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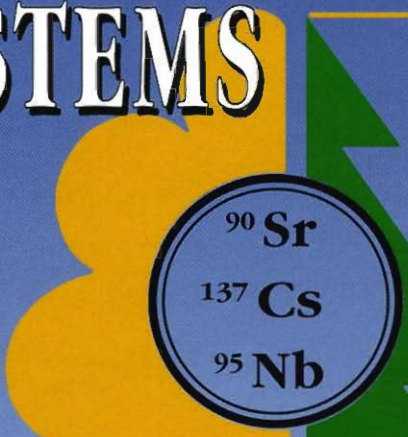

NAUKA

A. I. Shcheglov,
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BIOGEOCHEMICAL MIGRATION OF TECHNOGENIC RADIONUCLIDES IN FOREST ECOSYSTEMS

by the materials of a multiyear study in the areas severely contaminated due to the Chernobyl Accident

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The monograph is dedicated to the fate of Chernobyl-born radionuclides (^{137}Cs, ^{90}Sr, $^{238-240}Pu$, ^{106}Ru, ^{144}Ce, etc.) in the central part of the East-European Plain. It is based primarily on the original materials obtained in the course of a 12-year study in the temperate forest environments contaminated due to the Chernobyl accident. The monitoring network from which the data have been obtained covers a distance of 3 to 550 km from the Chernobyl Power Plant and includes sites which received deposition of radionuclides ranging from 200 kBq m^{-2} to 30 MBq m^{-2}. The research was designed at the outset to examine the behaviour and fate of radionuclides in all basic ecosystem components (soils, soil solutions, herbaceous and arboreal vegetation, fungi, etc.) at different levels (community, ecosystem, landscape, and climatic zone). The monograph considers in detail the role of forests in the initial distribution of the radioactive fallout and processes of the long-term radionuclide redistribution among forest soil and biota. The authors estimated the contribution of individual ecosystem components (including mycobiota) to the radionuclide transport in biogeocenoses and calculated the half-stay and quasi-equilibrium periods for the investigated radionuclides in various tree tissues, herbaceous plants, soil layers, and ecosystems.

The unique field data presented and discussed in the monograph will be interesting for anyone engaged in modelling and forecasting of the radiological impact of radionuclide contamination on temperate forests. The abundance of factual material and quantitative parameter values makes this book an indispensable reference source on radionuclide migration and fate in forest environments.

The book includes 87 tables, 97 figures, and reference list of 357 sources.

The monograph includes materials obtained within the framework of the International Atomic Energy Agency Co-ordinated Research Project (CRP No.10077, 1998-2001) and under support of the Russian Foundation for Basic Research (Projects 00-04-48024 and 01-04-48354)

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FOREWORD

As early as the 19th century, the founder of biogeochemistry V.I. Vernadskii wrote that the human impact on the evolution of the biosphere was becoming a major factor, almost of geological scale. However, the rate and extent of the environmental despoliation which occurred in the 20th century would be hardly imaginable even for him. The present state of the biosphere, with its shrinking species diversity, water pollution and soil degradation, not to mention the rising level of carbon dioxide in the global atmosphere, appears to indicate the devolution of Nature rather than her evolution.

The latter half of the 20th century was notorious for global radioactive pollution due to nuclear weapons testing and numerous accidents in nuclear installations across the world. The Chernobyl accident – the largest and most infamous in the history of nuclear energy – occurred in eastern Ukraine on April 26, 1986, and resulted in environmental pollution on a global scale. Vast areas of Ukraine, the Russian Federation, the Republic of Belarus, and many eastern and western European countries were exposed to severe radioactive fallout. A large proportion of the territories contaminated by this radioactive fallout was covered with forests. Scientists and decision – makers were unexpectedly challenged by a set of complex, interdependent and poorly investigated problems, such as the long-term forecasting of the radioactive contamination of these forests, estimation of the corresponding dose burden on various population groups, planning and development of large-scale protective and mitigation measures, control of the forestry activities in the territories exposed to the fallout and, ultimately, reclamation of the contaminated forestlands.

Solution of all these problems requires a detailed knowledge of how forests may affect the initial fallout distribution over the contaminated territory as well as the subsequent redistribution of radionuclides among the components of the forest ecosystem as time passes. The direct and indirect impact of radiation on the forest ecosystem also depends on radionuclide pathways and migration over a time scale of decades (wash out, leaching, root uptake, secondary accumulation, etc.) . Thus, an understanding of the long-term radioactive contamination of forests can only be obtained using an integrated biogeochemical approach, which makes it possible to interpret and understand radionuclide distributions and fluxes within the systems of natural biological and geological cycles. This is precisely the scope of this book.

This monograph is based primarily on information obtained by the Radioecology Laboratory of Moscow State University, which has been engaged in the study of radionuclide behaviour in the environment since the early 1950s.

Summarised herein are unique experimental data obtained in temperate forest ecosystems contaminated as a result of the Chernobyl accident. The monitoring network from which the data have been obtained covers a distance of 3 to 550 km from the Chernobyl Power Plant and includes sites which received deposition of radionuclides ranging from 200 kBq m^{-2} to 30 MBq m^{-2}. The key monitoring sites, established in 1986–1987, represent various climatic zones (from southern taiga to forest-steppe), a wide spectrum of soil types (from soddy-podzolic and meadow-bog to chernozems), and various phytocenoses (coniferous, broad leaved, mixed forests and meadow communities). The research was designed at the outset to examine the behaviour and fate of radionuclides in all basic ecosystem components (soils, soil solutions, herbaceous and arboreal vegetation, fungi, etc.) and at different spatial levels (community, ecosystem, landscape, and zonal), so that radionuclide fluxes could be determined at appropriate scales.

The monograph considers the role of forest ecosystems in the initial distribution of radioactive fallout over Chernobyl contaminated territories and their contribution to the long-term radionuclide redistribution among soils and biota. Grouping of the ecosystem components according to their accumulative capacity for radionuclides serves as a basis to evaluate the efficiency of biogeochemical barriers to retard and control radionuclide migration in various landscapes.

The authors have estimated the contribution of individual ecosystem components (including mycobiota) to radionuclide cycling in biogeocenoses and have calculated residence half times and quasi-equilibrium periods of Chernobyl-derived radionuclides in various tree tissues, herbaceous plants, soil layers and ecosystems. The data presented herein make it possible to identify so-called critical landscapes in which radionuclide migration is greatest. The specific behaviour of the technogenic radionuclides investigated suggests that their biogeochemical migration under natural conditions differs from that of natural isotopic and nonisotopic carriers.

