above is rather arbitrary since it reflects the distribution of the radionuclides under study in only one particular waterbody model. It is very likely that with a change in the biological, physicochemical, hydrodynamic, and some other parameters of such a model tropism of individual radionuclides in a waterbody will also undergo changes.

Chapter 5

EFFECT OF IONIZING RADIATION ON ORGANISMS AND THEIR COMMUNITIES

On entering the Earth's surface radionuclides contaminate soils and natural waters; this results in an enhancement of the background radiation in the environment and the appearance of additional sources of external and internal irradiation. In the case of external irradiation (resulting from radioactive contamination of the external environment of the organism), the radiation injury is inflicted by gamma and hard beta rays, whereas in the case of internal irradiation (arising through incorporated emitters), by alpha particles and beta rays.

The biological effect of external irradiation is primarily determined by the levels of radioactive contamination of the environment, by the pattern of distribution of the emitting nuclides between the constituents of a biogeocenosis, and by the radiosensitivity of organisms. The biological effect of internal irradiation depends on the accumulation of radionuclides in an organism, their distribution in this organism, and their rate of elimination.

The migration, accumulation, and distribution of radionuclides in the biogeocenotic links of interest to us (soil/solution, soil/plant, and water/hydrobiont) were considered in detail in the foregoing section. The present chapter deals with the general characteristics of radiosensitivity of terrestrial plants and hydrobionts; data on the effect of ionizing radiation on plant communities are also presented.

Radiosensitivity of Terrestrial Plants

The effect of ionizing radiation on plants is a subject that currently concerns a large area of radiobiology, and is covered by a copious literature, including over ten thousand papers. It is not our intention to review all of these works and we shall refer only to several monographs and reviews available to us (Breslavets, 1946; Sparrow *et al.*, 1958; Vasilyev, 1962; Berezina, 1964; Batygin and Savin, 1966; Preobrazhenskaya, 1971).

Current concepts of the general effects of ionizing radiation on plants may be reduced to the following: Extremely low doses, which differ but little from the natural background radiation due to cosmic radiation and the natural radioactivity of the Earth, do not produce any noticeable and detectable effects. Higher, but still relatively weak doses, generally of technogenic origin, produce some stimulation of plant growth,

development, and the total biomass buildup. With a further increase in radiation dose stimulation is replaced by an ever increasing inhibition of plant development and growth with an enhancement of plant mortality. Finally, sufficiently high doses elicit a lethal effect. Acute doses, particularly at earlier developmental stages of the plant, are generally more effective than chronic doses. Apart from the overall biological effect (stimulation, inhibition, and lethality to plants), ionizing radiation at the earlier developmental stages are teratogenic, while at the reproductive stage there are observed disorders in gametogenesis, resulting in a higher sterility; in maturing gametes the percentage of mutations increases. The radiosensitivity of various plant species may differ by one to two orders of magnitude of the stimulating, inhibiting, and lethal radiation doses, the general trend in sensitivity being progression from the lower plant forms to the higher ones.

Radiosensitivity of Plants to External Irradiation

The most important studies of the comparative radiosensitivity of higher terrestrial plants having externally irradiated seeds were carried out by Preobrazhenskaya (Preobrazhenskaya, 1959, 1961, 1967, 1971; Preobrazhenskaya and Timofeyev-Resovsky, 1962). Proceeding from the copious experimental material, covering about 700 species and varieties of plants, Preobrazhenskaya subdivides the plants studied into three major groups. Lethal doses, determined by plant survival rate at the end of the vegetative period, served as radiosensitivity indices. They were 15 to 25 kilorad for the highly radiosensitive, 25 to 100 kilorad for the moderately radiosensitive, and higher than 100 kilorad for the radioresistant group of plants. Among the angiosperms the monocotyledonous class is on the average more radiosensitive than the dicotyledonous class whereas the ligneous and shrubby forms within the dicotyledonous class are more radiosensitive than the herb plants (Tables 54 and 55). It is assumed that the plant species with a long evolution history are more radiosensitive than those with a short one.

Among the trees the least resistant to irradiation at all developmental stages are conifers. The critical doses of the external radiation for seeds of various species of pine, cedar, spruce, fir, and larch causing a 50% reduction in the germination rate and killing an appreciable share of the emergent sprouts, comprise the range from 1 to 6 kilorad (Gustafsson and Simak, 1958; Privalov, 1963; Gailis, 1965; Karaban, 1966, 1970; Clark *et al.*, 1967, 1968; Yushkov and Tarchevskaya, 1968; Tarchevskaya, 1970; Tarchevskaya and Yushkov, 1970).

The experiments run in our laboratory by Tarchevskaya and Yushkov have demonstrated that the LD_{50} , as determined by the seed germination rate in the laboratory, is 2.5 kilorad for pine (*Pinus silvestris* L.), 3 to 3.5 kilorad for spruce (*Picea excelsa* Link), and 5 to 7 kilorad for larch (*Larix sukachevii* Djl.). The energy of seed germination was

Table 54

Radiosensitivity of plants (Preobrazhenskaya, 1967)

Order, family, genus, species	Total No.		Radiosensitive		Moderately resis- tant		Radioresistant	
	Abs.	%	Abs.	%	Abs.	%	Abs.	%
<i>Gymnospermae. Coniferophyta</i>								
Order	1	100	1	100	0	—	0	—
Family	3	100	3	100	0	—	0	—
Genus	6	100	6	100	0	—	0	—
Species	11	100	11	100	0	—	0	—
<i>Angiospermae Monocotyledonae</i>								
Order	5	100	2	40	2	40	1	20
Family	5	100	2	40	2	40	1	20
Genus	43	100	18	42	24	56	1	2
Species	84	100	29	35	54	64	1	1
<i>Dicotyledonae</i>								
Order	27	100	8	30	11	40	8	30
Family	44	100	12	27	15	33	17	39
Genus	198	100	43	22	91	46	64	32
Species	324	100	52	16	152	47	120	37

Table 55

Radiosensitivity of ligneous, shrubby, and herbaceous forms from dicotyledon class (Preobrazhenskaya, 1967)

Plant group	Total No. of species studied	Radiosensitive		Moderately sensitive		Radioresistant	
		Abs.	%	Abs.	%	Abs.	%
Ligneous and shrubby	35	20	57	12	34	3	9
Herbaceous	159	18	11	78	49	63	40

found to be depleted in these three species of plants at still lower radiation doses (Table 56).

Table 57 shows the seed germination rate in the ground and the survival rate of pine seedlings as a function of the radiation dose. One can see that in the experimental variants involving irradiation at dose ranges from 100 to 1000 rad the sprouting began simultaneously with controls and lasted for 9 days. At a radiation dose of 2500 rad the

sprouting process was more protracted (up to 25 days). In seeds irradiated with 2500 rad the germination rate was 16% by the time the sprouting was completed in the control, and 32% by the end of the summer. Unfurling of the cotyledonary conifer needles and emergence of the primary needles in plants of this experimental group lagged 8 days behind the control group and, besides this, death of plantlets was recorded owing to root collar injury caused by fungi. Sixteen months after emergence of sprouts the development of plantlets lagged obviously when seeds were irradiated at doses of 1000 and 2500 rad (Table 58). The length of tip shoots, the number of lateral shoots and paired needles, and the weight of lateral shoots and primary needles in the plantlets in the experimental groups described were lower as compared to controls. When the radiation dose was 250 rad a radiostimulation effect was noted for a number of indices.

Damaging radiation doses for plantlets and adult coniferous trees vary within the range from 600 to 1500 rad (Woodwell and Miller, 1963; Gailis, 1965; Sparrow *et al.*, 1965; Capella and Conger, 1967; Clark *et al.*, 1968).

Investigations conducted in our laboratory showed that the chronic irradiation of Scotch pine plantlets in a dose of 50 rad/day arrested the growth of roots, stem, and primary needles within the first month following the initiation of irradiation. At the same time CO₂ absorption rate was lowered, the outflow of photosynthesis products from the needles was delayed, and the pattern of the assimilate distribution in the plant was changed. Irradiation of plantlets at single doses of 0.5 and 5.0 rad/day during 140 days following the emergence of seedlings did not elicit any noticeable deviations from controls (Yushkov and Kulikov, 1966, 1967, 1969, 1970; Yushkov, 1970; Yushkov *et al.*, 1970).

Some investigators attribute the high radiosensitivity of conifers to the presence of large-sized chromosomes in their cells since the radiosensitivity of many plant species is closely correlated with the size of the cell nucleus and the size of chromosomes in the interphase (Sparrow *et al.*, 1961, 1963; Sparrow and Woodwell, 1962; Woodwell and Sparrow, 1963b). One should note, however, that the sources of great differences in the radiosensitivity of various plant species have not been adequately studied. Within single taxonomic groups of plants there has been observed a dependence of radiosensitivity on seed oiliness (Gustafsson, 1946), on size of the seed and its anatomical structure (Timofeyev-Resovsky and Poryadkova, 1956; Barton, 1961), on ploidy and number of chromosomes (Sparrow and Woodwell, 1962), on degree of hybridity, and other biological peculiarities. At the same time none of these peculiarities by itself can account for the differing radiosensitivity of all of the plant species under study.

The radiosensitivity of plants within a species, and even within a variety, depends to a great extent on many attending factors, which makes the study of the problem still more difficult. It has been shown, for instance, that immature seeds are more radiosensitive than completely mature ones (Engel, 1952; Mericle and Mericle, 1957). The

Table 56

Germination energy and germination rate of pine, spruce, and larch seeds

Plant	Index	Percentage of			
		0	50	100	250
Pine	Germination energy	41 ± 0.6	46 ± 3	48 ± 4	44 ± 3
	Germination rate	87 ± 4	72 ± 5	91 ± 3	92 ± 2
Spruce	Germination energy	39 ± 3	37 ± 1	32 ± 2	29 ± 2
	Germination rate	75 ± 3	94 ± 2	71 ± 2	76 ± 2
Larch	Germination energy	12 ± 1	24 ± 1	12 ± 1	11 ± 1
	Germination rate	84 ± 3	89 ± 3	91 ± 2	83 ± 3

Table 57

Influence of preplanting irradiation of Scotch pine seeds with ^{60}Co

Dose (rad)	Number of shoots			
	June 21	June 23	June 26	June 30
Control	10.3 ± 1.3	30.3 ± 13.8	76.0 ± 3.0	78.0 ± 3.5
100	3.6 ± 1.7	22.3 ± 9.8	80.6 ± 3.3	86.6 ± 2.3
250	4.3 ± 2.3	19.0 ± 9.5	76.3 ± 2.9	80.6 ± 1.8
500	3.6 ± 2.7	10.6 ± 3.8	66.0 ± 4.4	71.0 ± 5.1
1000	1.6 ± 0.9	8.3 ± 3.7	55.0 ± 7.1	68.3 ± 3.8
2500	0	2.0 ± 1.5	16.6 ± 4.8	26.3 ± 6.1
5000	0	0	0	1.0

Table 58

Influence of preplanting irradiation of seeds with ^{60}Co gamma rays on

Dose (rad)	No. of plants measured	Length (mm)		No.	
		Hypocotyl	Tip shoot	Lateral shoots	Lateral buds
Control	70	28.1 ± 0.5	72.7 ± 1.2	1.8 ± 0.1	0
100	80	27.2 ± 0.6	68.7 ± 2	1.7 ± 0.1	0
250	70	28.7 ± 0.7	75.3 ± 2.5	2.5 ± 0.1	0.21 ± 0.06
500	80	27.0 ± 0.6	68.9 ± 1.6	1.9 ± 0.2	0.3 ± 0.08
1000	87	29.6 ± 0.6	57.9 ± 3.4	1.2 ± 0.1	1.1 ± 0.2
2500	30	27.1 ± 0.9	48.9 ± 3.1	0.8 ± 0.2	2.1 ± 0.8

as dependent on ^{60}Co gamma-radiation dose

germinating seeds at various radiation doses (rad)

500	750	1 000	1 500	2 500	4 000	5 000	7 500
37 ± 3	30 ± 2	26 ± 2	—	5 ± 1	—	0.3	0
90 ± 2	71 ± 3	86 ± 2	—	50 ± 6	—	1.3	1.6
34 ± 2	30 ± 2	23 ± 3	15 ± 1	0	0	0	0
81 ± 3	78 ± 2	89 ± 3	80 ± 2	76 ± 1	28 ± 2	0	0
8 ± 1	15 ± 1	5 ± 0.5	8 ± 1	5 ± 0.4	0	0	0
84 ± 3	88 ± 2	86 ± 2	91 ± 3	95 ± 3	74 ± 3	53 ± 2	48 ± 2

gamma rays on dynamics of sprouting and survival rate

(% of seeds planted)

July 15	July 21	August 15	October 17
77.6 ± 2.5	78.0 ± 2.5	78.0 ± 2.5	78.0 ± 2.3
86.0 ± 4.3	86.6 ± 4.3	86.0 ± 5.3	84.0 ± 5.2
83.0 ± 2.8	84.3 ± 3.5	84.0 ± 5.4	83.0 ± 4.2
75.6 ± 4.8	78.3 ± 3.6	80.0 ± 4.0	77.0 ± 5.5
72.3 ± 4.1	73.6 ± 3.8	75.0 ± 4.1	73.0 ± 3.3
38.6 ± 1.7	40.0 ± 5.7	32.3 ± 5.2	31.0 ± 6.5
0.6	0.7	0	0

growth and development of Scotch pine seedlings (aged 16 months)

per plant			Dry weight of 10 plants (gm)			
Tip shoots	Primary needles	Paired needles	Hypocotyl + tip shoot	Primary needles	Paired needles	Lateral shoots
2.4 ± 0.1	54 ± 1	30 ± 1	1.5 ± 0.1	0.73 ± 0.01	4.4 ± 0.3	0.43 ± 0.02
2.5 ± 0.1	47 ± 1	26 ± 1	1.5 ± 0.1	0.6 ± 0.03	4.3 ± 0.2	0.53 ± 0.07
2.9 ± 0.1	56 ± 2	33 ± 1	1.8 ± 0.1	0.78 ± 0.03	4.8 ± 0.3	0.95 ± 0.1
2.5 ± 0.1	53 ± 1	28 ± 1	1.5 ± 0.04	0.72 ± 0.06	4.2 ± 0.2	0.45 ± 0.05
2.3 ± 0.1	47 ± 1	23 ± 1	1.3 ± 0.03	0.51 ± 0.08	4.1 ± 0.3	0.37 ± 0.2
2.6 ± 0.1	47 ± 2	22 ± 1	1.2 ± 0.1	0.45 ± 0.04	4.3 ± 0.5	0.19 ± 0.09

radiosensitivity was found to depend on mature seed moistness (Caldecott, 1954; Savin, 1962; Poryadkova, 1963, 1965; Fesenko and Poryadkova, 1966; Nemtseva, 1967), on conditions of the seed stock growing, and its postirradiation storage (Yanushkevich, 1963; Ehrenberg, 1954; Wolff and Sicard, 1961; Preobrazhenskaya, 1970), and on conditions of the postirradiation growth of plants (Vasilyev, 1962; Moshkov and Savin, 1963; Batygin and Savin, 1966). The radiosensitivity also changes in the process of swelling and germination of seeds: the swollen and germinating seeds are severalfold more sensitive to radiation than are dry seeds (Atabekova, 1936; Afanasyeva, 1939; Timofeyev-Resovsky *et al.*, 1957a, b; Poryadkova *et al.*, 1960; Alshits, 1969). In the irradiation of vegetating plants generative developmental stages have been found to be more radiosensitive than vegetative stages (Sparrow and Woodwell, 1962).

In addition, the radiosensitivity of a plant depends on the type of radiation and the dose rate. Radiations with high linear energy transfer (alpha particles, neutrons) are as a rule more effective than beta and gamma rays, particularly in the case of the accumulation of the respective emitters in the plant tissues (Timofeyev-Resovsky, 1956; Timofeyev-Resovsky and Luchnik, 1960; Poryadkova *et al.*, 1960; Valeva, 1969). The dose rate is of great importance when plants are irradiated during the periods of their active growth. It has been found that accumulated doses inducing drastic inhibition of plant growth are much higher at low dose rates than at high dose rates (Gunkel *et al.*, 1953; Sparrow and Woodwell, 1962). Radiation injury of dry seeds appears to depend on the dose rate to a lesser extent, although recently evidence has been obtained that long-term exposure of dry seeds to a low dose rate is more effective than an acute exposure to a high dose rate (Kuzin *et al.*, 1960; Berezina, 1964; Tsarapkin and Tsarapkina, 1969a, b).