The importance of the monograph goes far beyond its formal scope. On the one hand, it contributes substantially to our knowledge of the biogeochemistry of technogenic radionuclides as a whole and, on the other hand, it expands our current knowledge of the rates and pathways of migration of other trace elements in forest ecosystems. Furthermore, the methodology developed and tested in this work is applicable to biogeochemical studies of any technogenic pollutant or nutrient in forest ecosystems. The unique field data presented and discussed within this monograph are extremely valuable for everyone engaged in modelling and forecasting of the radiological impact of radionuclide contamination of temperate forests. The abundance of factual material and quantitative parameter values makes this an indispensable reference source on radionuclide migration and fate in forest environments.

Prof. F. Tikhomirov

INTRODUCTION

The biogeochemical cycles of natural radioactive elements have been formed through the entire geological history of the Earth. Industrial activities, such as mining, processing, production, and utilisation of radioactive materials, lead to a large-scale redistribution of natural radionuclides and global environmental pollution by technogenic radionuclides. The main sources of technogenic radionuclides are nuclear weapon tests, improper disposal of nuclear wastes, and various accidents at nuclear installations. Technogenic impact on the long-term natural processes may provoke a deep ecological crisis on a global scale. This is especially true in the context of the Chernobyl accident (1986) that has challenged urgent development of special recommendations and technologies to keep industrial activities in the contaminated territories of Ukraine, Belorussia, and Russia.

Forest ecosystems are known to be the so-called "critical" environments that may accumulate and preserve technogenic radionuclides for a long time and provide the maximal dose burden on living beings, including humans due to both intensive external irradiation of the population and fast radionuclide migration through trophic chains [8, 232]. Thus, forests are of high ecological importance not only as an essential component of the biosphere sustaining its ecological equilibrium, but also as one of the factors directly affecting the radioecological situation in contaminated territories. Radionuclide behaviour in forest ecosystems is very specific and differs from that in natural meadow ecosystems and agroecosystems.

Biogeochemical studies of technogenic radionuclides in forest environments of different natural zones are of top priority for modern radioecology. Important data on the radionuclide fate in forest environments were obtained during special studies in the zones of the Kyshtym and some other nuclear accidents, and in a wide range of field and laboratory experiments. The results of these studies were considered in detail in well-known monographs by R.M. Alexakhin, Yu.A. Egorov, R.T. Karaban', D.A. Krivolutskii, N.V. Kulikov, I.V. Molchanova, M.A. Naryshkin, F.A. Tikhomirov, E.B. Tyuryukanova, etc. However, the global scale of the Chernobyl accident and the specific features of the Chernobyl-derived fallout make the biogeochemical studies in the forest environments contaminated due to the Chernobyl accident of top priority to solve numerous theoretical and practical problems.

In addition, a detailed knowledge on the behaviour of long-lived radionuclides in forest landscapes is crucial for many aspects of forestry, agriculture, and medicine. It is required for estimating the dose burden on various living organisms, including human beings, in contaminated areas.

The objective of this monograph is to apply theoretical and experimental principles of biogeochemistry to radioecological studies of forest ecosystems of the central part of the East European Plain. The research is focused on the following items:

- to show the role of forest ecosystems in the processes of the primary and secondary distribution (redistribution) of radionuclides over the contaminated territory;
- to reveal the peculiarities and dynamics of the radionuclides' redistribution among various components of forest ecosystems in particular climatic zones;
- to determine the leading factors of radionuclide migration in the soil-plant system and make quantitative evaluation of the relevant processes;
- to estimate basic parameters of radionuclide fluxes and evaluate their individual contribution to the biogeochemical radionuclide cycle in forest ecosystems;
- to determine physicochemical forms of radionuclides in the soils and characterise the dynamics of radionuclide availability for higher plants;
- to develop a conceptual model of the biological cycle of radionuclides in various forest ecosystems.

The work is based on the data of long-term monitoring and experimental studies carried out within the so-called exclusion zone of the Chernobyl Nuclear Power Plant (ChNPP)* and more distant contaminated territories of Ukraine and Russia. Being focused primarily on the original studies, the authors also analyse the published data on technogenic radionuclides released due to the weapon tests and the accidents in the South Ural (Kyshtym) and Chernobyl.

This book is dedicated to the mechanisms, dynamics, and parameters of the radionuclide migration rather than its mathematical modelling. The issues directly related to modelling of the radionuclide fate in forest environments are out of the scope, since these are considered in detail in a range of other publications [352, 350]**. We believe that mathematical modelling is an indispensable tool to study, estimate, and predict radionuclide migration as far as it is based on clear understanding of the key mechanisms and processes governing the radionuclide migration in real ecosystems. In this context, the book should be interesting and helpful for everybody engaged in modelling of technogenic radionuclides in the environment.

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* The exclusion zone is a 30 km wide, protected, unpopulated territory around the ChNPP established in 1986 to exclude any human activities in the severely contaminated area.

** See also: Mamikhin *et al.*, Dynamics of 137-Cs in the forests of the 30 km zone around the Chernobyl nuclear power plant. *Sci. Total Environ.* 193, 169–177 (1997); Avila, R. et al., Modelling of radionuclide migration in forest ecosystems. *SSI Report* 98:07 ISSN 0282–4434 (1998); *Contaminated Forests*, Eds. I. Linkov and W. Schell, Kluwer Academic Publishers, Netherlands (1999); *Final Report on the BIOMASS Forest Working Group Activities 1998–2000*, IAEA, Vienna, (2000), ISSN 1011-4289.

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Please, do not hesitate to share your any remark and commentary on the book with the authors:

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1. METHODOLOGICAL APPROACHES TO THE STUDY AND RADIOECOLOGICAL CHARACTERISTICS OF THE INVESTIGATED TERRITORY

1.1. METHODOLOGICAL APPROACHES

Our methodology is based on the landscape-biogeochemical approach that combines (i) the studies on radionuclide migration and accumulation in the geochemically linked (conjugated) landscapes (large scale), and (ii) the ecosystem studies at the level of mesotopography and microtopography (medium scale). This is complemented with the studies of the particular processes in the soil-plant, soil solids-liquids, forest litter-mineral soil and other systems (micro-scale). Such integral, multisided, and multilevel approach makes it possible (1) to characterise the radionuclide behaviour under natural conditions; (2) to forecast the peculiarities of the radionuclide dynamics and distribution among the ecosystem components, elementary landscapes, and systems of geochemically conjugated landscapes; and (3) to trace in detail the radionuclide pathways in a landscape as a whole.