Radiosensitivity of Plants under Conditions of Radioactive Contamination of Soil

Problems of plant sensitivity to chronic irradiation arising from the radioactive contamination of soil as well as from radionuclide accumulation in plant tissues have so far been studied to a lesser extent than the radiosensitivity of plants to irradiation from external sources. Specific features of the biological effect of radionuclides contaminating the soil are associated with the nonuniform distribution of the dosage burden between different parts and organs of plants throughout their lifespan; such distribution is defined by the processes of migration, accumulation, and redistribution of the radionuclides in both the soil and the plant itself.

Certain species of cultivated plants (wheat, oats, pea) exhibit signs of radiation damage at sufficiently high levels of radioactive contamination of the soil (0.5 to 1.0 mCi/kg). It was noted that the earlier

developmental stages of plants are more radiosensitive than the later stages (Yudintseva and Gulyakin, 1968).

Of interest were studies concerned with conifers (Karaban and Tikhomirov, 1967, 1968; Karaban, 1970; Tikhomirov, 1972). These authors have demonstrated that partial inhibition of pine, spruce, and larch seed germination in the ground occurs at levels of soil contamination with ^{90}Sr which bring the total dose of radiation of seeds during their germination to as high as 1000 rad, whereas a complete loss of germination occurs at a dose level of approximately 2500 rad. The former case is equivalent to a radioactive contamination density of the soil of 10 mCi/m^2 , whereas the latter case is equivalent to a contamination density of 40 mCi/m^2 .

A pronounced inhibition of growth and a higher mortality of plantlets were recorded during the second year following planting, at a soil contamination level of about 2 mCi/m^2 when the total radiation dose received by the terminal bud reached 600 to 800 rad (at a dose rate of 4 rad/day). An irreversible damage to plantlets occurred when soil contamination with ^{90}Sr reached 6 to 10 mCi/m^2 and the total radiation dose delivered to the terminal bud exceeded 3000 rad (at a dose rate of 10 to 15 rad/day). The crucial period in the life of plantlets growing in radioactively contaminated sites are the initial developmental stages, throughout which the plantlets are confined to the upper, most heavily contaminated, soil layer and the contiguous air layer. As the roots and tip shoots of plantlets recede from the maximally radioactive soil layer the radiation injury hazard diminishes.

Similar experiments with Scotch pine plantlets were conducted in our laboratory by Yushkov and Tarchevskaya. In these experiments the subgerminating pine seeds were planted in the spring in vegetative vessels charged with soddy-meadow soil mixed with washed lake sand in a 1:1 proportion. As a preliminary step, aliquot amounts of ^{90}Sr in the chloride form were added to all of the vessels at 0.6 mCi per vessel. In the first experimental variation the total amount of strontium was added to the 0- to 2-cm topsoil layer; in the second, the strontium-treated layer was 0 to 5 cm thick, and in the third, the layer thickness was 0 to 10 cm. In the fourth variation the entire amount of strontium was uniformly mixed with the soil to fill the entire volume of the vegetative vessel to a height of 20 cm. With this manner of ^{90}Sr addition, its initial concentration in the first experimental variation was 1 mCi/kg , in the second 0.4 mCi/kg , in the third 0.2 mCi/kg , and in the fourth variant it was 0.1 mCi/kg of dry soil.

Thus, an experimental model of radioactively contaminated sites was developed in which the initial amount of ^{90}Sr per unit surface was identical in all cases, while its distribution throughout the soil column was different. Under these conditions pine seeds and plantlets were under differing irradiation conditions from the very beginning of their development.

Observations showed that in all variations of the experiment the emerging shoots were normal, save for the variant with ^{90}Sr introduction in the uppermost soil layer, in which only occasional

Table 59

**Dry mass weight and some morphometric indices of 90-day-old pine
⁹⁰Sr in soil layers (M ± m)**

Depth of radioactive soil layer (cm)	Radioactivity of contaminated soil layer (mCi/kg)	Mean weight of single plant		
		Needles	Stem	Root
Shoots died 2 months after				
0-2	1.0			
0-5	0.4	30 ± 0.8	6 ± 0.2	23 ± 0.8
0-10	0.2	38 ± 1.4	9 ± 0.2	32 ± 1.2
0-20	0.1	30 ± 1.1	8 ± 0.4	22 ± 1.2
Control	0.0	39 ± 0.6	10 ± 0.2	37 ± 0.7

shoots were observed which emerged with a 5 to 7 days' delay and all died about 2 months later.

Ninety days after emergence of the shoots, the most inhibited of them, as compared with controls, were those grown under conditions of ⁹⁰Sr introduction to the 0- to 5-cm soil depth (Table 59). Of the remaining two variants of the experiment plantlets grown under conditions of ⁹⁰Sr distribution over the whole soil thickness (0 to 20 cm) were more inhibited, according to a number of indices, than plantlets grown in vessels with ⁹⁰Sr initially added to the soil depth of 0 to 10 cm. The last result suggests that at certain stages of development the plantlets tolerate radioactive contamination of the soil better when a portion of their root system develops outside the radioactive soil layer.

Table 60

**Accumulation of ⁹⁰Sr in different portions of pine seedlings
as a function of initial distribution of the radionuclide in soil layers**

Depth of radioactive layer (cm)	Radioactivity of contaminated layer (10 ⁻⁹ Ci/gm dry soil)	Plant portions	Plant radioactivity (10 ⁻⁹ Ci/gm dry matter)	Accumulation coefficient
0-5	400	Needles	359.9 ± 35.4	0.9
		Stem	509.1 ± 16.2	1.2
		Root	201.3 ± 59.6	0.5
0-10	200	Needles	227.9 ± 18.2	1.3
		Stem	223.7 ± 19.9	1.2
		Root	249.9 ± 25.7	1.2
0-20	100	Needles	181.4 ± 15.4	1.8
		Stem	185.3 ± 11.0	1.8
		Root	309.4 ± 22.8	3.0

portions (mg)	Mean length of tip shoot (mm)	Mean total length of stem (mm)	Average No. of needles per plant
Whole plant			
emergence			
59 ± 1.1	3 ± 0.5	20 ± 1.4	30 ± 2.3
79 ± 1.9	6 ± 0.3	24 ± 1.2	33 ± 0.4
60 ± 1.7	8 ± 0.5	28 ± 1.2	37 ± 1.3
86 ± 0.9	10 ± 0.5	30 ± 0.8	37 ± 1.3

The ^{90}Sr concentration in the above-ground parts of plantlets is somewhat higher when the nuclide is introduced into the surface soil layer, whereas the accumulation coefficients increase markedly with an increase in the radioactive soil layer around the roots (Table 60). This is due to the fact that with a superficial addition of the emitter a considerable part of plantlet roots remains outside the radioactive soil layer.

The experimental data presented bear out that the radiation damage to plants under conditions of radioactive contamination of the earth surface depends upon the vertical distribution of radionuclides in the soil, which in turn is defined by the capacity of radionuclides to migrate in the soil.

At similar levels of radioactive contamination per unit area, the greater is the part of radioactive substances retained for long in the uppermost soil layers and in the litter, the more likely is a radiation damage to plants. This is particularly true with regard to seed regeneration among the radiosensitive forms of plants, as, for example, members of the genus *Pinus*.

Radiostimulation

As noted above, the irradiation of vegetative objects at relatively low doses of ionizing radiation elicits a radiostimulation effect that is manifested by acceleration of growth and an increase in the biomass, as well as by changes in the quality and structure of the yield of many plant species (Breslavets, 1946; Zhezhe, 1955; Kuzin, 1955; Timofeyev-Resovsky and Luchnik, 1960; Poryadkova *et al.*, 1960; Vlasyuk *et al.*, 1963; Berezina, 1964). Radiostimulation may be evinced by the most varied types of radiation exposure: by presowing maceration of seeds in solutions of radioactive substances in appropriate concentrations; by exposure of seeds to X or gamma rays; by introduction of emitters into the soil; and by chronic irradiation of plants in the course of vegetation.

For the practical application of radiostimulation in agriculture and in selection, the most acceptable method is that of external irradiation of seeds prior to sowing and irradiation of vegetating plants in gamma fields.

We have observed the stimulating effect of radiation on tomato, radish, white cabbage, and collard seeds (Kulikov, 1958a, b, 1961b; Poryadkova *et al.*, 1960).

Within a month after planting (by the time of singling), the number of tested plants per unit area in an experimental variant involving irradiation in doses of 250 to 1000 rad was somewhat higher than that in controls (Table 61). The mean height of the plants irradiated at the time of transplantation into the ground exceeded very significantly that of the controls. The count of flowering plants at the time of efflorescence showed that on the count day the number of flowering plants among the irradiated specimens exceeded 3 to 4 times that in the control. At the stage of mass flowering and infructescence of the first racemation the number of plants among the irradiated specimens was also significantly greater than on the control plots. It thus follows that the plants grown from the irradiated seeds developed more congruously and rapidly than those in the control.

The beneficial effect of the irradiation manifested itself in the results of harvesting. The mean weight and mean number of tomato fruits on a single plot, as well as the mean weight of a single tomato fruit over 3 cm in diameter, and the relative weight of fruits on plants proved to be higher in the irradiated specimens than in the controls. It was only with a radiation dose of 16000 rad that the mean weight of one fruit was somewhat reduced. The number of ripening (yellow) fruits in the irradiated groups was 2 to 2.5 times greater than that in the control plots. Since at the time of harvesting the ripening fruits were scarce, we picked, as far as possible, 100 identical and well-shaped green fruits for each experimental variant, and kept them indoors to allow complete ripening. Subsequent checks and counts of fruits that reached marketable quality permitted to notice that the ripening process of fruits picked from the control plants was more protracted than that of fruits whose seeds had been irradiated prior to planting. On September 26 the number of ripe fruits among the irradiated specimens was much greater than that in controls; subsequently this difference leveled off, although even in later periods the number of ripe fruits in control was lower than that in the irradiated variants.

Irradiation of radish seeds in doses ranging from 250 to 2000 rad resulted in a somewhat higher mean number of plants, weight of root vegetables, and weight of radish leaves than on the control plots. Higher radiation doses inhibited the development of plants. It should be noted that our data on the pre-sowing irradiation of radish seeds are in close agreement with the experimental results of other investigators (Berezina, 1964).

In experiments with white cabbage and collard we met with success in ascertaining the optimal range of gamma-radiation doses beneficial for the development of these plants. Radiation doses in a range from 250

Table 61

Effect of preplanting irradiation of seeds with ^{60}Co gamma rays on development and yield of tomatoes
(Gruntovy Gribovsky variety, mean values for 12 replications)

Dose (rad)	No. of plants at singling	Mean height of transplanted plants		No. of flowering plants, June 28	No. of plants with ovaries on first raceme, July 12	Mean weight of fruits ($d^* > 3$ cm) in one plot		No. of yellow fruits per experi- mental varia- tion	Average No. of all fruits ($d > 3$ cm) in one plot	Mean weight of one fruit with $d > 3$ cm (gm)	Relative weight of fruits and ovaries (%)
		cm	P**			kg	P**				
Control	149	11.5 \pm 0.3		11	51	8.2 \pm 0.7		16	194 \pm 16	42.3	68
250	172	14.6 \pm 0.3	< 0.0001	33	72	9.5 \pm 0.6	0.17	37	206 \pm 17	46.4	71
500	160	14.8 \pm 0.2	"	42	83	10.0 \pm 0.4	0.04	36	225 \pm 14	44.2	70
1 000	1171	14.9 \pm 0.2	"	43	88	10.1 \pm 0.4	0.02	40	233 \pm 11	43.6	73
2 000	148	14.2 \pm 0.2	"	40	80	9.5 \pm 0.4	0.10	47	216 \pm 11	44.2	72
4 000	150	14.5 \pm 0.2	"	34	77	9.8 \pm 0.3	0.05	33	226 \pm 13	43.4	72
8 000	148	14.6 \pm 0.3	"	31	76	9.3 \pm 0.4	0.18	31	216 \pm 13	43.5	72
16 000	148	12.8 \pm 0.2	"	28	64	8.9 \pm 0.4	0.38	22	224 \pm 8	39.9	69

* d—diameter.

** P—statistical significance level.

to 8000 rad stimulated white cabbage, whereas those from 500 to 4000 rad stimulated collard.

It is important to note that a pre-sowing irradiation of seeds in booster doses causes no harmful aftereffects. The yield after planting radiostimulated seeds of pea, wheat, barley, and rice was even somewhat higher than in the control (Breslavets, 1946; Poryadkova *et al.*, 1960).

Apart from radiostimulation, preliminary irradiation in low doses has in a number of cases a protective effect with regard to later irradiation in sublethal doses. This phenomenon was recorded by many investigators experimenting on animals (Luchnik and Kulikova, 1956; Pomerantseva and Ramaya, 1962; Arbuzov and Lomonos, 1962; Bacq and Alexander, 1961; Kulikov *et al.*, 1969).

There are also data on the radioprotective effect of preliminary irradiation in plants (Schoen and Magnus, 1953).

A similar effect was recorded in our laboratory in experiments with seeds of pea ("Capital" variety) and Scotch pine (Tarchevskaya and Kulikov, 1969; Kulikov *et al.*, 1971a). In experiments with pea the dry dormant seeds were preliminarily irradiated with ^{60}Co gamma rays in doses of 50, 100, 250, 1000, and 2500 rad at a dose rate of 25 rad/min. The irradiation of the seeds in a dose of 2500 rad was repeated in the swollen state after their 24 hr maceration in water at room temperature. The total interval between the first and the second irradiation was 500 hr. The choice of the dose for the second irradiation was determined by the fact (as demonstrated by preliminary tests at this dose level) that by the end of the first postirradiation mitosis the number of cells with chromosome aberrations in the anaphase present in the radicle embryonic meristem reached 40 to 50%.

The incidence of cells with chromosome aberrations in the radicle meristem of sprouts, and the weight of the dry mass of the above-ground parts 30 days after the emergence of shoots, were adopted as criteria of the radiobiological response in all variations of the experiments. The plants were grown in vegetative vessels under greenhouse conditions (33 seeds per vessel). Each experiment was performed in triplicate. The chromosome aberrations in the radicle meristem were read from provisional squash preparations stained with acetolacmoid (an organic dye used in cytological experiments).

Whereas the percentage of spontaneous chromosome mutations in the control (no irradiation) run was comparatively low (Table 62), the incidence of cells with chromosome mutations following irradiation of the swollen seeds with 2500 rad (without preliminary irradiation) was appreciably higher (34.5%). Preliminary irradiation in a dose of 100 rad reduced the incidence of damaged cells by a factor of 2 and more ($t = 5.4$; $P = 0.007$). The 250- and 500-rad doses elicited no appreciable effect on the total production of injured cells (the recorded differences are nonsignificant), whereas a preliminary irradiation in a dose of 1000 rad obviously boosted the effect of radiation injury. The maximal reduction in the injured cell number per unit dose is likewise noted with a preliminary irradiation in a dose of 100 rad; this dose also produced

Table 62

Yield of cells with chromosome aberrations in root meristem of embryos and weight of dry superterranean mass of 30-day-old plants as a function of dose of preliminary irradiation of dry seeds subsequently irradiated at seed swelling stage in a dose of 2500 rad

Preliminary radiation dose (rad)	No. of anaphases studied	Injured anaphases (%)	No. of injured cells per 1000 rad	Mean weight of single plant (mg)
0	200	34.5 ± 7.3	27	144 ± 9
50	—	—	—	137 ± 9
100	250	14.0 ± 3.4	10	188 ± 21
250	250	25.6 ± 4.7	18	140 ± 13
500	200	38.5 ± 4.3	25	134 ± 7
1000	200	54.0 ± 8.1	30	119 ± 7
2500	—	—	—	139 ± 7
Non-irradiated control	250	2.8 ± 1.5	—	194 ± 14

an increase in the mean weight of one plant, as compared with the variant without a preliminary irradiation.

Rather similar results were obtained in the experiments with the Scotch pine seeds. The dormant pine seeds (6 to 7% moistness) were preliminarily irradiated with ^{60}Co gamma rays in doses of 50, 100, 250, 1000, and 2000 rad at a dose rate of 25 rad/min. Thereupon the seeds were put on filter paper in Petri dishes (100 seeds per dish) containing moist sand, and were allowed to stand in a thermostat at 22 to 23°C. In 3 days the swollen seeds were subjected to repeated irradiation at 1000 and 2500 rad, and returned to the thermostat for further germination. Seeds that did not undergo preliminary irradiation served as control for each experimental variation. The energy of germination and the germination rate were used as criteria of the radiobiological response. Each experiment was conducted in triplicate.