A network of experimental key sites was established to cover different climatic zones and the most representative ecosystems in the contaminated territory. In some regions, the network was specially designed to constitute a complete geochemical profile, i.e. a series of the key sites were arranged in such a way that the whole system represented a chain of geochemical catenas most typical of the investigated region (*Fig.* 1) [277]. The biogeocenoses* (BGC) chosen to represent the elementary landscapes within the geochemical profiles exhibited the most characteristic features of the vegetative and soil cover, as well as the composition and spatial distribution of the initial fallout in the investigated area.

A detailed description of a monitoring network including the distance of the key sites from the accidental unit of ChNPP, the features of landscape, the soil and vegetative cover, deposition, and composition and properties of the initial fallout is presented in Table 1 and the maps enclosed.

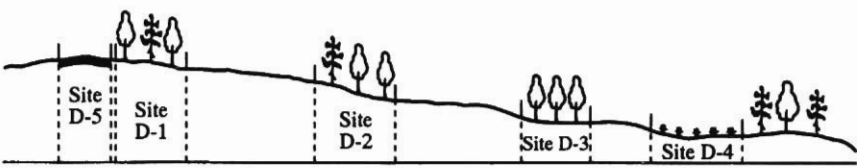

Fig. 1. Principal arrangement of the key sites down a geochemical profile

* The term "biogeocenosis" in the context of this study is much similar to the term "ecosystem", but lays an emphasis on the soil as an essential component of the ecosystem.

Table 1. Basic characteristics of the investigated key sites

KEY SITE: (Direction and distance from the Chernobyl Nuclear Power Plant)	Soil type*	Type of landscape	Type of vegetation (phytocenosis**)	Deposition, MBq/m^2
		Tula region (Russia)		
Pl-1 (550 km to NE)	Chernozem (podzolized), clay-loamy	Eluvial	Mixed, broad-leaved/ small leaved forest	0.37
Pl-2 (550 km to NE)	Chernozem (podzolized), clay-loamy	Eluvial	Pine plantations	0.38
		Bryansk region (Russia)		
Zl-1 (165 km to NE)	Podzolic, illuvial-iron, sandy soil	Transit	Pine forest with herbaceous cover	1.41
Km-1 (200 km to NEE)	Podzolic, illuvial-iron, sandy soil	Eluvial	Pine plantation	0.12
Kl-1 (200 km to NEE)	Podzolic, illuvial-iron, sandy soil	Transit-accumulative	Mixed, pine/small-leaved forest	0.64
Kl-2 (200 km to NE)	Peat podzolic, surface-gleyic soil	Accumulative	Mixed, pine/broad-leaved forest	0.58
		Bryansk region (Russia)		
Kr-1 (220 km to NNE)	Soddy-podzolic soil	Eluvial	Mixed, pine/broad-leaved forest	2.26
Nz-1 (175 km to NE)	Podzolic, illuvial-iron, sandy soil	Transit	Pine plantation	1.65

		Kaluga region (Russia)		
Nz-2 (175 km to NE)	Podzolic, weakly stratified, sandy soil	Eluvial	Pine forest with mixed herbaceous/moss cover	0.6
Kh-1 (440 km to NEE)	Podzolic, illuvial-iron, sandy soil	Eluvial	Pine forest with herbaceous cover	0.41
Kh-2 (440 km to NEE)	Soddy-podzolic, sandy soil	Eluvial	Mixed, pine/broad-leaved forest	0.41
Kh-3 (440 km to NEE)	Alluvial, boggy, clay-peat soil	Accumulative	Sedge bog on a floodplain	0.44
Kh-4 (440 km to NEE)	Alluvial, boggy, peat-gley soil	Transit	Alder forest with herbaceous cover	0.45
E-1 (430 km to NEE)	Podzolic, illuvial-humus-iron, sandy soil	Transit	Mixed, pine/broad-leaved forest	0.42
E-2 (430 km to NEE)	Podzolic, gleyic, sandy soil	Transit	Pine plantations	0.41
E-3 (430 km to NEE)	Podzolic, illuvial-humus-iron, sandy soil	Transit	Spruce forest with moss cover	0.41
		Kiev region, Ukraine (30 km exclusion zone of ChNPP)		
K-2 (6.5 km to SE)	Secondary-podzolic, sandy soil	Transit-accumulative	Pine forest with mixed grass/moss cover	2.90
K-3 (6 km to SE)	Podzolic, iron-illuvial, sandy soil	Transit-accumulative	Pine forest with mixed grass/moss cover	3.23
K-4 (5.8 km to SE)	Alluvial-bog, clay-humus, gley soil on floodplain deposition	Accumulative	Floodplain meadow and grass/sedge valley bog	2.43
K-5 (5,5 km to SE)	Soddy-podzolic, old-tilled, sandy soil	Eluvial	Old-tilled fallow with mixed grass/cereal cover	1.48

Table 1. (continuation)

KEY SITE: (Direction and distance from the Chernobyl Nuclear Power Plant)	Soil type*	Type of landscape	Type of vegetation (phytocenosis**)	Deposition, MBq/m^2
K-6 (3.5 km to S)	Secondary-podzolic, sandy soil	Eluvial	Pine plantations	6.90
K-7 (3 km to S)	Bog, humus-gley soil	Accumulative	Valley grass/sedge bog	6.02
Ch-1 (5.2 km to SW)	Podzolic, gleyic, sandy soil	Transit-accumulative	Pine forest with mixed grass/moss cover	3.91
Ch-2 (5.9 km to SW)	Secondary-podzolic, sandy soil	Transit	Pine plantations	3.42
Ch-3 (6.5 km to SW)	Secondary-podzolic, sandy soil	Eluvial	Pine plantations	3.67
Ch-4 (4.8 km to SW)	Soddy-podzolic, old-tilled, sandy soil	Transit-accumulative	Old-tilled fallow with mixed grass/cereal cover	7.20
	Kaluga region (Russia)			
L-1 (450 km to NEE)	Soddy-podzolic, weakly stratified, sandy soil	Eluvial	Mixed, pine/small-leaved forest	0.75
L-2 (450 km to NEE)	Bog, sphagnum-peat, gleyic soil	Accumulative	Open pine forest on sphagnum peat	0.43
L-3 (450 km to NEE)	Soddy-weak-podzolic, sandy soil	Eluvial	Pine plantation	0.76
	Kiev region, Ukraine (30 km exclusion zone of ChNPP)			
D-1 (28.5 km to S)	Podzolic, iron-illuvial, sandy soil	Eluvial	Mixed, broad-leaved-pine forest	0.24

D-2 (27 km to S)	Podzolic, gleyic, sandy soil	Transit-accumulative	Mixed, broad-leaved-pine forest	0.24
D-3 (26 km to S)	Bog, valley peat-gley, soil	Accumulative	Alder forest with herbaceous cover	0.24
D-4 (25.5 km to S)	Meadow-bog, peat-gley soil	Accumulative	Valley bog with herbaceous (grass-sedge) cover	0.21
D-5 (29 km to S)	Soddy-podzolic, old-tilled, sandy soil	Eluvial	Old-tilled fallow with mixed grass/degenerated cereal cover	0.13
K-1 (7 km to SE)	Secondary podzolic, sandy soil	Eluvial	Pine forest with mixed grass/moss cover	2.54
Sh-1 (6 km to W)	Weak-podzolic, weakly stratified, sandy soil	Eluvial	Mixed, broad-leaved-pine forest	44.73
Sh-2 (5.2 km to NW)	Secondary-podzolic, sandy soil	Eluvial	Pine plantations	5.05
Sh-3 (5 km to NW)	Bog, humus-clay, gley soil	Accumulative	Valley, sedge bog	6.49
Sh-4 (5 km to NW)	Soddy-podzolic, old-tilled, sandy soil	Transit-accumulative	Old-tilled fallow with mixed grass/cereal cover	4.90
Sh-5 (9 km to W)	Soddy-weak-podzolic, weakly-stratified, sandy soil	Eluvial	Pine forest with mixed grass/moss cover	5.09
C-1 (3 km to NE)	Soddy-gley	Accumulative	Mixed grass/cereal cover on floodplain	23.52
U-1 (6.5 km to N)	Sod, sandy, superficial peat soil	Transit-accumulative	Mixed, broad-leaved-pine forest	10.04
U-2 (6.5 km to N)	Soddy-podzolic, sandy soil	Eluvial	Pine plantations	14.85

* Soil nomenclature by [138]

** Vegetation nomenclature by [236]

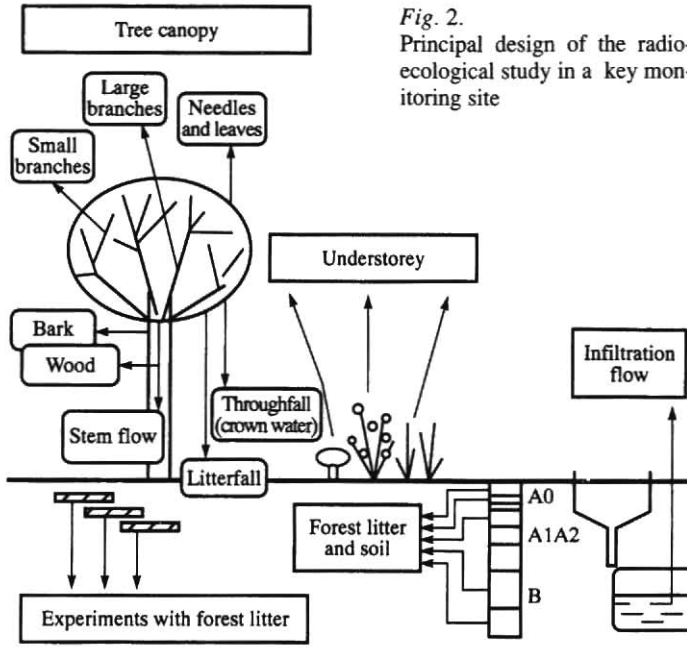

Fig. 2.
Principal design of the radioecological study in a key monitoring site

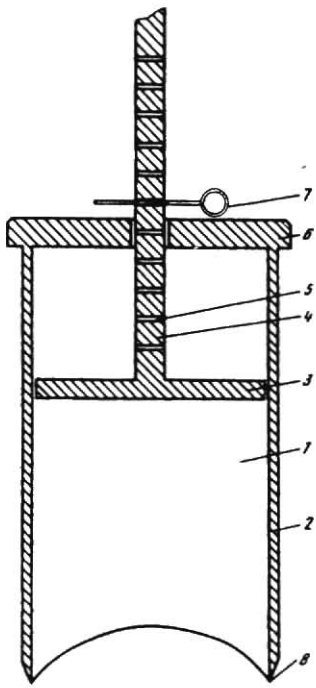

Fig. 3. Longitudinal section of the soil sampler (scheme):
1 – sampling cylinder; 2 – cylinder side; 3 – pushing piston; 4 – retractable rod; 5 – section indexing hole; 6 – sampler lid; 7 – stopping pin; 8 – cutting edge

A comprehensive monitoring scheme was designed at each key site to study, in the maximum detail, the following processes affecting the radionuclide migration: (i) downward transport to the soil surface with litterfall and throughfall; (ii) redistribution of radionuclides in the vertical soil profile via the mechanisms of mechanical replacement, diffusion, and infiltration; and (iii) lateral migration and large-scale redistribution in a system of geochemically linked elementary landscapes (*Fig.* 2). In addition, field experiments were carried out at some key sites to estimate different parameters of biological cycles, and laboratory experiments were conducted to study physicochemical forms of radionuclides in the liquid and solid soil phases.

Sampling. Specific character of radioecological studies requires special methods of sampling and preparation. In our case, soil

samples were taken using sequential, layer-by-layer sampling of the forest litter and mineral soil layers. Three sublayers of the forest litter were collected accurately from a known area (20 × 20 sq. cm) using a special steel frame. Then a 20-cm high soil column was taken out from the litter-free soil and separated into the sections of 1, 2, 5, etc. centimetres using a special sampling device (*Fig.* 3).

The structural components of the ecosystem were sampled as presented in *Fig.* 2 using standard methods [17].

The total phytomass of the arboreal vegetation was calculated from the experimental data and the known quantitative relations between different structural components (organs) of the tree species [54, 58, 64, 167, 169, 199, 200, 220, 281, 282].

To determine the fractional composition of the phytomass of tree species, an empirical dependence between the phytomass of individual fractions (organs) and the known indices, commonly used in forest taxation, were applied to the conditions of the investigated key sites. The equations of direct or inverse regression were used as approximation models. The variables of the regression were the stand height (h), the average stem diameter at the height of 1.3 m (d), the product of the squared stem diameter to the stand height (d^2h), and/or the stand age. The basic, region-adjusted equations for calculation of the tree phytomass are presented in Table 2.

Since the radioactive contamination of the particular components of tree stands is in close correlation with their adsorption area and position in the tree crown, two additional phytomass fractions were introduced: the branches were divided into the fractions of small and large branches, and the bark was divided into the fractions of "external bark" (external dead cork exposed to the initial fallout) and "internal bark" (alive, inner part of the bark and phloem) (Table 3).

The downward radionuclide flux was studied by standard methods described in [17]. This part of the research included (1) regular sampling of the litterfall, and estimation of the rate of its decomposition; (2) sampling of the throughfall and stem flow; and (3) long-term lysimetric studies.