The preliminary irradiation of dry seeds in doses of 50 and 100 rad not only does not reduce but even somewhat increases the germination energy and rate as compared with the experimental variant omitting preliminary irradiation (Table 63). Higher doses of preliminary irradiation led to increased sensitivity to repeated irradiation.

Examination of the results of experiments with pea and pine seeds indicates that preliminary irradiation of dry seeds with relatively low gamma-radiation doses enhances the resistance of the seeds to subsequent irradiation with sublethal doses. Moreover, the radioprotective effect is shown not only in external signs of plant development (germination and sprouting rate, increase in the weight of plants), but also in a reduction of radiation injury to genetic structures. Since radiation doses eliciting the radioprotective effect are found within the range of stimulating doses for seeds of both pea (Berezina, 1964) and pine (Tarchevskaya, 1970; Karaban, 1970), it may be assumed that the radiobiological effects of radiostimulation and of increased

Table 63

Germination energy and overall germination rate of pine seeds as a function of dose of preliminary irradiation of dry seeds and of their subsequent irradiation at seed swelling stage

Preliminary radiation dose (rad)	1 000 rad			2 500 rad		
	Germination energy (%)	Germination rate (%)	Relative germination rate (% of control)	Germination energy (%)	Germination rate (%)	Relative germination rate (% of control)
0	24.6	51 ± 1	100	16.0	24 ± 2	100
50	45.0	63 ± 5	123	29.3	39 ± 3	164
100	34.0	60 ± 5	118	17.6	30 ± 7	126
250	16.6	47 ± 5	91	10.0	20 ± 7	84
500	18.0	48 ± 6	93	9.3	14 ± 3	57
1 000	7.0	38 ± 1	74	4.6	9 ± 2	36
2 000	2.0	17 ± 3	34	1.6	2 ± 1	8

radiosensitivity, due to preliminary irradiation in low doses, are controlled by a single mechanism. Radiostimulation appears to be manifested not only by acceleration of the growth and development, and by increase of the biomass, but also by increased radioresistance of seeds to subsequent irradiation.

Unfortunately, the mechanisms responsible for the radiostimulation phenomenon are still insufficiently clear. Current ideas regarding this issue have been discussed in detail in a monograph by Berezina (1964). It is to be hoped that a more detailed study of the manifestations of the radioprotective effect of low radiation doses, using the seeds of radiobiologically well-studied plants, will be instrumental in a better understanding of the mechanisms of the stimulatory effect of radiation.

Radiosensitivity of Freshwater Organisms

The general pattern of the biological effect of the ionizing radiation, as dealt with in the preceding section and exemplified by vegetative objects, holds true for all living organisms, including hydrobionts. It should only be noted here that the general radiobiology of hydrobionts, and their comparative radiosensitivity in particular, have been less well studied to date than those of terrestrial plants.

Radiosensitivity of Freshwater Organisms under Conditions of External Irradiation

It is unnecessary to review in detail all of the literature concerned with the sensitivity of hydrobionts to external irradiation, since this has already been fully and circumstantially done by Polikarpov (1964). It follows from Polikarpov's review that the radiosensitivity of aquatic organisms to a single external irradiation dose increases from the lower to the more highly organized forms and decreases from the early

developmental stages of these organisms to the later ones. Thus, for instance, the LD₅₀ for bacteria equals approximately 1 Mrad, whereas for algae and some Protozoa they vary from 4500 to 120,000 rad, for Crustacea from 1000 to 50,000 rad, and for adult fish and certain amphibians from 700 to 3000 rad. The LD₅₀ for the rainbow trout (*Salmo gairdnerii*) at a single cell stage following fertilization is 58 rad, whereas for adult specimens it increases up to 1500 rad (Welander *et al.*, 1948, 1949; Welander, 1954).

Especially drastic changes in fish radiosensitivity are recorded in the course of fertilization and early cleavage of the zygote. In experiments with eggs of the mud loach (*Misgurnus fossilis* L.) and starred sturgeon (*Acipenser stellatus* Pall.) it was found that the first radiosensitivity peak corresponds to the stage of approximation of the male and female pronuclei, while the subsequent peaks precede the interkinetic state of the nucleus between divisions of cleavage (Belayeva and Pokrovskaya, 1959; Neyfakh, 1959a, b). Similar results were obtained by us in experiments with pike eggs (Kulikov, 1970a, b). In these experiments the eggs were divided into several portions (150 to 200 eggs each) immediately after insemination and placed into Koch dishes with lake water. Twenty minutes after insemination the first portion was irradiated with ⁶⁰Co gamma rays in a dose of 200 rad at a dose rate of 26 rad/min. Each subsequent portion of eggs was then irradiated in the same dose 10 minutes after beginning the irradiation of the preceding portion, taking into account the 8 min required to irradiate the eggs and the 2 min required to install by hand the dish containing the new egg portion over the nuclide. In all, the experiment included three dishes containing the control (nonirradiated) eggs and 20 dishes containing the eggs irradiated in a dose of 200 rad at different developmental stages following insemination. The developmental process proceeded at a water temperature of 15°C. In the course of the experiment the water in the dishes was replaced twice or thrice a day and dead embryos were removed.

The survival rate of embryos, the average lifespan to hatching, and the number of embryos with various morphological abnormalities (scoliosis, caudal fin hypoplasia, changes in heart structure and blood vessels) were taken into account as criteria of radiosensitivity.

In all cases irradiation of the eggs markedly decreased the percentage of larvae hatching and increased the number of larvae having morphological abnormalities as compared to the control (Table 64). The average embryonic life until hatching became shorter and the coefficient of variation of the average lifespan increased. The latter circumstance indicated that the kill of embryos developed from the irradiated eggs was more protracted than the same process in the control. Also noteworthy is a sharp change in the egg radiosensitivity in the course of development after insemination (Fig. 41). The first radiosensitivity peak was noted long before cleavage of the zygote, in the interim between 30 and 50 min following insemination; irradiation of the eggs at this stage resulted in the death of all embryos in the course of 3 days. The second peak was observed shortly before the appearance of the first

Table 64

Radiosensitivity of pike eggs during initial developmental stages after insemination

Time elapsed after insemination (min)	Total No. of eggs	Hatching (%)	Abnormal specimens (%)	Average lifespan of embryos prior to hatching (days)	Coefficient of variation for average lifespan (C)
Control	467	83.7	16.0	5.80 ± 0.34	29.3
20	186	25.8	79.1	4.10 ± 0.15	43.9
30	162	9.8	87.5	3.20 ± 0.08	31.6
40	165	0.0	—	3.01 ± 0.10	43.2
50	136	6.6	88.8	2.91 ± 0.10	37.1
60	136	44.1	65.0	4.01 ± 0.24	51.4
70	184	65.8	48.7	4.33 ± 0.30	52.2
80	167	55.6	62.3	5.41 ± 0.20	35.1
90	143	55.9	68.7	3.40 ± 0.25	57.6
100	138	36.8	88.2	3.20 ± 0.20	50.0
110	151	33.7	78.4	3.72 ± 0.20	51.0
120	156	27.5	86.0	3.60 ± 0.14	41.1
130	167	2.4	100.0	2.60 ± 0.06	33.4
140	162	15.4	48.0	3.03 ± 0.10	46.2
150	171	74.2	51.9	4.59 ± 0.29	42.2
160	167	71.2	33.6	3.59 ± 0.30	60.2
170	147	59.8	78.4	3.39 ± 0.20	53.1
180	138	56.5	84.6	3.50 ± 0.20	48.6
190	139	46.0	82.8	5.49 ± 0.20	32.8
200	113	13.2	73.3	3.03 ± 0.10	31.4
210	120	2.5	100.0	2.36 ± 0.10	31.4

cleavage fissure, between 120 to 140 min following insemination, while the third peak was evident at the 4- to 8-blastomere stage.

The described pattern of the radiosensitivity changes in the course of fertilization and early cleavage of pike eggs is in good agreement, both by the time course and by its external manifestations, with the data presented earlier by other investigators who irradiated eggs of mud loach and starred sturgeon (Belayeva and Pokrovskaya, 1959; Neyfakh, 1959a, b). As mentioned above, these works showed that the initial period of the enhanced radiosensitivity corresponded to the stage of male and female pronuclei approximation, whereas the following periods

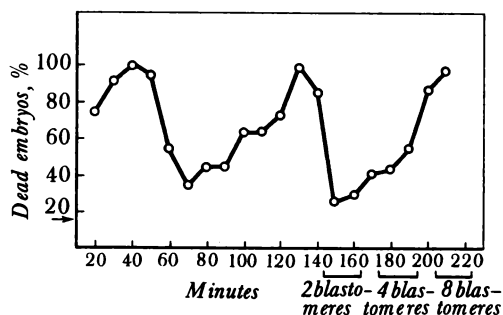


Fig. 41. Change in radiosensitivity of pike eggs in the course of fertilization and early cleavage. Arrow indicates percentage of dead embryos in control.

were associated with different phases of the cycles of initial embryonic mitoses. There is thus reason to believe that in our experiments with pike eggs the periods of higher radiosensitivity correspond to the same developmental stages. Absence of a well-pronounced radiosensitivity peak preceding the 4-blastomere stage in our experiments may probably be explained by the fact that the most sensitive transitional period between the 2-blastomere and the 4-blastomere stages coincided in time with that moment of eggs irradiation when the total dose did not yet attain the necessary level because of the low power of the irradiation source.

We followed the change in radiosensitivity of embryos throughout the entire period of embryogenesis using eggs of tench (*Tinca tinca* L.) and of a freshwater mollusk (*Lymnaea stagnalis* L.).

In the tench experiments (Kulikov, 1969, 1970b), the eggs fertilized under laboratory conditions were irradiated at different developmental stages with ^{60}Co gamma rays in a dose of 250 rad at a dose rate of 24.5 rad/min and incubated concomitantly with the nonirradiated controls in Koch dishes containing pure lake water at a temperature of 20 to 23°C prior to hatching. In the course of incubation of the eggs mortality, time of hatching, and the number of larvae, as well as the number of larvae with morphologic abnormalities were recorded. Twenty-four hours after hatching, part of the larvae (100 specimens from each variant of the experiment), was additionally irradiated in a dose of 1500 rad and the survival rate throughout the next 18 to 20 days was recorded. Another, nonirradiated, portion of larvae served as the control during the same time period.

Irradiation of the eggs prior to the first division of the zygote (15 min after insemination) resulted in the appearance of various signs of injury in all embryos (Table 65). At later developmental stages the embryonic radiosensitivity was reduced; however, at the 4- and 16-blastomere stages, and at the mid-blastula stage it noticeably increased. These radiosensitivity peaks are apparently due to changes in the radiosensitivity in the course of initial embryonic mitoses, as described above. These changes have recently been regarded as being associated with DNA transinduction phenomena, i.e., with those processes that affect the quality and volume of the genetic information necessary for completion of each next developmental stage of the embryo (Kuzin and Yusifov, 1967; Kuzin, 1970).

Mass hatching occurs from 52 to 56 hr following insemination. In the case of eggs irradiated earlier than the beginning of the first division of the zygote, the hatching process is appreciably more prolonged ending 72 hr following insemination. Mass hatching under such experimental conditions is observed in the interim between 56 and 72 hr. If the eggs are irradiated at later developmental stages the hatching process ends within 8-hour period between 48 and 56 hr after insemination, as in the control. However, the bulk of larvae in this instance is hatched approximately 4 hr or, in the case of irradiation occurring at the 32-blastomere or mid-blastula stages, 8 hr earlier than in controls. Thus, with the exception of the effects of embryonic injury

Table 65

Radiosensitivity of tench embryos at different developmental stages (single irradiation in a dose of 250 rad)

Developmental stage at the time of irradiation	Time elapsed after insemination	Total No. of eggs	No. of larvae hatched (%)	Including abnormal larvae (%)	No. of injured embryos (%)*
Precleavage	15 min	491	44.8	100	100
2-blastomere	1 hr 0.5 min	442	80.2	30.0	44.1
4-blastomere	1 hr 20 min	398	75.9	70.0	77.1
8-blastomere	1 hr 35 min	578	88.1	31.4	39.6
16-blastomere	2 hr 05 min	680	84.9	45.4	53.6
32-blastomere	2 hr 35 min	572	81.5	28.9	41.9
Mid-blastula	4 hr 50 min	604	85.3	54.0	60.7
Early gastrula	9 hr 20 min	708	84.7	10.0	23.7
Mid-gastrula	10 hr 50 min	683	85.2	18.6	31.0
Late gastrula	12 hr 20 min	608	85.5	9.6	22.8
Initial segmentation	21 hr 20 min	639	82.6	15.7	30.3
Initial movement	27 hr 40 min	692	86.9	22.4	31.8
Beginning of hatching	45 hr 20 min	678	85.0	14.3	27.4
Control	—	1 113	86.5	7.7	20.1

* Injured embryos: total of dead eggs and abnormal larvae.

Table 66

Average lifespan of larvae hatched from eggs irradiated at different developmental stages and subjected to additional irradiation in a dose of 1 500 rad 24 hr after hatching

Developmental stage at the time of irradiation	Average lifespan (days)			
	No additional irradiation	Coefficient of variation for average lifespan (C)	After additional irradiation	Coefficient of variation for average lifespan (C)
Precleavage	2.76 ± 0.22	68.8	0.00	—
2-blastomere	10.19 ± 0.36	35.3	4.92 ± 0.05	9.1
4-blastomere	8.20 ± 0.67	62.6	3.56 ± 0.15	28.9
8-blastomere	11.22 ± 0.42	36.1	4.96 ± 0.10	19.6
16-blastomere	12.23 ± 0.51	41.4	5.15 ± 0.09	16.0
32-blastomere	12.16 ± 0.49	40.3	5.31 ± 0.08	15.2
Mid-blastula	12.07 ± 0.55	45.5	4.38 ± 0.09	17.3
Early gastrula	14.30 ± 0.40	28.2	6.02 ± 0.04	6.6
Mid-gastrula	16.94 ± 0.18	11.0	5.30 ± 0.05	9.4
Late gastrula	17.00 ± 0.21	12.4	5.96 ± 0.06	10.4
Initial segmentation	15.20 ± 0.22	15.3	5.72 ± 0.05	8.6
Initial movement	14.82 ± 0.50	33.7	5.88 ± 0.04	5.9
Beginning of hatching	14.95 ± 0.23	15.2	7.41 ± 0.06	8.2
Immediately after hatching	15.97 ± 0.14	9.1	7.55 ± 0.06	7.8
Control	15.32 ± 0.11	7.2	6.87 ± 0.005	7.0

described above, irradiation of tench eggs in a dose of 250 rad at all developmental stages (except for the most radiosensitive stage preceding the first division of the zygote) results in a marked acceleration of hatching.

Data on the average lifespan of larvae hatched from eggs irradiated at different developmental stages demonstrate that the larvae hatched from eggs irradiated before the first division of the zygote are the first to die (Table 66). The lifespan of larvae from eggs irradiated at later developmental stages is, as might be expected, much longer. For stages of irradiation from the first division of the zygote (2-blastomere) to the mid-blastula stage, the lifespan of the larvae approximated 8 to 12 days and for subsequent stages it was 14 to 17 days, the mean lifespan period for controls being about 15 days.

Additional irradiation of larvae 24 hr after hatching in a dose of 1500 rad drastically reduced their lifespan in both controls and particularly experimental variants with preliminary irradiation. An exception were the last two experimental variants with preliminary irradiation of embryos immediately before and immediately after hatching. In these cases the average lifespan of larvae was somewhat longer than that in controls, i.e., in the larvae hatched from the nonirradiated eggs. As will be demonstrated below, this phenomenon is associated with the radioprotective effect of preliminary irradiation.

It should be noted that the death of larvae hatched from irradiated eggs was in all cases more protracted than in controls. This is borne out by higher variation coefficients as compared to controls for the average lifespan in experimental variants receiving irradiation. Following additional irradiation in a dose of 1500 rad, variation coefficients were sharply diminished, but for larvae preliminarily irradiated at earlier stages of eggs development, the coefficients were on the whole higher than in other experimental variants.

In the experiment with the mollusk *Limnaea stagnalis* L. (Kulikov and Famelis, 1970) clutches with eggs at different developmental stages were irradiated with ^{60}Co gamma rays in a dose of 300 rad at a dose rate of 36 rad/min. Thereupon the eggs were incubated concomitantly with the nonirradiated control in Koch dishes with lake water at a temperature of 18 to 20°C. The development of embryos was observed under an МБС-1 microscope. In experimental variants the death rate, number of embryos with different morphological abnormalities, and number of hatched normal specimens were recorded. In order to determine the presence or absence of a shell, all teratic embryos were examined under an МБИ-6 microscope in polarized light using cross-polarized lenses. Six replications of the experiment were run with 160 to 1600 eggs used for each experimental variant (Table 67).