Table 2. Regression equations describing a dependence between the tree components (%) and basic indices of the stand valuation (age, diameter, and heights)

Fraction of the phytomass, Y	Argument, X	Regression	r**	R^2	σ^2	m
		Pine				
Wood	Age	$Y = 38.86 \cdot X^{0.14}$	0.84	69.78	0.38	0.06
Bark	d	$Y = 18.45 \cdot X^{-0.40}$	0.75	53.01	0.31	0.01
Total branches	Age	$Y = 71.52 \cdot X^{-0.60}$	–0.72	51.79	6.08	0.38
Large branches	% of branches	$Y = 1/(9.88*10^{-3} + + 2.48 \cdot 10^{-4} \cdot X)$	0.79	62.03	0.001	0.001
Needles	Age	$Y = 151.41 \cdot X^{-0.98}$	–0.85	72.59	22.03	0.39

Table 2. (continuation)

Fraction of the phytomass, Y	Argument, X	Regression	r**	R^2	σ^2	m
Needles > > 1-y old	Age	$Y = 1.50 \cdot X^{-0.07}$	–0.72	51.70	1.23	0.19
Roots	Age	$Y = 10.77 + 0.06 \cdot X$	0.68	45.89	0.55	0.42
		Spruce				
Stem	h	$Y = 35.59 \cdot X^{0.21}$	0.88	77.46	0.51	0.09
Wood*	Age	$Y = 20.17 \cdot X^{0.28}$	0.76	58.36	1.59	0.16
Total branches	Age	$Y = 76.17 \cdot X^{-0.58}$	–0.66	44.04	5.20	0.47
Small branches	Age	$Y = 603.65 \cdot X^{-0.64}$	–0.91	83.55	1.84	0.17
Needles	Age	$Y = 413.64 \cdot X^{-1.04}$	–0.87	76.14	9.76	0.35
Roots	Age	$Y = 32.23 \cdot X^{-0.14}$	–0.68	47.85	0.55	0.15
Oak						
Wood	d^2h	$Y = e^{(4.16-0.00002 \cdot X)}$	–0.82	67.03	0.05	0.08
Bark	d^2h	$Y = e^{(2.63-0.00002 \cdot X)}$	–0.94	88.92	0.04	0.04
Branches	Age	$Y = 2.42 \cdot X^{1.03}$	–0.65	42.54	5.22	0.63
Leaves	h	$Y = 5.42-0.21 \cdot X$	–0.82	66.43	0.83	0.31
Roots	d^2h	$Y = e^{(2.77-0.00004 \cdot X)}$	0.74	54.53	0.32	0.22
		Birch				
Wood*	Age	$Y = e^{(4.64-0.00514 \cdot X)}$	–0.78	60.86	0.93	0.21
Bark*	Age	$Y = 1.75 \cdot X^{0.47}$	0.70	48.59	0.89	0.30
Branches*	d	$Y = e^{(0.13 \cdot X-0.37)}$	0.73	53.34	4.49	0.55
Leaves*	Age	$Y = e^{(0.014 \cdot X-0.25)}$	0.84	70.93	5.42	0.47
Roots	Age	$Y = 1/(6.67 \cdot 10^{-3} + + 1.53 \cdot 10^{-3} \cdot X)$	0.98	96.20	0.004	0.01
		Alder				
Wood*	h	$Y = 53.52 \cdot X^{0.10}$	0.77	58.78	0.01	0.03
Bark*	h	$Y = 31.50 \cdot X^{-0.31}$	–0.88	78.04	0.24	0.07
Branches*	Age	$Y = e^{(1.81 + 0.00999 \cdot X)}$	0.87	76.21	0.20	0.11
Leaves*	Age	$Y = 1.38 \cdot X^{-1.04}$	–0.97	94.07	0.14	1.34
		Alder				
Wood	Age	$Y = 25.20 \cdot X^{0.32}$	0.95	89.84	0.01	0.02
Bark	Age	$Y = 1/(0.04 + + 1.12 \cdot 10^{-3} \cdot X)$	0.78	61.18	0.01	0.003

Table 2. (continuation)

Fraction of the phytomass, Y	Argument, X	Regression	r**	R^2	σ^2	m
Branches	Age	$Y = 3010.92 \cdot X^{-1.64}$	–0.90	80.77	0.43	0.13
Leaves	Age	$Y = 290686.31 \cdot X^{-36.60}$	–0.86	74.80	2.25	0.33
Roots	Age	$Y = 678.58 \cdot X^{-1.27}$	–0.71	52.13	0.32	0.30

* % of the above-ground phytomass

** r is the coefficient of correlation; R^2 is the confidence interval; s^2 is the standard deviation; m is the correlation error

The intensity of redistribution of radionuclides in the system of geochemically linked landscapes was determined from data on the total pool of radionuclides in every particular elementary landscape within the catena. Data on external irradiation dose were taken into account as an indirect index.

Table 3. Bark proportions in various tree species, (weight %)

Species	Index			
	Inner (alive) bark			
	Stem top	Stem middle	Stem basement	Weighted average
Pine	43.2	22.5	14.9	20.1
Birch	77.2	72.2	34.7	69.4
Oak	60.5	46.8	31.0	45.4
Spruce	69.1	60.5	41.0	58.2
Aspen	69.4	68.5	30.4	64.8
Alder	53.8	31.5	26.1	37.6
Species	Index			
	External bark (cork)			
	Stem top	Stem middle	Stem basement	Weighted average
Pine	56.8	77.5	85.1	79.9
Brich	22.8	27.9	65.4	30.59
Oak	39.5	53.2	69.0	54.61
Spruce	30.9	39.5	59.0	42.8
Aspen	30.6	31.5	69.6	35.18
Alder	46.2	68.5	73.9	62.44

1.2. RADIOECOLOGICAL CHARACTERISTICS OF THE INVESTIGATED TERRITORY

By 1995, the area of Bryansk region contaminated by the radioactive fallout due to the Chernobyl accident comprised 171 thousand hectares (th. ha) (according to the data submitted by the Federal Forest Service of the Russian Federation). The deposition was distributed over the territory as follows: 37–185 KBq/m^2 (60% of the total contaminated area), 185–555 KBq/m^2 (23%), 555–1480 KBq/m^2 (15%), and higher than 1480 KBq/m2 (about 2%). The total contaminated area in Kaluga region reached 177.8 th. ha: 37–185 KBq/m^2 (75%), 185–555 KBq/m^2 (almost 25%), 555–1480 KBq/m^2 (less than 1%). The total contaminated area in Tula region was 74.8 th. ha: 37–185 KBq/m^2 (97%) and 185–555 KBq/m^2 (3%) [205]. In Ukraine, by 1995, the total contaminated area covered about 9 million hectares (mil. ha) with 1.75 mil. ha occupied by forests [189]. In Bryansk and Kaluga regions, the contaminated forest area was about 1900 and 810 sq. km, respectively.

A specific feature of the Chernobyl-derived fallout is high spatial and temporal heterogeneity of the radionuclide composition and physicochemical forms. Most of the radionuclides were incorporated into low-soluble, polydispersed, fine particles from several tenths to hundreds of micrometer in diameter (the so-called "hot particles"*). Chemical composition of the hot particles was, as a rule, represented by various uranium oxides (up to 90%) with some admixtures. The radionuclide composition of the hot particles is close to that of the irradiated nuclear fuel mixed in various proportions with volatile fission products [92, 111, 140, 142, 302]. Sometimes, hot particles are represented by the Fe–Zr matrix enriched with ^{103}Ru and ^{106}Ru (the so-called "ruthenium particles"). Hot particles may be enriched also with ^{144}Ce, ^{144}Ce+^{106}Ru, ^{144}Ce+^{134,137}Cs, or caesium only [314, 319, 323].

Larger hot particle (more than 200 μm in size) precipitated primarily in proximity to the accidental unit (the so-called "near zone"). The near zone contaminated during the first hours of the accident was enriched with the most insoluble particles composed basically of UO_2 [192]. Distant investigated territories (Bryansk, Tula, and Kaluga regions of Russia, Baltic countries, and West Europe) were contaminated by the fallout composed of more finely dispersed particles and condensed aerosols considerably enriched with iodine and (after iodine decay) caesium isotopes (the so-called "far zone"). The fallout in these regions consisted of the condensed component by 80-90% [72], and only of 10 to 20% of the deposition was attributed to rare "hot" particles of various radionuclide composition [179]. Thus, the fallout dispersion tends to increase and the proportion of fuel component decreases as the distance from the accidental unit increases [110].

In general, surface deposition over the central part of the East European Plain varied by 5–6 orders of magnitude: from several kBq/m^2 to hundreds MBq/m^2. In Russia, this index varied from several kBq/m^2 to several MBq/m^2. Minimum variation was characteristic of Tula region of the Russian Federation,

* The term "hot particles" is defined as solid radioactive particles of various nature (fuel, condensed, or absorbed) with specific activity higher than 0.1 Bq/μg.

Table 4. Average radionuclide composition of gamma-irradiating radionuclides and ^{90}Sr in the investigated territory in 1986 [15, 75, 81, 167].

Region	Radionuclide, %									
	I-131	Ru-103	Ru-106	Cs-134	Cs-137	Zr-95 + + N-95	L-40 + +B-140	Ce-141	Ce-144	Sr-90
Russia (reduced to 23.05.1986)										
Bryansk	25.0	25.5	–	13.2	25.0	2.7	7.8	–	–	0.8
Kaluga	24.6	24.8	–	14.1	26.1	1.8	8.1	–	–	0.5
Tula	20.9	33.6	–	12.5	24.0	1.9	6.5	–	–	0.6
30 km exclusion zone of ChNPP, Ukraine (reduced to 10.05.1986)										
"Near zone" 5–10 km	5.5	14.1	3.5	0.7	1.2	40.1	5.1	15.1	13.1	1.7
"Middle zone" 10–30 km	38.4	13.8	3.7	2.6	4.7	21.7	–	7.8	6.5	0.8

Table 5. Radionuclide composition in the soil of the basic key sites in 1987–88 (%)

Key site*	Radionuclide composition					
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{95}Zr	^{95}Nb
Russia (1988)						
Pl-1	–	18.4	75.2	6.4	–	–
Pl-2	–	17.9	76.7	5.4	–	–
Kl-1	0.04	18.3	75.9	5.8	–	–
Kl-2	0.05	18.4	75.3	6.2	–	–
Zl-1	1.50	18.1	74.2	6.2	–	–
30 km exclusion zone of ChNPP, Ukraine (1987)						
D-1	52.5	6.0	18.1	19.5	1.7	2.3
K-2	52.1	5.8	17.9	20.7	1.1	2.5
Sh-1	57.1	4.9	14.5	19.1	1.4	2.8

* See Table 1 for the site parameters

Table 6. Multiyear dynamics of the exposition dose rate of gamma-irradiation in the investigated key sites, mR/h

Key site	h, m*	Years							
		1987	1988	1989	1990	1991	1992	1994	1995
				Russia					
Pl-1	0	nd***	0.20	0.08	0.07	0.06	0.06	nd	nd
	1	nd	nd	nd	nd	0.05	0.04	nd	nd
Pl-2	0	nd	0.18	0.07	0.07	0.06	0.05	nd	nd
	1	nd	nd	nd	nd	0.05	0.04	nd	nd
Kl-1	0	nd	0.40	0.16	0.14	0.12	0.11	nd	nd
	1	nd	nd	nd	nd	0.09	0.08	nd	nd
Kl-2	0	nd	0.39	0.15	0.12	0.11	0.10	nd	nd
	1	nd	nd	nd	nd	0.09	0.07	nd	nd
Zl-1	0	nd	0.71	0.35	0.28	0.25	0.20	nd	nd
	1	nd	nd	nd	nd	0.20	0.15	nd	nd
				30 km exclusion zone of ChNPP, Ukraine					
D-1	0	0.68**	0.22	0.13	0.10	0.05	0.05	0.04	0.04
	1	nd	nd	0.07	0.05	0.03	0.04	0.03	0.03
D-3	0	0.73**	0.23	0.11	0.10	0.05	0.05	0.04	0.03
	1	nd	nd	0.07	0.05	0.04	0.04	0.03	0.02
K-2	0	nd	1.90	1.36	0.91	0.43	0.47	0.36	0.31
	1	nd	nd	0.72	0.52	0.34	0.33	0.27	0.23
Sh-1	0	95.0	57.60	36.5	12.93	6.32	5.94	5.04	4.47
	1	nd	nd	19.1	6.99	4.53	4.18	3.52	3.18

* Height measurements: 0 on the soil surface; 1 at 1 m above the surface

** Data of 1986

*** No data

Table 7. Multiyear dynamics of the deposition/dose rate ([σ]/[γ-dose rate]) : $(Ci/m^2)/(mR/h)$ in the basic key sites