Against a general background of reduction of radiosensitivity of embryos from earlier to later stages (resembling results in the tench eggs experiments), several periods of increased radiosensitivity may be distinguished. As with tench eggs, the first peak of radiosensitivity corresponds to the stage preceding the first division of the zygote; the second peak occurs in the period of transition from the 4- to

Table 67

Radiosensitivity of *Lymnaea stagnalis* L. embryos irradiated in a dose of 300 rad at different developmental stages

Developmental stage at the time of irradiation	Total No. of eggs	No. of injured embryos	Including (%)		Total	Probability of absence of differences between experimental variations (P)
			Abnormal specimens	Dead specimens		
Precleavage	169	94	42.0	13.6	55.6 ± 7.5	—
2-blastomere	853	228	18.2	7.8	26.1 ± 2.9	< 0.01
2-4-blastomere	243	74	22.5	7.2	29.7 ± 5.5	> 0.05
4-blastomere	447	206	42.5	9.4	51.9 ± 4.2	< 0.05
4-8-blastomere	157	94	47.4	13.1	60.5 ± 7.8	> 0.05
8-blastomere	346	99	27.9	7.5	35.5 ± 4.2	< 0.05
32-blastomere	252	39	11.1	3.7	14.8 ± 3.8	< 0.01
64-blastomere	132	6	1.5	3.1	4.6 ± 2.9	< 0.01
Final cleavage	592	152	19.3	4.7	24.0 ± 2.8	< 0.01
Early gastrula	619	103	12.7	4.6	17.3 ± 2.9	< 0.01
Mid-gastrula	801	140	13.1	3.9	17.0 ± 2.9	< 0.05
Late gastrula	850	170	13.6	2.4	16.0 ± 2.8	> 0.05
Trochophore	203	18	3.9	4.9	8.8 ± 2.1	< 0.05
Veliger	420	18	3.1	1.2	4.3 ± 1.4	< 0.05
Control	1 646	49	1.6	1.4	3.0 ± 0.8	—

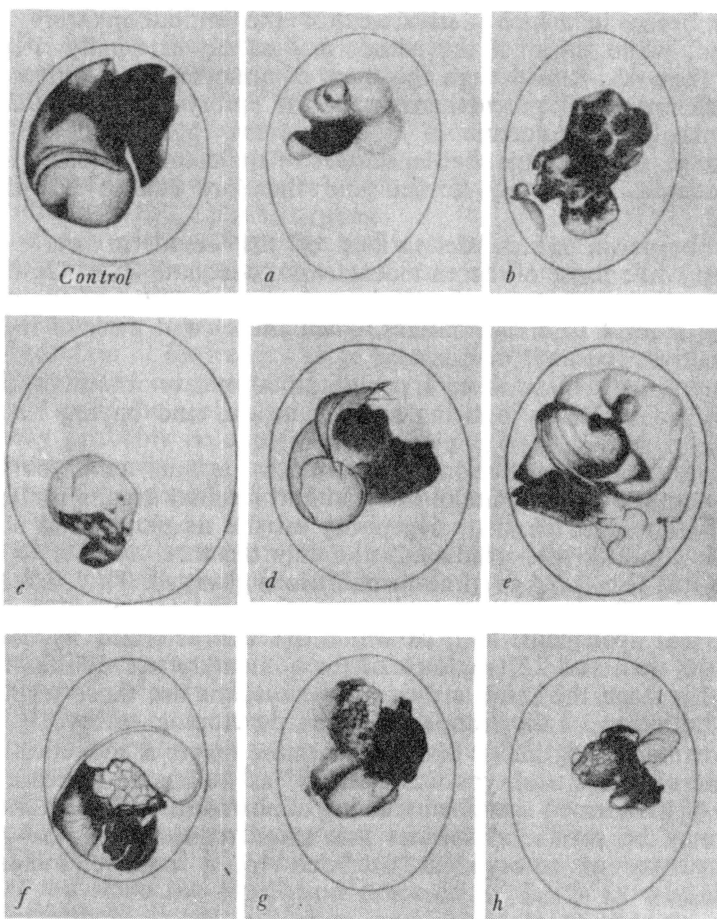


Fig. 42. Types of radiation-induced abnormalities in greater pond snail embryos ($\times 27.5$): (a), (b), (c) complex disorder in development of external and internal organs; (d), (e), (f) preponderant disorder in development of internal organs; (g), (h) preponderant disorder in development of ectoderm derivatives.

8-blastomere stage, and the third, less prominent peak is observed at the end of cleavage.

All of the numerous radiation-induced embryonic abnormalities observed in our experiments may be divided into the three following types, according to morphological evidence:

1. Embryos with a complex disorder of development which had markedly underdeveloped derivatives of the ectoderm (shell, mantle) and endoderm (absence of alimentary sac, reduction of yolk size, and other changes in internal organs). This type of teratism arises chiefly after irradiation at early developmental stages, although it may also occur after irradiation at later stages, except for the veliger stage (Fig. 42a-c).

2. Embryos in which derivatives of the endoderm were underdeveloped, while those of the ectoderm had no externally detectable damage (Fig. 42d-f). Although this type of abnormalities also occurred when different developmental stages of the embryo were irradiated, it was primarily characteristic of embryos from eggs irradiated at the veliger stage. At this stage the derivatives of the ectoderm (shell, mantle) are practically completely formed and therefore cannot be seriously damaged.

3. Embryos in which derivatives of the ectoderm were underdeveloped while those of the endoderm were adequate (Fig. 42g-h). This type of teratism was observed only in embryos irradiated at the stage of transition from 4 to 8 blastomeres, when the second peak of increased radiosensitivity became manifest.

Embryos with these three types of radiation abnormalities differed from control embryos in being smaller in size and having less body pigment.

The relative distribution of mollusk teratic embryos having developmental abnormalities of external and internal organs is shown in Table 68. One can see that, depending on the developmental stage at which the mollusk was irradiated, not only the total number of teratic embryos but also the type of abnormalities is changed. Thus, irradiation of eggs at the stage preceding cleavage results in a relatively high number of abnormal specimens, 78% of which are characterized by complete lack of the shell and 22% by lack of the alimentary sac. It thus follows that at this stage the more radiosensitive systems are those responsible for the initiation of the ectoderm in the developing embryo.

Following irradiation at the 2-blastomere stage, a reduction in the total yield of teratic embryos was recorded as well as an increase in the number of malformed specimens having abnormalities of the alimentary sac. It may be presumed that at this stage reduction of the general radiosensitivity of embryos is attended by a relative increase in

Table 68

Relative distribution of greater pond snail embryos with abnormalities of external and internal organs after irradiation in a dose of 300 rad at different developmental stages

Developmental stage at the time of irradiation	Total No. of experimental eggs	No. of abnormal embryos		
		Total	No shell	No alimenta- ry sac
Precleavage	169	42.0	77.7	22.3
2-blastomere	853	18.2	37.2	62.8
2-4-blastomere	243	22.5	52.5	47.5
4-blastomere	447	42.5	38.0	62.0
4-8-blastomere	157	47.4	33.0	67.0
8-blastomere	346	27.9	32.2	67.8
Final cleavage	592	19.3	9.0	91.0
Gastrulation	2 440	13.1	7.0	93.0
Trochophore	203	3.9	2.0	98.0
Veliger	420	3.1	0.0	100

radiosensitivity of those systems of a developing embryo which eventually lead to the formation of the endoderm.

Irradiation of embryos at the stage transitional between 2 and 4 blastomeres results in an approximately equal percentage of abnormalities in development of the external and internal organs, whereas irradiation at the subsequent stages of cleavage once again increases the relative number of embryos with developmental abnormalities of the internal organs.

At the early gastrulation stage and later stages the radiosensitivity of embryos is reduced once again, as indicated by a sharp decline in the total yield of teratic specimens. The overwhelming number of malformations are abnormalities of tissues derived from endoderm, while irradiation at the veliger stage results in 100% of abnormalities of this type. Such a great reduction in number of developmental abnormalities of the shell of mollusks irradiated at later developmental stages may probably be explained by a higher rate of specialization of the ectodermal cells as compared with the endodermal cells. After completing the specialization the cells become significantly more radioresistant. Indeed, at the trochophore and veliger stages the derivatives of the ectoderm function as fully formed integuments, whereas the specialization of endoderm derivatives is far from complete at these particular stages.

Finally, it should be noted that all abnormal embryos of the mollusk were unable to emerge from the egg capsule, and sooner or later died. After emergence of normal juvenile specimens from the clutches into the outer environment, the integrity of the cocoons was impaired and the remaining embryos were as a rule attacked by fungi.

The dependence of mollusk radiosensitivity on stage of embryonic development is demonstrated by the data in Tables 69 and 70, where the results of experiments with varying irradiation doses are presented. These data show the LD₅₀ of irradiated embryos to be about 450, 900, and 2000 rad when the irradiation occurred at the 2- to 4-blastomere stage, the embryonic motility stage, and the stage of shell formation, respectively.

In experiments with pike eggs, the dependence of embryonic radiosensitivity on water temperature was studied (Alshits *et al.*, 1970; Kulikov *et al.*, 1971b). The eggs were obtained from adult female pikes during the spawning season at Lake Maloye Miassovo. The fertilization was performed by the dry method; eggs obtained from a single female were mixed with the seminal fluid from two or three males. Immediately after fertilization (prior to first cleavage of the zygote) the eggs were irradiated with ⁶⁰Co gamma rays and then incubated in lake water at a water temperature of 10 to 20°C. The overall hatching data, the number of larvae with morphological abnormalities, and the number of cells with chromosome aberrations were recorded.

For cytological analysis the eggs were fixed at the late blastula stage with Carnoy fluid (6 parts alcohol, 3 parts acetic acid, 1 part chloroform). Fifteen minutes after introducing the eggs into the fixative, the egg membranes were pierced and the fixative was exchanged; 3 hr after

Table 69

Development of mollusk *Lymnaea stagnalis* L. embryos after irradiation of eggs at early cleavage stage (2 to 4 blastomeres)

Radiation dose (rad)	No. of eggs	No. of larvae hatched	% of total No. of eggs	P	No. of abnormal embryos	No. of embryos that died at different developmental stages (% of total No. of dead specimens)		
						Cleavage, gastrulation	Organogenesis	Shortly before hatching
Control	1 352	1 305	95.0	—	2	2.1	4.2	93.7
15	1 066	1 029	96.5	> 0.05	2	17.0	26.0	57.0
150	687	635	92.4	< 0.001	10	25.0	52.0	23.0
300	299	209	69.9	< 0.001	51	9.0	10.0	81.0
600	280	71	25.3	< 0.001	26	45.0	23.0	32.0
1 200	370	47	12.7	< 0.001	38	79.0	20.0	19.0
5 000	450	0	—	—	—	100	0	0

Table 70

Development of mollusk *Lymnaea stagnalis* L. embryos irradiated at stages of embryonic motility and initial formation of shell

Radiation dose (rad)	Embryonic motility stage				Initial shell formation stage			
	No. of eggs	No. of larvae hatched	% of total No. of eggs	No. of abnormal embryos	No. of eggs	No. of larvae hatched	% of total No. of eggs	No. of abnormal embryos
Control	466	453	97.2	0	707	692	97.7	2
15	498	490	98.6	0	488	477	98.0	5
150	458	446	97.5	0	556	534	96.0	9
300	471	463	98.0	2	568	538	95.0	2
600	494	485	98.5	2	493	480	97.6	4
1 200	580	9	1.5	3	581	555	95.5	7
1 500	480	0	0	—	430	263	61.2	13
5 000	540	0	0	—	550	44	7.5	—

beginning the fixation process, the eggs were transferred into 70% ethyl alcohol for storage.

The quantitative determination of chromosome aberrations was performed in squash acetolacmoid-stained preparations. Prior to staining the dissected germ disks were placed for 10 min into 30% lactic acid solution for maceration. From the germ disks stained in this manner provisional squash preparations were prepared which had an advantage of having all germ-tissue cells arranged in one layer and containing no artifacts in the form of fragments produced by sectioning with a microtome. In each preparation 50 anaphases were counted and the percentage of damaged cells was determined. A cell was regarded as damaged if it contained at least one bridge or fragment. The hatching of larvae was noticeably reduced in comparison with controls when the material was irradiated in doses exceeding 100 rad (Table 71). An increase in the number of abnormal embryos was noted within approximately the same dose range. A dose of 800 rad was absolutely lethal causing death of the embryos at the early gastrula stage. In the case of a water temperature of 20°C, a decrease in overall hatching was observed at a dose of 200 rad, whereas an increase in the yield of abnormal embryos occurred with 50 to 100 rad doses (Table 72). Under these conditions, as distinguished from an experiment at a lower temperature, the 400 rad dose proved to be absolutely lethal. The dependence of pike eggs radiosensitivity on the temperature level is particularly evident in Fig. 43. It shows that at the 10°C water temperature the number of abnormal larvae increases when relatively high radiation doses are applied, whereas at 20°C the number of abnormal larvae increases beginning with a 50 rad dose.

Since at 20°C the embryonic development of the pike was twice as rapid as at 10°C, it may be presumed that the observed reduction in radiosensitivity of embryos at a lower temperature is associated with recovery processes after irradiation which are more effective when the development of embryos is more protracted.

Table 71

Effect of ^{60}Co gamma-irradiation on early developmental stages of pike embryos at a water temperature of 10°C

Dose (rad)	No. of eggs	Larvae hatched					
		Total		Normal		Abnormal	
		No.	%	No.	%	No.	%
Control	488	343	71.0 ± 2.0	311	90.7 ± 1.6	32	9.3 ± 1.6
25	322	251	78.7 ± 1.9	235	93.6 ± 1.5	16	6.4 ± 1.5
50	452	347	77.0 ± 2.3	318	91.7 ± 1.6	29	8.3 ± 1.9
100	274	182	68.0 ± 2.8	170	93.4 ± 1.9	12	6.6 ± 1.8
200	302	145	47.5 ± 2.8	128	88.3 ± 2.8	17	11.7 ± 2.7
400	218	73	33.6 ± 3.2	143	59.0 ± 1.7	30	41.0 ± 5.8
800	229	Embryos died at early gastrula stage					

Table 72

Effect of ^{60}Co gamma-irradiation on early developmental stages of pike embryos at a water temperature of 20°C

Dose (rad)	No. of eggs	Larvae hatched					
		Total		Normal		Abnormal	
		No.	%	No.	%	No.	%
Control	207	146	70.7 \pm 3.1	131	89.7 \pm 2.5	15	10.3 \pm 2.5
25	394	325	83.0 \pm 2.4	275	87.7 \pm 1.8	40	12.3 \pm 1.8
50	278	206	74.0 \pm 2.6	173	84.0 \pm 2.6	33	16.0 \pm 2.7
100	326	249	75.0 \pm 2.4	172	72.8 \pm 2.8	67	27.2 \pm 2.8
200	246	122	49.5 \pm 3.3	53	43.0 \pm 1.4	69	57.0 \pm 4.6
400	305	Embryos died shortly before hatching					
800	290	Embryos died at late blastula stage					

It is noteworthy that insofar as the number of injured mitoses is concerned, differences between experimental embryos and controls are already noticeable at radiation doses of 25 to 50 rad (Fig. 44); with respect to the number of hatched abnormal larvae, the differences were noted at dosages of 50 and 200 rad, respectively, depending on the experimental conditions, while the differences in the total number of hatched larvae became evident only beginning with a 200 rad dose. Thus, chromosome aberrations in the cells of pike embryos appear to be a somewhat more sensitive criterion of radiobiological response as

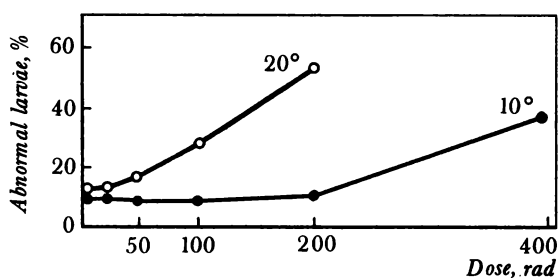


Fig. 43. Yield of abnormal pike larvae, incubated at different water temperatures, as a function of radiation dose received by the eggs.