Site	Height measurements, m*	Year							
		1987	1988	1989	1990	1991	1992	1994	1995
				Russia					
P1-1	0	nd***	50.8	128.3	148.0	173.1	187.6	nd	nd
	1	nd	nd	nd	nd	215.4	278.4	nd	nd
Pl-2	0	nd	56.9	152.0	166.8	193.8	202.0	nd	nd
	1	nd	nd	nd	nd	238.5	313.4	nd	nd
Kl-1	0	nd	42.7	109.3	124.5	141.5	152.2	nd	nd
	1	nd	nd	nd	nd	202.2	205.7	nd	nd
Kl-2	0	nd	39.7	101.3	127.4	134.1	149.8	nd	nd
	1	nd	nd	nd	nd	170.1	209.8	nd	nd
Zl-1	0	nd	52.2	103.4	128.3	141.5	178.3	nd	nd
	1	nd	nd	nd	nd	178.6	233.1	nd	nd
				30 km exclusion zone of ChNPP, Ukraine					
D-1	0	8.8	24.7	43.3	55.5	106.7	109.5	140.0	146.4
	1	nd	nd	89.7	113.6	192.0	153.3	233.3	256.2
D-3	0	9,7**	23.9	60.0	62.2	140.5	125.0	162.9	195.5
	1	nd	nd	93.8	130.2	200.0	161.3	231.6	307.7
K-2	0	nd	35.0	51.8	75.5	157.1	139.1	171.4	195.3
	1	nd	nd	98.6	133.3	200.0	200.0	230.8	266.4
Sh-1	0	12.6	16.1	30.4	81.3	158.5	160.2	171.0	188.4
	1	nd	nd	58.1	150.4	221.2	227.8	245.0	265.0

*Height measurements: 0 on the soil surface; 1 at 1 m above the surface
**Data of 1986
***No data

and the maximum absolute values (and variation) were found in the 30 km exclusion zone of ChNPP. The total initial deposition in this territory (except for ^{131}J) varied from 1.4 MBq/m^2 (in the marginal parts of the exclusion zone) to 370 MBq/m^2 (near the accidental unit).

Current spatial heterogeneity of the deposition usually obeys the logarithmically normal distribution [7, 93, 173]. The averaged coefficient of variation of this index in the exclusion zone is about 30–35%. The closer the accidental unit, the more manifested is the microspot, mosaic-like character of the deposition pattern because of higher proportion of the fuel particles and non-random distribution of the fallout over the territory [344].

Radionuclide composition of the fallout also varied depending on the distance from ChNPP. In the "near zone," it was close to the fuel composition in the reactor at the moment of the accident. The proportion of volatile elements (iodine and caesium) increased as the proportion of infusible radionuclides (Ce, Zr, Nb, etc.) decreased with distance from ChNPP (Table 4).

Radionuclide composition of the fallout deposited on the forests in Bryansk, Kaluga, and Tula regions was roughly the same. ^{134}Cs and ^{137}Cs were the main dose-forming radionuclides in these territories for several years after the accident. By September of 1988, the proportion of caesium isotopes in the deposition was more than 90%, whereas the proportion of ^{90}Sr did not exceed 1–2% (absolute deposition of ^{90}Sr was 7.4–29.6 kBq/m^2) (Table 5).

The total caesium proportion in the exclusion zone varied within the range of 19–24%, with somewhat increase toward marginal parts of the territory (August, 1987).

The above features were among the leading factors that predetermined the initial distribution of radionuclides in the soils, their biological availability, and accumulation by plants particularly in the first years after the accident.

The irradiation dose rate also varied over the territory depending on both the deposition rate and the composition of radionuclides. Shortly after the accident, the dose rate varied from 0.2 to 0.7 mR/h in the territory of the Russian Federation and, from 0.7 to 95 mR/h in the exclusion zone (Tables 6, 7).

The dose rate quickly decreased in time, and the rate of this decrease declined as well. On the one hand, this was due to fast radioactive decay of the short-lived isotopes (currently the dose rate is almost completely determined by ^{137}Cs), and on the other hand, forest litter prevented the radionuclides from fast downward migration.

No linear dependence between the deposition and dose rate coefficient A was observed, which is reflected in the following equation:

$$A = \sigma/(DR_{actual} - DR_{background}),$$

where σ is the deposition, Bq/km^2; DR_{actual} is the actual (measured) value of the dose rate; and $DR_{background}$ is the average value of the dose rate before the accident (10 μR/h).

Thus, in spite of the agreed dynamics of the dose rate and deposition, the last value may unlikely be estimated from the first one directly, since the dose rate depends on a wide range of other parameters: downward radionuclide migration; intensity of biological cycles, microtopography, etc.

1.3 VEGETATION AND SOIL COVER

1.3.1. Vegetation

Basic characteristics of the phytocenosis (number of vertical levels in the forest ecosystem, structure of the levels, tree height and crown projection, vegetative stage, etc.) and species composition play an important role in the primary interception of the radionuclides by the overstorey, since all these factors determine the area of effective interception of the fallout. In the middle-age stand (quality class 0.8) the index of leaf surface is 2.1–2.2 ha/ha* in pine forests, 2.2–3.1 ha/ha in aspen forests, and 3.2–4.3 ha/ha in birch forests [48]. In other words, upon similar conditions, birch stand is able to retain much higher proportion of the fallout compared to the other tree species. Obviously, other tree components (organs) directly exposed to the fallout (leaves, needles, branches, and external bark) were contaminated initially at a maximum extent, whereas the covered tissues and organs, such as wood and roots, were less contaminated.

Secondary contamination of forest vegetation is accounted for the root uptake of the radionuclides along with the soil nutrients and stable elements. It is reasonable to suppose that the degree of secondary contamination is in general correlation with the indices of biological production, i.e., the maximum intensity of the radionuclide uptake is characteristic of those tree components and organs characterised by the maximum growth rate. Therefore, the radionuclide content in the permanently growing organs (stem, roots, branches, etc.) reflects the accumulative radionuclide dynamics, whereas the contamination of the short-lived tree components (leaves, needles, generative organs) is in closer correlation with actual radionuclide uptake from the soil.

The Chernobyl fallout has affected a vast territory comprising various natural zones and provinces [194]. The variation of local soil conditions, stand age, structure, and species composition has determined a range of specific features of the vegetation-fallout interaction.

Pine forests occupy most of the contaminated forest area. In the investigated territory of Ukraine, pine forests occupy 51% of the total forest area, and the corresponding indices for the Bryansk, Kaluga, and Tula regions are 42% and 20–40%, respectively [25, 42, 62, 187]. The green-moss, bilberry, and sphagnum types of the understorey correspond to the "fresh", "wet", and "moist" forest types, which are very common in the investigated territory. By species composition, the investigated forests are represented by monodominant pine plantations (*Pinus sylvestris*) and co-dominant mixed forests composed of *Quercus robur* and *Pinus sylvestris*, with an admixture of lime tree (*Tilia parvifolea*). Such species as *Betula verrucosa, Populus tremula,* and *Picea abies* are also of wide occurrence [58, 186]. Pure pine forests are very common in the Ukrainian Polessie. In Tula, Kaluga, and Bryansk regions they yield dominance to mixed, pine-broad-leaved forests.