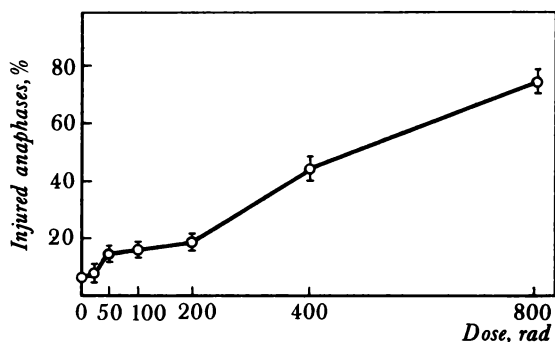


Fig. 44. Number of damaged anaphases in cells of pike embryos as a function of radiation dose (water temperature 10°C).

compared to the other criteria at issue. Irradiation of tench eggs in doses ranging from 25 to 200 rad does not change the total percentage of larvae hatched, yet the yield of larvae with various morphological abnormalities increases noticeably already at a dose of 50 rad (Table 73). A radiation dose of 400 rad at a water temperature of 20°C was found to be absolutely lethal, just as was the case with pike eggs: all embryos died immediately before hatching. With 800 rad the embryos died at the late gastrula stage, and with 1200 rad at the early gastrula stage (approximately 5 hr after irradiation). Survival data for hatched larvae demonstrated that the death of larvae hatched from the eggs irradiated in a dose of 200 rad occurred at a much higher rate than in controls. With radiation doses ranging from 25 to 100 rad the difference in survival of the experimental and control larvae is statistically nonsignificant.

It may be assumed that outwardly normal larvae hatched from eggs irradiated in relatively low doses have latent radiation damage which will adversely affect their survival rate after an additional irradiation of these larvae in a massive dose. To verify this assumption special experiments were undertaken (Kulikov *et al.*, 1969, 1970d). From the total number of the 2-day-old larvae hatched from the nonirradiated eggs, and from the larvae irradiated in doses of 25, 50, 100, and 250 rad, 200 outwardly normal larvae from each group were selected and divided into two subgroups of 100 specimens each. One subgroup was irradiated in a dose of 4000 rad while the other subgroup served as a control not subjected to additional irradiation. During the next 10 days the survival rate of the larvae was recorded (Fig. 45).

In the absence of additional irradiation the survival rate decreased

Table 73

Effect of a single ^{60}Co gamma-irradiation of tench eggs at 2-blastomere stage and at a water temperature of 20 to 23°C on development of embryos

Dose (rad)	No. of eggs	Larvae hatched						P
		Total		Normal		Abnormal		
		No.	%	No.	%	No.	%	

Control	2 341	1 992	82.5 ± 2.3	1 854	92.8	138	7.2 ± 2.1	—
25	3 151	2 657	84.0 ± 4.2	2 516	94.6	141	5.4 ± 1.7	>0.05
50	2 234	1 847	83.0 ± 2.8	1 678	90.8	169	9.2 ± 2.2	<0.05
100	2 016	1 675	83.0 ± 3.1	1 458	87.0	217	13.0 ± 4.2	<0.01
200	2 544	2 022	80.0 ± 4.0	1 391	67.3	322	32.7 ± 3.0	>0.001
400	3 010	Embryos died shortly before hatching						
800	2 950	Embryos died at late gastrula stage						
1200	2 116	Embryos died at early gastrulation stage						

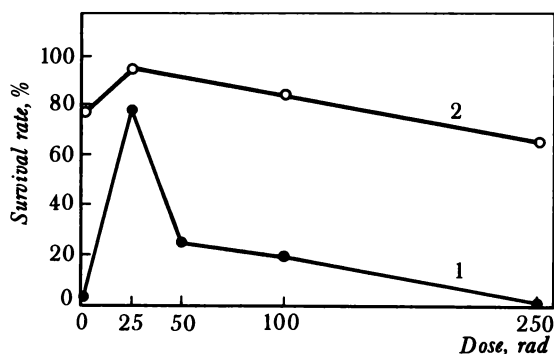


Fig. 45. Survival rate of tench larvae on day 10: (1) after additional irradiation in a dose of 4 kilorad; (2) without additional irradiation.

only in the case of larvae hatched from eggs irradiated in a dose of 250 rad. In the variants of the experiment with eggs irradiated with 25 and 100 rad the relative number of survivors was even somewhat greater than that in the control. However, after additional irradiation in a dose of 4000 rad all of the larvae hatched from the control eggs died by the tenth day after the irradiation. Similar effect was recorded when the eggs were irradiated in a dose of 250 rad. After the additional irradiation the survival rate of larvae irradiated in doses of 25, 50, and 100 rad was fairly high, the maximum value being 78% at 25 rad. Thus, the latent radiation effects which we expected to occur after irradiation of the eggs in relatively low doses not only did not reduce the survival rate of larvae hatched from these eggs after their additional irradiation in a sublethal dose, but, on the contrary, had a marked radioprotective action similar to that described in the foregoing section with respect to pea and pine seeds. The radioprotective effect of low-dose irradiation appears to be of general biological importance and closely associated, as has already been mentioned, with the mechanism of radiostimulation.

Radiosensitivity of Freshwater Organisms under Conditions of Radioactive Contamination of the Aqueous Medium

In recent years, because of the hazard of radioactive contamination of waterbodies detrimental to their bioproductivity, there has been growing interest in the biological effect of higher radiation levels arising from radioactive contamination of the habitats of hydrobionts and the accumulation by hydrobionts of radionuclides.

The extremely high radioresistance of the waterbody bacterial flora deserves emphasis. Even at a $^{90}\text{Sr}/^{90}\text{Y}$ concentration in water reaching 10^{-3} Ci/liter, no changes in the development of nitrifying bacteria and *Escherichia coli* could be found (Zhogova, 1961). In the water in which the Los Alamos atomic reactor is submerged, there were discovered bacteria *Pseudomonas* which retained their reproductive ability in the water that accumulated a dose of 10 Mrad per 8 hr of irradiation (Dubinin, 1961).

In experiments with freshwater periphyton at a concentration of a nonseparated mixture of uranium fission fragments in water of from 10^{-6} to 10^{-3} Ci/liter radiostimulation of the development of this organism was noted. Along with stimulation of the total mass of the community, a marked rearrangement of its species composition took place (Timofeyeva-Resovskaya, 1963). Stimulation of the development of freshwater algal periphyton was also observed when it was chronically irradiated for 2 months in a gamma-field in doses ranging from 5 to 50 rad/day (Gileva *et al.*, 1964). In experiments with protococcal algae and infusoria lasting for 23 days no noticeable changes in the development and accretion of the biomass of these organisms were observed under conditions of contamination of the medium with the radionuclide mixture* to a level of 10^{-7} to 10^{-5} Ci/liter (Guskova *et al.*, 1971a, b).

The inhibitory effect of $^{90}\text{Sr}/^{90}\text{Y}$ on the reproduction of the freshwater flea *Daphnia magna* was noted only when the organism was cultivated for 80 days in water with a radioactivity level of 10^{-5} Ci/liter (Telitchenko, 1958).

In another study a natural population of the *Chironomus tentans* mosquito was cytologically investigated; great quantities of the larvae of this mosquito are present in the bottom deposits of White Oak Lake and White Oak Creek, into which radioactive waste was for a long time discharged by the United States Oak Ridge National Laboratory. A population of this mosquito at the larval stage was irradiated for 22 years in a yearly dose of about 230 rad. It was found that under these conditions the frequency of new chromosome aberrations in the mosquito population was somewhat increased, but later these aberrations were eliminated by natural selection; adaptability of the population to new conditions did not undergo any essential changes (Blaylock, 1966).

Our laboratory carried out experiments to study the development of embryos of the freshwater mollusk *Lymnaea stagnalis* L. in water with a varying $^{90}\text{Sr}/^{90}\text{Y}$ content (Kulikov *et al.*, 1966; Kulikov and Timofeyeva, 1967; Kulikov, 1971a; Timofeyeva *et al.*, 1971). Decrease in the percentage of hatching larvae was recorded only at the level of radioactivity of the water being in the order of 10^{-4} to 10^{-3} Ci/liter when the total radiation dose to the eggs during the incubation period exceeded 350 rad (Table 74). In solutions with a concentration of 10^{-3} Ci/liter, there was, along with an increase in death of embryos, a noticeable delay in embryonic development, as well as a weaker pigmentation and diminution of embryo size. Death of embryos, as observed in the experiments at high radionuclide concentrations, occurred at the late developmental stages shortly before hatching. It was only at a concentration of 10^{-2} Ci/liter that the embryos kept in the radioactive water died during the first day. It was found that a single acute irradiation of the mollusk eggs with ^{60}Co gamma rays at the early

* The radionuclide mixture had the following composition: 52% of rare-earth radionuclides (^{91}Y , $^{141,144}\text{Ce}$, ^{143}Pr , ^{147}Nd , ^{140}La) and 18% of alkaline-earth radionuclides (^{89}Sr , ^{140}Ba) and also ^{95}Zr and ^{95}Nb .

Table 74

Effect of $^{90}\text{Sr}/^{90}\text{Y}$ on embryonic development of greater pond snail
(*Lymnaea stagnalis* L.)

Water radio-activity (Ci/liter)	8-day radiation dose (rad)	Total No. of eggs	No. of larvae hatched	% of total No.	P	No. of abnormal embryos
Control	—	553	544	98.0	—	1
1×10^{-9}	0.0035	539	527	98.0	—	0
1×10^{-7}	0.35	497	481	97.2	—	2
1×10^{-6}	3.5	266	262	98.5	—	1
1×10^{-5}	35.0	653	625	96.0	—	5
1×10^{-4}	350.0	602	584	97.0	> 0.05	3
5×10^{-4}	1700.0	366	335	91.5	< 0.001	4
1×10^{-3}	3500.0	501	308	61.5	< 0.001	19
1×10^{-2}	35000.0	550	0	0	—	—

cleavage stage was more effective than their chronic irradiation at about the same dose during incubation in the $^{90}\text{Sr}/^{90}\text{Y}$ solutions (Kulikov *et al.*, 1966).

The above examples demonstrate that members of different taxonomic groups of freshwater organisms, from bacteria to developing mollusk eggs, can for some time tolerate fairly high levels of radioactive contamination of the aqueous medium, escaping apparent injuries. Of great interest in this connection are data on the extremely high radiosensitivity of rapidly developing eggs of some fish species (Polikarpov and Ivanov, 1961, 1962; Polikarpov, 1964; Fyodorov *et al.*, 1964; Ivanov, 1967). These investigators have shown that incubation of fertilized eggs in an aqueous solution of $^{90}\text{Sr}/^{90}\text{Y}$ at a concentration of 10^{-10} Ci/liter and above increases the number of hatched larvae with various morphological abnormalities, and that at concentrations of 10^{-8} to 10^{-7} Ci/liter there is, in addition, a considerable elevation of the embryo mortality rate. Similar results were obtained in experiments with an aquarium fish (*Zebra danio*) when the eggs of this fish were incubated in solutions of a radioactive mixture of salts from rain water (Mikami *et al.*, 1956). At the same time there are data pointing either to the absence of such effects in individual fish species when the radioactive contamination of water is quite intensive (Brown and Templeton, 1964; Guskova *et al.*, 1971a, b; Pitkanen, 1971) or to the fact that such effects are observed when the incubated eggs remain for very long periods (over 100 days) in water with a ^{90}Sr concentration of 10^{-8} to 10^6 Ci/liter (Neustroyev and Podymakhin, 1966).

To correctly appraise the biological effectiveness of irradiation of fish embryos developing in water contaminated with various nuclides it is necessary to have an idea of the dose burden sustained by the eggs during the incubation period. Such calculations have been performed for eggs of some freshwater fish species developing in aqueous solutions of the major fission fragment radionuclides at a concentration of 10^{-5}

Ci/liter (Kulikov *et al.*, 1970a, b; 1971c). Two extreme variants of the distribution of the nuclides in the eggs were considered: (1) that the radionuclides derived from the environment are accumulated predominantly on the external surface of the egg membrane, while the radioactivity of the inner egg content equals that of water; and (2) that the radionuclides are evenly distributed throughout the egg volume.

In the former case the dose rate in the egg center is derived from the radioactivity of its content and that of the environment, and also from the radioactivity of the egg membrane. If we assume that the radioactivity of the liquid filling the egg does not differ from that of the environmental water, and that the membrane that sorbs nuclides is infinitely thin, the total dose rate (D) may be defined according to the following equation:

$$D = 1.44 \frac{TAL_\beta}{\rho} (1 - e^{-\frac{0.693t}{T}}) (1 - e^{-\mu R_m} + \frac{\mu R_n C_A}{3} e^{-\mu R_n}) \text{ rad/sec} \quad (1)$$

In the second case the dose rate in the egg center depends on the radioactivity of the aqueous medium and on that of the entire egg, the accumulation coefficient also being taken into account. The total dose rate is then defined as

$$D = 1.44 \frac{TAL_\beta}{\rho} (1 - e^{-\frac{0.693t}{T}}) [C_A (1 - e^{-\mu R_n}) + e^{-\mu R_n} - e^{-\mu R_m}] \text{ rad/sec} \quad (2)$$

where T is the half-life period in seconds; t is the duration of egg incubation in the radioactive water in seconds; A is the water radioactivity, equal in our case to 10^{-2} $\mu\text{Ci/ml}$; ρ is the water density equal to 1 gm/ml; μ is the attenuation coefficient of beta particles (cm^2/gm); R_m is the maximum range of beta particles in water (gm/cm^2); R_n is the egg radius (gm/cm^2); C_A is the accumulation coefficient;

$$L_\beta = 3.7 \times 10^4 \times 1.6 \times 10^{-8} \bar{E}_\beta = 5.9 \times 10^{-4} \bar{E}_\beta \frac{\text{rad} \cdot \text{gm}}{\mu\text{Ci} \cdot \text{sec}}, \text{ where}$$

$$\bar{E}_\beta = \sum_i^n E_i \eta_i; \bar{E}_i \text{ is the energy of single lines of the beta spectrum;}$$

and η_i is the relative yield of these lines in the particular radionuclide spectrum.

Calculations have shown that in the case of surface distribution of radionuclides on the egg membrane total radiation doses during the incubation period are somewhat lower than in the case of uniform distribution of radionuclide throughout the egg volume. This is associated with the energy characteristics of the radionuclides which are described by different equations (1) and (2). The real values of the radiation doses should lie somewhere between the two extreme variants we considered, since the radionuclides accumulated in the spawn are not completely retained by the membranes but pass partially inside the eggs.

Table 75 shows coefficients of accumulation for six fission fragment radionuclides with respect to eggs of the carp (*Carassius carassius* L.), tench (*Tinca tinca* L.), perch (*Perca fluviatilis* L.), and pike (*Esox lucius* L.). In the same table irradiation doses for the eggs are shown as calculated for the case of the uniform distribution of radionuclides throughout the egg volume. Duration of the incubation period for the eggs of the carp and tench (at a water temperature of 20°C) was 7 and 2 days, respectively, and for the eggs of the perch and pike (at a water temperature of 10°C) it was 15 and 8 days, respectively.

Table 75 demonstrates that the accumulation coefficients are fairly variable, depending on the biological features of various fish species and the chemical nature of different radionuclides. Accordingly, radiation doses fluctuate between 0.07 rad for the tench and perch eggs in a $^{106}\text{Ru}/^{106}\text{Rh}$ solution and 28.66 rad for the perch eggs in a $^{90}\text{Sr}/^{90}\text{Y}$ solution.

The experiments designed to study the impact of different levels of radioactive substances in the aqueous medium on the development of embryos of freshwater fish were conducted using solutions of ^{90}Sr in equilibrium with ^{90}Y (Kulikov *et al.*, 1968c; Timofeyeva and Alshits, 1970; Timofeyeva *et al.*, 1971).

In experiments with fertilized tench eggs that developed in the lake water contaminated with ^{90}Sr at concentrations ranging from 10^{-10} to 10^{-5} Ci/liter, we failed to find significant differences in comparison with the control either with respect to the development rate of embryos or with respect to the number of normal or abnormal larvae hatched from these eggs (Table 76). The survival rate of larvae in different variations of the experiment during the first 10 days following hatching was also similar.

It might be expected that, despite their externally normal state, the larvae from the eggs incubated in radioactive solutions would be less resistant to subsequent irradiation with gamma rays of ^{60}Co . For this

Table 75

Coefficients of radionuclide accumulation for eggs of freshwater fish (per wet weight) by end of incubation period and approximate radiation doses received by embryos during their development until larva hatching in aqueous solutions having a radioactivity of 10^{-3} Ci/liter

Nuclide	Accumulation coefficient				Radiation dose (rad)			
	Crucian carp	Tench	Perch	Pike	Crucian carp	Tench	Perch	Pike
^{90}Sr	8.0	4.0	5.0	1.2	3.18	0.41	6.00	0.80
^{90}Y	31.0	11.0	25.0	54.5	10.04	1.16	22.66	25.13
$^{95}\text{Zr}/^{95}\text{Nb}$	—	23.7	5.1	—	—	1.24	1.90	—
$^{106}\text{Ru}/^{106}\text{Rh}$	—	26.2	6.8	—	—	0.07	0.07	—
^{137}Cs	—	1.5	1.7	—	—	0.41	1.75	—
$^{144}\text{Ce}/^{144}\text{Pr}$	—	40.5	17.8	—	—	1.44	3.82	—

Table 76

Effect of different levels of $^{90}\text{Sr}/^{90}\text{Y}$ content in lake water on development of tench embryos

Nuclide concentration in water (Ci/liter)	Radiation dose (rad)	Total No. of eggs	Larvae hatched				
			% of total No. of eggs	Normal		Abnormal	
				No.	%	No.	%
Control	—	2810	75.0 ± 4.1	1963	93.2	143	6.8
10 ⁻¹⁰	0.0000157	2931	72.3 ± 5.1	1968	93.0	150	7.0
10 ⁻⁹	0.000157	3045	70.2 ± 8.1	2009	94.0	127	6.0
10 ⁻⁸	0.00157	2772	74.0 ± 5.1	1900	92.5	154	7.5
10 ⁻⁷	0.0157	2951	71.0 ± 7.2	1965	93.9	127	6.1
10 ⁻⁵	1.57	1597	77.0 ± 2.4	1146	93.1	85	6.9

purpose the 3-day-old larvae obtained in different experimental variations were irradiated in a dose of 800 rad and their survival rate was then observed during 5 days following their irradiation. However, even in this case the survival rate of larvae in all variations of the experiment was similar. Thus, in the experiments described none of the indices we adopted showed any deviations from the norm when tench eggs developed in the water containing ^{90}Sr in concentrations from 10^{-10} to 10^{-5} Ci/liter.

Absence of a biological effect may be explained by a too low dose level in the irradiation of embryos during their development in radioactive solutions. The maximal irradiation dose even at the highest concentration of ^{90}Sr in water (10^{-5} Ci/liter) was only 1.6 rad. This dose accumulated gradually throughout the period of embryogenesis and the principal dose burden fell on the later stages of embryo development, characterized by a higher radioresistance. If we consider that the minimal dose of a single gamma-irradiation of tench eggs resulting in detectable disorders in embryo development is 50 to 100 rad (Table 73), a dose several scores lower, and, besides, highly prolonged in time, must obviously be still less effective.

In experiments with perch eggs we have also failed to detect any disorders in embryo development in water contaminated with $^{90}\text{Sr}/^{90}\text{Y}$ in concentrations ranging from 10^{-10} to 10^{-5} Ci/liter. The highest radiation dose for the perch eggs at a water radioactivity level of 10^{-5} Ci/liter, calculated for the case of uniform distribution of radionuclides throughout the egg volume, was about 29 rad. The real radiation dose should be 3 to 4 times lower since an appreciable proportion of radionuclides accumulated in the spawn is concentrated on egg membranes, thus reducing the total radiation dose.

At a $^{90}\text{Sr}/^{90}\text{Y}$ solution concentration of about 10^{-4} Ci/liter, corresponding to a total radiation dose of about 260 rad, an increase in the percentage of hatched pike larvae with morphological abnormalities is

Table 77

Effect of different levels of $^{90}\text{Sr}/^{90}\text{Y}$ content in lake water on development of pike embryos

Nuclide concentration in water (Ci/liter)	Radiation dose (rad)	Total No. of eggs	Larvae hatched				
			% of total No. of eggs	Normal		Abnormal	
				No.	%	No.	%
Control	—	445	72 ± 2	289	89.8	33	10.2 ± 1.9
10 ⁻⁹	0.00259	400	74 ± 2	256	86.5	40	13.5 ± 2.0
10 ⁻⁸	0.0259	316	72 ± 3	204	89.5	24	10.5 ± 2.0
10 ⁻⁷	0.259	369	66 ± 3	208	85.0	37	15.0 ± 2.3
10 ⁻⁵	25.93	318	80 ± 2	223	88.0	31	12.0 ± 2.0
10 ⁻⁴	259.3	378	75 ± 2	224	79.0	60	21.0 ± 4.1

noted (Table 77). Similar changes are observed when the pike eggs are externally irradiated with ^{60}Co gamma rays in a single dose of about 50 to 100 rad.

Thus, at similar doses of radiation a single external gamma-irradiation of eggs at early cleavage stages is biologically more effective than prolonged irradiation of embryos during incubation in water containing a corresponding concentration of $^{90}\text{Sr}/^{90}\text{Y}$ (10⁻⁵ to 10⁻⁴ Ci/liter). The number of damaged cells significantly increased, compared to controls, beginning with a $^{90}\text{Sr}/^{90}\text{Y}$ concentration of 10⁻⁵ Ci/liter.

In the experiments we carried out with eggs of a freshwater greater pond snail (*Lymnaea stagnalis* L.), as well as developing tench, perch, and pike eggs, we failed to find significant differences from the control either in the mortality rate of embryos or in the quantitative yield of abnormal larvae when the range of $^{90}\text{Sr}/^{90}\text{Y}$ concentrations in water was 10⁻⁹ to 10⁻⁵ Ci/liter. It was only through cytological analyses of the pike germ

Table 78

Number of injured anaphases and telophases in pike embryo cells at late blastula stage as dependent on $^{90}\text{Sr}/^{90}\text{Y}$ concentration in water

Nuclide concentration in water (Ci/liter)	No. of dividing cells studied	Injured anaphases and telophases		Cells with bridges		Cells with fragments	
		No.	%	No.	%	No.	%
Control	500	57	11.4 ± 1.4	6	1.2 ± 0.5	48	9.6 ± 1.3
10 ⁻⁹	500	44	8.8 ± 1.3	5	1.0 ± 0.4	42	8.4 ± 1.3
10 ⁻⁸	500	55	11.0 ± 1.4	14	2.8 ± 0.7	47	9.4 ± 1.3
10 ⁻⁷	500	78	15.6 ± 1.6	7	1.4 ± 0.5	46	9.2 ± 1.3
10 ⁻⁵	500	88	17.6 ± 1.7	15	3.0 ± 0.8	88	16.6 ± 1.7
10 ⁻⁴	500	142	28.4 ± 2.1	30	6.0 ± 1.1	115	23.0 ± 1.9

cells that a higher yield of injured cells was detected, beginning with a concentration of 10^{-5} Ci/liter and over. The data obtained support the conclusion that with similar doses of radiation a single external gamma-ray irradiation of the mollusk and fish eggs at early cleavage stages is biologically more effective than a prolonged irradiation of embryos during their development in water containing $^{90}\text{Sr}/^{90}\text{Y}$ (Table 78).

It should be noted that currently available data on the damaging action of a low level radioactive contamination of the aqueous medium on the embryogenesis of some sea and freshwater fish are not consistent. This shows that the problem of radiosensitivity of fish at early ontogenetic stages is not sufficiently studied, and also points to the likelihood of great differences in the sensitivity of various fish species to irradiation resulting from the radioactive contamination of the aqueous medium and the accumulation of radioactive substances by developing embryos.

For the successful solution of these problems, we believe, it would be necessary, in addition to an expansion of studies on the effect of radioactive substances on the early ontogenesis of various fish species, to include in the sphere of radioecological studies as many members of other taxonomic groups of hydrobionts as possible. Convenient objects should be found permitting more prolonged studies that would cover the entire cycle of the ontogenetic development of organisms. In carrying out research of this type it is extremely important to employ radiogenetic methods.

Effect of Ionizing Radiation on Communities of Organisms

Under conditions of natural habitats, living organisms are represented in terms of more or less complex communities (biocenoses) in which all of the forms are closely associated both with each other and with environmental components, through multiform ecological links. In such a system any disturbance in the development or living conditions of a single species (especially among the species dominating a particular biocenosis) usually entails a number of secondary phenomena leading to a disturbance of the previously existing dynamic equilibrium and to a rearrangement of the entire community. Therefore, in the intricate complex of responses of a biocenosis to irradiation, one should not only note the primary phenomena associated with the immediate effect of radiation on individual organisms, but also the secondary responses that manifest themselves in the form of various rearrangements of a biocenosis as a whole, produced by disorders in biocenotic bonds between the components of the biocenosis. Owing to a broad variability in the radiosensitivity of various species, their responses to the same radiation dose will be different. At the same time the degree of radiation response is defined by both the radiosensitivity of a particular species and its competitive interrelations with other organisms in a biocenosis

(Kulikov *et al.*, 1962; Aleksakhin, 1968; Aleksakhin *et al.*, 1970; Tikhomirov, 1972).

A comprehensive study of changes occurring in biocenoses under the effect of ionizing radiation may be exemplified by the work carried out in the United States in radiation fields around powerful radiation sources, including various natural forest areas, discarded fields and cultivated plots (Sparrow, 1960; Ohba and Simak, 1961; Woodwell and Sparrow, 1963a, b; Woodwell, 1962, 1965; Brower, 1965; McCormick, 1966; Sparrow and Woodwell, 1962; Woodwell and Oosting, 1965; Sparrow *et al.*, 1965; Platt, 1963).

These investigations corroborated an extremely high radiosensitivity of coniferous tree species and made it possible to estimate the radiation dose levels resulting in various shifts in the composition and structure of the communities under study attributable to the radiation exposure profile, season, density of planting, and other factors.

For experimental studies of the effects of ionizing radiation, apart from radiation fields, other methods can be used. For instance, it is feasible to isolate small plots inhabited by a certain type of phytocenosis and to introduce into these plots definite amounts of radionuclides thus modelling radioactive fallout, and to record the changes occurring in such a phytocenosis. Additionally, it is possible to build up under experimental conditions artificial phytocenoses by planting in small plots seed mixtures of various plant species. In such studies the radioactive substances may be introduced into community habitats, or such phytocenoses may be formed by the planting of seeds preliminary wetted in radioactive solutions having differing concentrations of radionuclides. Finally, such plant communities may be set up by planting a mixture of seeds subjected to external irradiation prior to planting.

Our laboratory has conducted a series of experiments involving both the introduction of emitters into community habitats and the preplanting wetting of seeds in solutions of these emitters (Kulikov, 1957a, b; Kulikov *et al.*, 1962; Timofeyev-Resovsky, 1957; Timofeyev-Resovsky *et al.*, 1957a, b; Timofeyeva-Resovskaya, 1963).

An experiment was carried out to test the overall effect of radioactive substances on phytocenoses. Specifically matched communities comprising 15 plant species were grown on beds having a differing soil content of nonseparated uranium fission fragments. In the beds with lower amounts of emitters (1 and 5 Ci/m²) the number and weight of plants during the vegetative season were markedly higher than in the control beds. At the same time in beds with a higher content of radioactive fission fragments (25, 50, and 100 mCi/m²), inhibition of phytocenoses was evident (Fig. 46).

An analysis of plant communities according to constituent species has demonstrated that various species and species groups composing a community respond in different ways to the introduction of emitters into the soil. One group of plants (wheat, barley, millet, flax, pea, pea-vine, lentil, and blue lupine) showed the usual effects of radiostimulation at low and inhibition at higher concentrations of radionuclides in the soil. Another plant group (European yellow lupine, sainfoin, lucerne, and

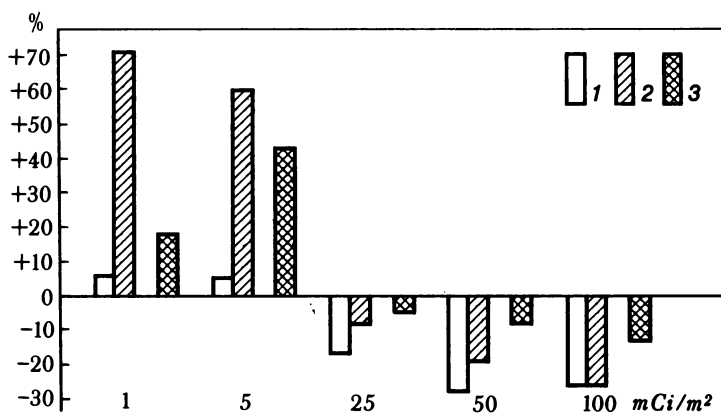


Fig. 46. Percent deviation from control of the number (1), total weight at age of 7 to 8 weeks (2), and total dry weight at final harvesting (3) of plants grown on experimental beds with varying radioactivity levels.

clover) was also stimulated at low emitter doses, but had no appreciable deviations from the control at higher doses, except one. With the highest radionuclide concentration in the soil (100 mCi/m²) the plants once again produced the biomass in an amount appreciably exceeding that of the control community (Fig. 47). One can note that this unusual response to high radionuclide concentrations in the soil is shown only by the species that constitute the lowest storey of the plant community (Fig. 48).

Especially drastic changes in the composition and structure of a plant community were noted in experiments involving the preplanting wetting of a seed mixture using different concentrations of uranium fission fragment solutions. Based on the average agrotechnical seeding standards, seeds of 30 species of cultivated plants were selected for these experiments. In the control the seeds were subjected to 24 hr wetting in tap water, while in three experimental variations wetting was performed

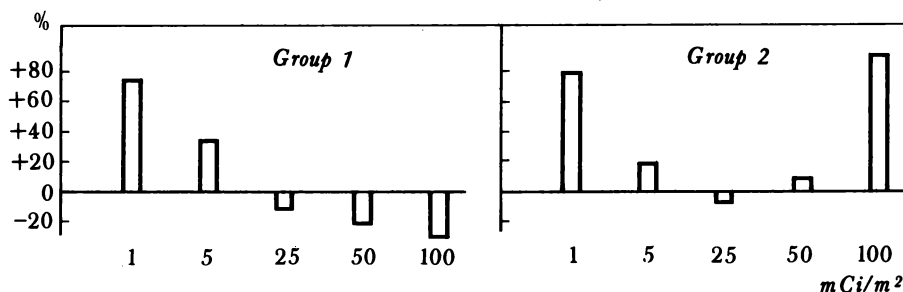


Fig. 47. Percent deviation in weight of plants on five beds with increasing amount of emitters in soil. Averaged data for two groups of plant species; group 1: wheat, barley, oats, millet, flax, pea, peavine, lentil, blue lupine; group 2: yellow lupine, sainfoin, lucerne, clover.

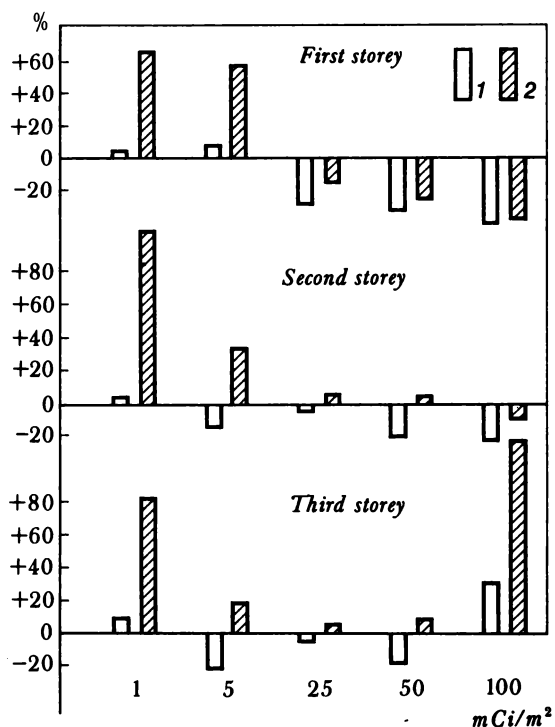


Fig. 48. Percent deviation from control of the number (1) and weight (2) of plants through three storeys on five beds with differing content of emitters in the soil.

in a solution of uranium fission fragments stored for a long time and having concentrations of 2, 20, and 200 mCi/liter (for convenience these concentrations will be subsequently referred to as the first, second, and third doses, respectively).

The solution of uranium fission fragments consisted chiefly of ^{89}Sr , ^{90}Y , ^{95}Zr , ^{106}Ru , ^{137}Cs , and ^{144}Ce .

After wetting the seeds were rinsed with tap water, dried, and planted in a 1×10 m plot in five replicate lots. In each plot as much as 1816 gm of seeds were planted with the resulting overcrowding. This overcrowding, creating harsh conditions of fighting for edaphic space, rendered the community much more responsive (with respect to qualitative and quantitative composition) to various exposures. Experimental plots were laid out in a single row with the successive distribution of experimental variations and replications. The total number of planted plots was 20.

During the vegetative season visual observations were carried out and time of emergence of shoots, of florescence, and maturation was recorded for each species of the community, including time of emergence of the ear (panicle) for annual cereals. Twice during the season and on termination of the experiments the number of specimens was counted separately for each species grown in the plots. In the course of this work the storey to which each species was confined in the community was

recorded and the biomass was assayed (Fig. 49).

When the first two doses of emitters were applied, the community structure closely resembled that of the control; in these three variations (control, first, and second doses) during the vegetative period and until the second determination of the plant distribution between storeys, the rearrangement was negligible. The third dose community, however, sharply differed from controls already at the onset of vegetation: bean and haricot fell out of the community, chick pea passed from the first to the third storey, mustard shifted from the second to the first storey;

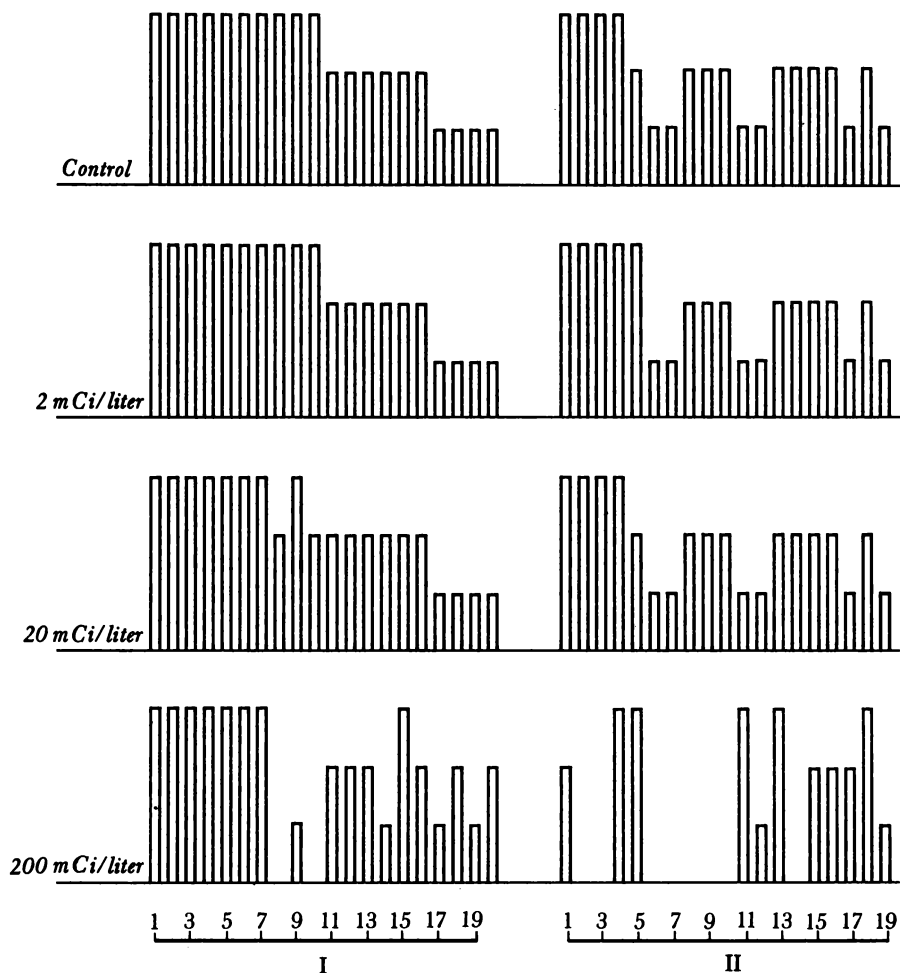


Fig. 49. Histogram of storey distribution of species in 3- to 4-week old (I) and 2.5-month old (II) plant communities that developed under effect of differing doses of emitters: (1) wheat; (2) oats; (3) barley; (4) vetch; (5) hemp; (6) lentil; (7) rye; (8) beans; (9) chick pea; (10) haricot; (11) millet; (12) perennial cereals; (13) pea; (14) soybean; (15) mustard; (16) buckwheat; (17) clover; (18) falseflax; (19) carrot; (20) spinach.

soya moved from the second to the third storey, and falseflax and spinach moved from the third to the second storey. Later the picture underwent still greater changes. Apart from bean and haricot, oats, barley, rye, lentil, chick pea, soya, and spinach dropped out of the community. Wheat went from the first to the second storey, pea and falseflax passed from the second to the first storey, and the perennial cereals shifted to the third storey; finally, millet ascended from the third to the first storey, and clover from the third to the second storey. Thus, in this variation, there was formed, in comparison with the control, an absolutely different, much scantier community whose distribution over the storeys was also altered.

The general impression produced in the wake of all observations of the plant community development may be summarized as follows: the first dose resulted in a slight stimulation of the community with no perceptible disturbance of its structure; the second dose induced a slight inhibition of some species leading to hardly discernible shifts in the community structure; the third dose resulted in complete elimination of a number of the species dominant in the control community, and this, in turn, led to a scantier composition of the community, to a reduction of its biomass, and to essential rearrangements in its structure.

A detailed quantitative analysis of the communities, involving the counting and weighing of all plant species in different vegetative periods, and a quantitative determination of the storey dynamics during community development, fully corroborate the qualitative appraisal of experimental phytocenoses made during visual observation, and to a large measure improve the accuracy of this appraisal.

The number of plants in the communities formed on exposure to the first radiation dose somewhat increased throughout the vegetative period as compared with the control, whereas the mean weight of a single plant differed hardly at all from that in the control community; as a result the total biomass per unit area was somewhat greater than that in the control. After the second dose the number of plants during the entire vegetative period was lower than that in the control community, but the mean weight of a single plant was markedly higher and the resultant total weight of the plant mass per square meter was almost the same as in the control. When the third dose of emitters was applied the community, from the very beginning, was quite sparse. Later, the number of plants decreased still further, so that at the height of vegetation their total number was one-fourth that of the control community. However, since this reduction was attended by a substantial increase in single-plant weight, the total weight of the plant mass per square meter, though being lower than in the control community, was still not as much reduced as the number of plants (Fig. 50).

The relative distribution of total plant numbers for each species of the first radiation dose community was very close to that of the control, with a tendency to slightly exceed the control values.

In the communities formed under the effect of the second dose of radiation the number of hemp plants decreased sharply and that of millet specimens increased. The number of the remaining species was

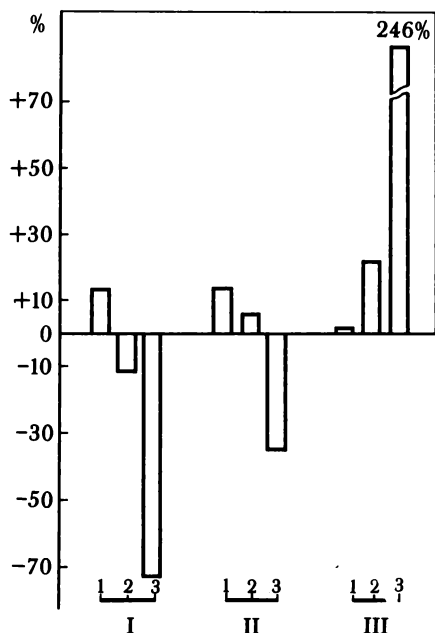


Fig. 50. Percent deviation from control of total number of plants per square meter (I), total weight of plants per square meter (II), and mean weight of a single plant (III) in communities formed under effect of differing doses of emitters: (1) 2 mCi/liter; (2) 20 mCi/liter; (3) 200 mCi/liter.

more or less uniformly decreased in accordance with the general slight decrease in the number of plants per unit area. In the third dose communities all cereals and a part of the annual leguminous plants were almost completely eliminated, while the total number of other plants (vetch, lentil, pea, buckwheat, hemp, and others) was decreased in proportion to the overall decrease in the number of plants per square meter. Some species (some perennial leguminous plants and millet) in this experimental variant unexpectedly increased in number almost twofold over that in the control community.

Considering the weight of plants by groups and by individual species it is notable that when the highest dose of emitters was applied there was a significant increase in the mean weight of single plants even in those species of which only a few specimens were left in this experimental variant. Both the total weight of the plant mass and the mean weight of single plants of millet, perennial leguminous, and cruciferae plants were especially sharply increased (Figs. 51 and 52).

In order to demonstrate the difference in relative position and the degree of shift in the contribution of various species to the control community and to the community exposed to the highest concentration of radioactive solution (200 mCi/liter), Fig. 53 shows the relative contributions of six different plant species to the production of the total community biomass in these two variations of the experiment. Wheat and barley yielded in the control a biomass many times as large as that in the experiment, the biomass of pea and vetch was the same in the two

variations, while clover, and particularly millet, produced a considerably greater amount of biomass in the community treated with emitters than in the control. Such a drastic increase in the number and biomass of some plant species in the communities which received the highest concentration of emitters was due to the elimination from these communities of the most prolific species of the first storey and to the vacation in this way of the edaphic space for species which in the control community had an inferior position in the lowest storey.

On the whole the experiments described here on the introduction of radionuclides into the soil and on the preplanting wetting of seeds in radioactive solutions demonstrated that with relatively low concentrations of emitters the total biomass of the annual herbaceous phytocenosis increased substantially, while its qualitative and basic structure did not undergo appreciable changes. At high concentration of emitters, along with a general reduction in the biomass and an impoverishment of the species composition of phytocenoses, marked rearrangements of the storey structure are observed. In some cases, against a background of general stimulation of the community biomass by low concentrations of emitters, growth of some species turns out to

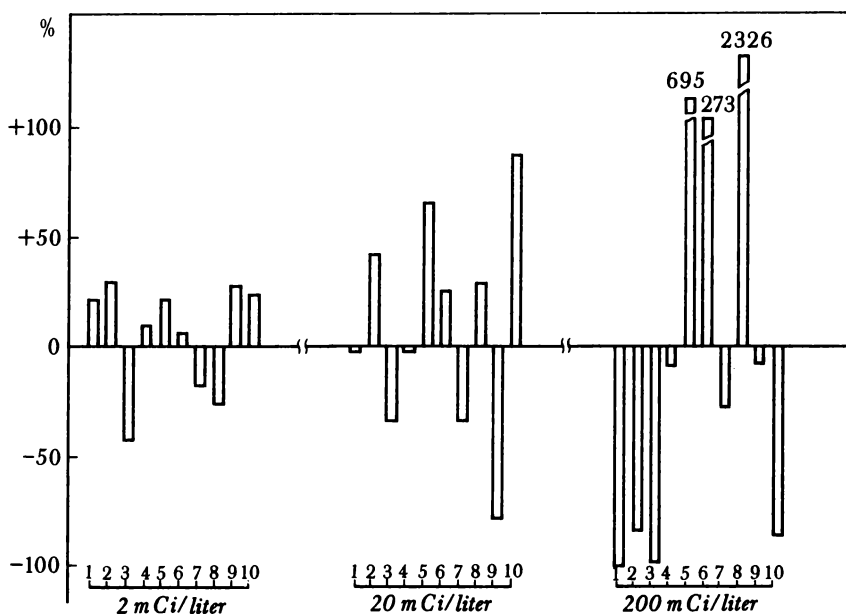


Fig. 51. Percent deviation from control of plant weight per square meter according to species and groups of species in 2.5-month old communities formed under effect of differing doses of emitters: (1) annual cereals; (2) perennial cereals; (3) first group of annual leguminous plants (beans, haricot, soybean); (4) second group of annual leguminous plants (pea, vetch, lentil); (5) perennial leguminous plants; (6) cruciferae; (7) umbellate plants; (8) millet; (9) hemp; (10) buckwheat.

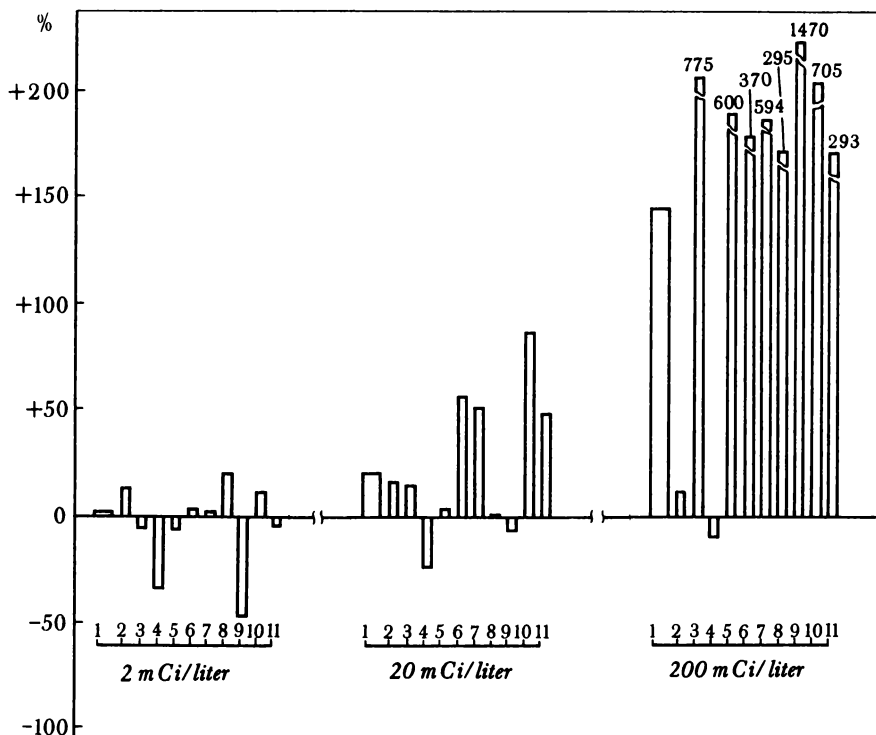


Fig. 52. Percent deviation from control of mean weight of a single plant according to species and groups of species in 2.5-month old communities formed under effect of differing doses of emitters: (1) weight of a single plant averaged for all species; (2) annual cereals; (3) perennial cereals; (4) first group of annual leguminous plants (beans, haricot, soybean); (5) second group of annual leguminous plants (pea, vetch, lentil); (6) perennial leguminous plants; (7) cruciferae; (8) umbellate plants; (9) millet; (10) hemp; (11) buckwheat.

be unexpectedly inhibited, whereas in experimental variants with high concentrations that generally inhibit the biomass growth some species appear to be stimulated. Analysis of these phenomena on the basis of the storey structure of the community demonstrated that such cases are associated with the process of community rearrangement. In an overcrowded community radiostimulation at low doses of the most prolific and rapidly growing species of the first storey results in a slight inhibition of the slowly growing species of the third storey, while a marked impoverishment and radioinhibition of first storey species at high radiation doses results, owing to increased edaphic space, in a sort of stimulation of the development of those species of the third storey that were inhibited in overcrowded communities in the remaining variations of the experiment. Similar phenomena of a "pseudoradiostimulation" caused by disturbances to competitive interrelations of the organisms in an irradiated community, were also noted with regard to the soil bacterial flora composition both with introduction of

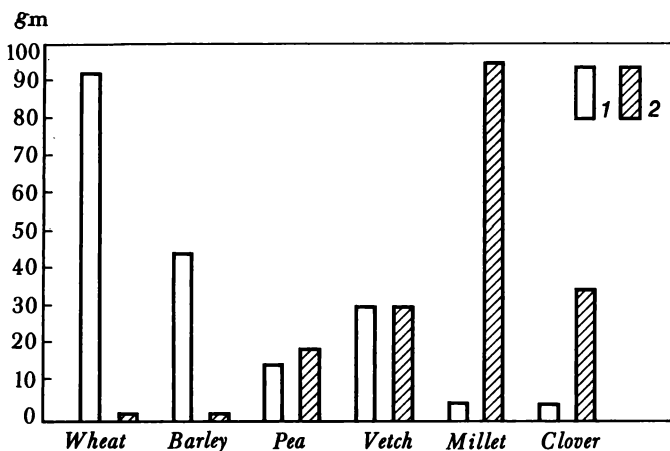


Fig. 53. Contributions of six plant species to phytocenosis biomass (grams per square meter of area) in control (1) and under effect of the highest dose of emitters (200 mCi/liter) (2).

radionuclides into the soil and with an external irradiation of microbopedocenoses with gamma rays (Timofeyev-Resovsky, 1957; Kashkina and Abaturov, 1968; Kashkina, 1969).

In experiments with freshwater periphyton developing in radioactive water contaminated with a nonseparated mixture of uranium fission fragments, considerable shifts in the community structure, including changes in its biomass, were also noted.

Thus, despite considerable differences in the ways of irradiation of communities of different types (terrestrial phytocenoses, soil bacteria, and freshwater periphyton), the responses of these communities to irradiation proved similar. Under the effect of relatively low emitter concentration (radiation doses) the total community biomass increases somewhat, whereas the species composition and the basic structure of the community changes only slightly. At sufficiently high emitter concentrations (radiation doses) profound changes in the composition and structure of the community are evident, attended by distinct changes in the biomass of species that form this community.

CONCLUSIONS

This book considers two closely interrelated aspects of continental radioecology: biogeochemical and radiobiological. The former includes work concerned with the migration and distribution of radionuclides in the soil/solution, soil/plant, and water/hydrobiont biogeoecotic links, while the latter deals with investigations concerning the biological effect of radionuclides on terrestrial and freshwater biogeoecoses.

Experiments conducted with the soil/solution system have demonstrated that the rate and degree of absorption of trace amounts of

^{59}Fe , ^{60}Co , ^{90}Sr , ^{91}Y , ^{137}Cs , and ^{144}Ce from solution in the soil are defined (other things being equal) by the chemical nature of these elements, and are independent of soil properties. This is attributable to the fact that the absorption capacity of the soils studied is sufficiently ample to retain the radionuclide forms capable to be sorbed from a solution. A change in solution pH from 3 to 10 does not affect the soil absorption of ^{60}Co , ^{90}Sr , ^{91}Y , and ^{137}Cs , whereas the absorption of ^{59}Fe and ^{144}Ce declines noticeably. This is due to the change, in an alkaline medium, of iron and cerium to colloidal forms that are poorly sorbed by the soil. The extent of soil absorption of iron and cerium also depends on their concentration in solution.

It was found that the soil sorption of cobalt, yttrium, and cerium radionuclides depends on the content in a solution of stable isotopes of iron and aluminum: the sorption is more reduced the higher the concentration of these elements in the initial solution. The mechanism of this phenomenon may be associated with a process of co-sedimentation and adsorption of radionuclides with the colloid ferrous and aluminum hydroxides.

Experiments with artificial chelating agents have shown that radionuclides in the chelated form are poorly absorbed by the soil, and that the effectiveness of chelating agents with respect to various radionuclides is directly related to the durability of chelate compounds as characterized by their stability constants.

Aqueous extracts from vegetative litter enhance the soil mobility of all the radionuclides under study except ^{137}Cs . The effect of plant extracts is on the one hand attributable to their containing organic substances capable to convert some chemical elements into stable, water-soluble complex compounds, and on the other hand to an increased concentration in these extracts of other cations that can partly displace radionuclides from the soil into solution.

^{59}Fe and ^{137}Cs were nuclides the most firmly retained by all of the soils studied, the rest followed in the sequence: $^{91}\text{Y} > ^{144}\text{Ce} > ^{60}\text{Co} > ^{90}\text{Sr}$. According to their ability to retain radionuclides the soils can be arranged in the sequence: soddy-meadow > chernozem > red soil > soddy podzolic soil. Since these soils differ from each other by their content of humus and their silt fraction in about the same sequence, it was postulated that organic matter and finely dispersed silt particles can reduce the mobility of radionuclides in the soil. Specific experiments with organic substance-free soils have demonstrated that their ability to retain radionuclides is much inferior to that in native soils.

Among different cations used in the experiments as desorbents, "specific replacers" for some radionuclides were distinguished. Aluminum is such a replacer for ^{59}Fe , copper and iron for ^{91}Y , and iron is a replacer for ^{144}Ce . With regard to ^{60}Co and ^{90}Sr , the desorbing effect of cations increases with an increase in their atomic weight and valency. The latter factor suggests the presence in soil of an ion-exchange mechanisms for absorption of these radionuclides.

The strong desorbing effect of "specific replacers" can be explained by the fact that in the soil they form largely the same compounds that

the radionuclides they replace. Therefore, the cations which are "specific replacers" can be regarded as nonisotopic carriers of the respective radionuclides in the soil.

With an increase in soil moisture, the total content of all radionuclides in the liquid phase increases. In soils with the maximal and minimal values of the moisture coefficient differences in content of the dissolved radionuclide forms reach about one order of magnitude for ^{60}Co , and two and three orders of magnitude for ^{144}Ce and ^{137}Cs , respectively, whereas for ^{90}Sr this difference is only three to fourfold. As a result, if the degree of mobility of different radionuclides in the soil is judged on the basis of their total content in the liquid phase, it follows that the ^{90}Sr mobility in moist soil exceeds that of the remaining radionuclides by a factor of from 10 to several hundred. In soil suspensions, however, these differences substantially level off.

The data recorded during the greenhouse experiments generally are consistent with the main results obtained in studies of radionuclide mobility in the soil/solution system. Thus, the higher mobility of ^{59}Fe in the soil/solution system (due to the presence in solution of poorly sorbed forms of iron), and of ^{90}Sr (due to its relatively poor retention by the soil) corresponds to the high mobility of these radionuclides in the soil/plant system. This agreement is manifested by a greater uptake (as compared with other radionuclides) of ^{59}Fe and ^{90}Sr by plants. This agreement between the laboratory and greenhouse experimental results suggests a conclusion that the entry of radionuclides into plants depends largely on the degree of these radionuclides mobility in the soil/solution system. This latter factor is defined in turn by the chemical nature of radionuclides, the form in which they are present in the soil solution, and by the physicochemical properties of the soil. The uptake of radionuclides by plants depends partially on the biological peculiarities of these plants.

Soils with a higher content of humus and a greater silt fraction better retain radionuclides, therefore their uptake by plants from heavily textured soils with a high humus content is lowered. With a rise in soil moisture from the level of withering to that of full water capacity the total accumulation of radionuclides by above-ground parts of plants increases. This increase, however, is due only to a more prolific growth of the plant biomass on more moist soils, and not to a change in the nuclide concentration in plants, which remains practically invariable under all of the moistening regimes studied.

It was found that the entrance of ^{59}Fe , ^{60}Co , and ^{91}Y into plants was considerably increased when these radionuclides were introduced into the soil in the form of chelate compounds incorporating an artificial chelating agent EDTA. Here also a good agreement was noted between the results of the laboratory and greenhouse experiments. Under laboratory conditions the chelating agent, while diminishing the sorption of radionuclides in the soil, enhanced their mobility in the soil/solution system; under conditions of greenhouse experiments this mechanism led to an increase in the radionuclide uptake by plants. EDTA did not perceptibly affect the mobility of strontium, cesium, and cerium

radionuclides, since the chelate compounds of these elements in the soil are insufficiently resistant to chemical and biological destruction.

With an increase in excessive concentrations of a chelating agent in a medium (soil, nutrient solution) the accumulation of cobalt by plants diminishes. This phenomenon is believed to be associated with the formation, in the presence of an excessive amount of a chelating agent in the medium, of compounds of this chelating agent with other cations which compete with Co/EDTA compound for being taken up by plants. This peculiarity should be taken into account when, for example, complex compounds are used as microfertilizers, since an excess of a chelating agent in relation to a microelement in the initial preparation may substantially impair the effectiveness of a microfertilizer.

It is to be emphasized that the results of the series of investigations conducted on plots of natural biogeocenoses were on the whole in fairly good agreement with the results of the laboratory and greenhouse experiments. ^{59}Fe and ^{144}Ce , being more mobile in the soil/solution system, under natural conditions also exhibited a greater capacity for migration than did the other radionuclides. As indicated above, the higher ability of iron and cerium to migrate in the soil is based on the capacity of these elements to change, in the pH range characteristic of many soils, to colloidal forms that are poorly sorbed by the soil.

Under natural conditions ^{90}Sr proved to be less mobile than might be expected from the results of the laboratory and greenhouse experiments. One of the causes of such a reduction in ^{90}Sr mobility may be its conversion with time into the nonexchangeable state as a result of an isomorphic replacement of calcium by strontium in soil minerals, a fact that escapes the attention of investigators in laboratory experiments of short duration. Another cause may be seasonal fluctuations of natural soil moisture, which appear to facilitate leveling of the rates of migration of different radionuclides with the soil solution. However, under conditions of excessive moistening, and owing to peculiarities of the Far North vegetation, the migration capacity of ^{90}Sr is greater than that of ^{137}Cs , resulting in the enrichment in ^{90}Sr of accumulative areas in the tundra landscape.

Investigations of the behavior of radionuclides in the water/hydro-biont system confirmed that the migration capacity of radionuclides in this system (as in the soil/plant system) is defined by their chemical nature, biological peculiarities of organisms, and physicochemical properties of the medium.

When the radionuclides accumulated by plants are derived from the aqueous medium the accumulation coefficients are higher than when the nuclides are derived from the soil, since the soil, being a potent natural sorbent, firmly retains trace amounts of chemical elements. Thus, for instance, the accumulation coefficients for the majority of the radionuclides under study attain, in freshwater plants, the order of ten thousand, whereas in terrestrial plants these coefficients are less than unity. Thus, the soil has a function of a powerful barrier along the path of radionuclide migration in a biogeocenosis.

Based on data for ^{90}Sr and ^{137}Cs , it is shown that the values of the accumulation coefficients established for hydrobionts experimentally and under laboratory conditions may in a number of cases be lower than those found in a natural waterbody. This can be explained by the inability to reproduce in a simplified laboratory model the complex of conditions arising in a natural waterbody. Moreover, during the long-term interaction of plants with water contaminated with radionuclides, a certain proportion of the absorbed radionuclides changes with time in the plant tissues to nonexchangeable or slightly exchangeable forms which results in a gradual increase of accumulation coefficients. Since this is a relatively slow process it cannot be detected in laboratory experiments of short duration.

An analysis of data from the literature and the results of investigations conducted in our laboratory permit the inference that within the range of microconcentrations (all other things being equal) the concentration of radionuclides in hydrobionts is directly proportional to their concentration in water and, as a consequence, their accumulation coefficients remain invariable. Within the range of macroconcentrations in water the accumulation coefficients diminish; they also diminish when the water concentration of some nonisotopic carriers (i.e., stable elements with similar chemical properties) of the respective radionuclides increases.

A discrimination of ^{90}Sr with respect to its chemical analog, calcium, and of ^{137}Cs with respect to potassium was noted when these elements were accumulated by hydrobionts. The value of the discrimination coefficient depends on specific biological features of hydrobionts and does not depend, in a wide range of values, on the macrocomponent concentration in water.

The excretion of chemical elements from fish with the roe during spawning is considered a peculiar pathway of the biological migration of radionuclides in a waterbody and a means of fish self-clearance of the incorporated nuclides. It has been found that ^{137}Cs and potassium are excreted from freshwater fish (tench, pike) in a comparatively larger amount than ^{90}Sr , calcium, and magnesium.

The accumulation of radionuclides by hydrobionts depends on the ecological characteristics of a waterbody. It was demonstrated that the coefficients of accumulation of ^{90}Sr and ^{137}Cs for plants growing in a dystrophic waterbody were markedly higher than for plants in an oligomesotrophic waterbody. One of the main causes of an increase in the accumulation coefficients of radionuclides in a dystrophic lake is a lower water content of macroelements, i.e., calcium, magnesium, potassium, and sodium, because of hydrobiont overcrowding.

Being a chemical analog of calcium, ^{90}Sr enters the same links of the biogeochemical migration as does calcium. In particular, it is involved with calcium in carbonate formation processes, changing in this way from the water-dissolved state to a slightly soluble sediment deposited on the waterbody bottom. The investigations described indicate that ^{90}Sr co-precipitates with calcium carbonates with a discrimination coefficient of about 0.3. The value of this coefficient does not depend

either on the specific biological features of the carbonate-forming plants or on the calcium concentration in water. The process of carbonate formation, which in the final analysis is a result of the vital activity of an organism, presumably proceeds outside the plant organism, at the plant/aqueous solution interface, whereas the carbonate sediment is formed as a result of carbon dioxide absorption by plants from bicarbonates.

The work described shows that, along with living plants, an important role in the fate of radionuclides in a waterbody is played by dying plants and detritus. The coefficients of accumulation of ^{90}Sr and ^{137}Cs in dead remains were of the same order of magnitude, while those of ^{106}Ru and ^{144}Ce were severalfold higher than in living plants. Moreover, ruthenium and cerium are retained by organic matter of dead plants more firmly than by the tissues of living plants. Of all the radionuclides studied only ^{137}Cs was retained by live tissues more firmly than by dead organic remains. Owing to a firm retention by the mass of dying plants, ruthenium and cerium nuclides must pass with the detritus to the bottom deposits of a waterbody, while ^{90}Sr and ^{137}Cs , which are bound less firmly, may easily return to solution and once again enter the biological links of migration.

It should be stressed that the methodologic approach used for the present investigation and aimed at a comparative study of the migration capacity of radionuclides in the crucial biogeocenotic links of soil/plant and water/hydrobiont by means of experimental division of these links into simpler systems proved to be quite justified. As the results of our experiments showed, this approach enabled us to provide not only the general comparative phenomenologic characteristics of the behavior of the radionuclides studied in systems of varying complexity, but also to reveal in individual cases the particular physicochemical mechanisms controlling the mobility of radionuclides in these systems.

Our research has confirmed that the problems of migration and distribution of radionuclides between the components of biogeocenoses are closely related to another highly important goal of radioecology—studies of biological effects of radioactive substances and their radiation.

We admit that, although the data presented concerning the comparative radiosensitivity of various species of living organisms as dependent upon a number of attendant factors give an idea of the most vulnerable links of natural biogeocenoses in case of their irradiation, these data are nevertheless insufficient for the scientifically substantiated prediction of possible consequences of the radioactive contamination of the particular biogeocenoses. In this connection it is imperative to include in radioecological investigations as many new biological objects as possible, studying their radiosensitivity in relation to their developmental stages, physiological state, modifications of environmental factors, and irradiation regime. Since the main forms of existence of living organisms in a natural habitat are population and biocenosis, it is necessary, along with studies of the radiosensitivity at the organismic level, to promote research on the radiosensitivity of individual

species at the levels of population and biocenosis. Suitable radiosensitivity criteria in such investigations may be the population size, age composition, reproduction and death rate of individual species, biomass, rearrangements in the composition and structure of communities, and other parameters used for descriptions of biological systems at the levels of population and biocenosis.

Under natural conditions the effects of ionizing radiation on organisms are combined with the influence of other ecological factors. In the cases when the effect of ionizing radiation manifests itself against a background of adverse environmental factors the radiosensitivity of organisms generally increases. In turn, any radiation injury reduces the resistance of organisms to adverse environmental factors. It is well known that organisms weakened by irradiation decrease their resistance to various infections, parasites, pest insects, predators, etc. (Ohba and Simak, 1961; Woodwell and Miller, 1963; Brower, 1965; Woodwell, 1965). Moreover, as indicated in this book, under the effects of radiation and because of secondary changes in the composition and structure of overcrowded communities, there can occur phenomena of pseudoradioinhibition of individual species at stimulating radiation doses, and phenomena of pseudoradiostimulation at very high radiation doses. In radioecological studies increasing attention should be given to the combined effect of ionizing radiation and other environmental factors, and to the quantitative appraisal of individual factors involved in radiation injuries to organisms.

The ultimate goal of radioecology is the development of a general theory of the migration and biological effects of radioactive substances in different biogeocenoses. This theory should be based on modern concepts of the structure and functioning of biological macrosystems characterized by a great number of interacting factors. We are facing quite a number of difficulties on the road to solution of this complicated problem, but we believe these may be successfully overcome in future with the aid of analog computers modelling the functioning of biogeocenoses.

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A. A. Neyfakh and M. Ya. Timofeeva

**MOLECULAR BIOLOGY OF DEVELOPMENT
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The authors head research groups on molecular biology of development at two Moscow institutes of the USSR Academy of Sciences: the Institute of Developmental Biology and the Institute of Molecular Biology.

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Nikolai Vasilyevich Kulikov, D.Sc. (Biology), is Head of the Laboratory of Radioecology of the Institute of Plant and Animal Ecology, the Urals Scientific Center, Academy of Sciences of the USSR. His field of research is the migration, accumulation, and biological effects of radioactive substances in terrestrial and freshwater ecosystems. He has published more than 150 works, including two monographs (jointly with I. V. Molchanova), "Radionuclides in the Soil/Plant System" and "Continental Radioecology. Soil and Freshwater Ecosystems."

Inna Vladimirovna Molchanova, Cand.Sc. (Biology), is a Senior Researcher of the Laboratory of Radioecology of the Institute of Plant and Animal Ecology, the Urals Scientific Center, Academy of Sciences of the USSR. She has conducted extensive research in the radioecology of the soil-plant cover and investigated the migration of radionuclides in the soil/plant system. She is the author of more than 50 scientific works, including two monographs (written jointly with N. V. Kulikov), mentioned above.