Along with *Pinus sylvestris*, the stands in the Ukrainian Polessie also include oak (*Quercus robur*),birch (*Betula verrucosa and Betula pubescens*), aspen (*Populus tremula*), spruce (*Picea abies*), and black alder (*Alnus glutinosa*). A complex topography of the contaminated territory and variable mois-

* $ha_{leaves} / ha_{territory}$

Table 8. Fractional structure of the tree phytomass in 1986 – 1987 (% of total phytomass): numerator is the range of variation (max-min), denominator is the mean value

Species	Wood	Bark		Branches		Needles/Leaves		Roots
		Inner (alive)	External (cork)	Small	Large	1st year	>1 year old	
Pine	68.8–71.7 / 70.8	1.0–1.3 / 1.1	3.8–5.1 / 4.2	0.7–1.1 / 0.8	5.2–6.3 / 5.6	0.9–2.3 / 1.1	1.7–2.5 / 2.0	13.8–14.9 / 14.4
Birch	62.9–72.5 / 68.9	5.7–8.4 / 7.0	2.6–3.6 / 3.1	1.7–6.4 / 3.5	2.0–8.5 / 4.1	1.1–1.8 / 1.5	–	8.8–14.8 / 11–6
Oak	55.7–61.9 / 59.5	5.4–6.1 / 5.8	6.5–7.3 / 7.0	1.1–1.9 / 1.5	2.4–4.1 / 3.2	2.2–3.0 / 2.5	–	18.1–24.9 / 20.5
Aspen	65.4–69.7 / 67.9	7.1–8.3 / 7.6	3.9–4.5 / 4.1	2.3–3.9 / 3.3	4.5–6.8 / 5.5	1.7–3.0 / 2.2	–	7.6–11.1 / 9.6
Spruce	53.0–56.4 / 54.8	8.1–8.8 / 8.5	5.9–6.4 / 6.17	2.5–3.4 / 3.0	3.9–4.1 / 4.0	0.9–1.1 / 1.0	4.0–5.4 / 4.7	17.4–18.3 / 17.9
Alder	74.1–75.9 / 75.0	3.3–3.5 / 3.4	5.5–5.8 / 5.6	2.1–2.6 / 2.4	3.8 / 3.8	2.0 / 2.0	–	7.5–8.2 / 7.9

Table 9. Annual weight increment of various tree components (% of the total tree phytomass increment)

Species	Wood	Bark		Branches		Needles/Leaves	Roots
		Inner (alive)	External (cork)	Small	Large		
Pine	44.7	0.3	1.2	1.0	0.8	42.1	10.3
Birch	30.2	6.5	2.8	3.9	5.8	51.1	No data
Oak	25.3	2.5	2.9	1.7	3.5	46.6	17.6
Aspen	25.1	0.6	0.3	1.2	5.0	72.5	No data
Spruce	41.8	3.9	2.8	1.0	1.8	40.8	8.7
Alder	40.2	1.3	2.1	1.2	1.0	49.7	4.5

ture conditions and soil fertility determine a high typological diversity of the forests. Long-term anthropogenic activities in the investigated territory have resulted in the replacement of primary forest vegetation by secondary, small-leaved species [176].

In 1986, soon after the accident, a large-scale radioecological monitoring network was established in the most typical landscapes (eluvial, trans-eluvial, and accumulative) which represented a wide diapason of environmental conditions and radionuclide deposition.

Arboreal vegetation constitutes 95 to 99% of the total above-ground phytomass. It yields 72–84% of the total annual phytomass in forest phytocenoses of temperate climatic zone [25, 158, 281]. Arboreal vegetation determines basic parameters of carbon and nutrient accumulation and cycling in the investigated forest ecosystems, in spite of considerable typological differences and wide variation of the ecosystem parameters.

Phytomass Composition and Reserves

The phytomass reserves, structure, and growth dynamics are of prime importance for radionuclide retention by arboreal vegetation. These parameters have a great impact on the rate of radionuclide accumulation by both surface and inner tissues of the trees. Knowledge on (i) fractional composition and (ii) reserves of forest phytomass is necessary for any ecological (and biogeochemical) research, since these two factors affect (i) the intensity of nutrient (radionuclide) uptake and (ii) the total capacity of the biological cycle.

The structure (composition) of the tree phytomass is considered in a range of fundamental research works [9, 48, 79, 158, 214]. According to these authors and our experimental data and calculations, wood constitutes more than 50% of the total phytomass stock (Table 8). Minimum contribution (< 1%) is typical of generative organs [58, 214].

The calculations confirmed considerable differences in the phytomass structure of different species, which depends on the crown structure, peculiarities of photosynthesis, conditions of root nutrition, etc. For instance, the ratio wood/roots/branches/bark/needles (leaves) is 21/4/2/2/1 for pine, 8/3/1/2/1 for spruce, 27/9/6/3/1 for oak, 41/6/7/4/1 for birch, 24/4/3/5/1 for aspen, and 38/4/5/3/1 for alder.

The composition of the annual phytomass production in the mature stand is considerably different from the fractional structure of the total phytomass (Tables 8, 9). The most productive tree parts are wood and leaves (needles) characterised by a similar annual increment (30–50% of the total phytomass). The branch phytomass grows slower, and the least increment is characteristic of bark and, sometimes, roots. In this connection, the wood of tree rings formed after the Chernobyl accident, as well as leaves and needles have the highest potential for radionuclide accumulation.

The maximum phytomass reserves in the territory of Kiev and Bryansk regions were found to be in pine stands. In Kaluga and Tula regions, the highest phytomass reserves are concentrated in spruce and birch (*Betula verrucosa*) stands, respectively.

The average annual increase in the phytomass in the investigated territory during the post-accidental years was estimated at 3–7 t/ha, which is about 7–10% of the total arboreal phytomass by the time of the fallout.

The investigated territory comprises two climatic belts, i.e., boreal and subboreal, and includes two natural zones, i.e., south taiga and forest-steppe. In the taiga zone, it includes the soddy-podzolic sub-zone with two soil facies (soil geographical class): shortly freezing and freezing soils of temperate climatic zone (the Belorussian and Central Russian soil provinces, respectively). In the forest-steppe zone, the soils represent the facies of temperate freezing soils (the Oka–Don province) [77]. A great diversity of soils in the investigated area makes it possible to trace the impact of different soil processes and properties on the migration of radionuclides.

The investigated soils represent the following soil types: chernozems, soddy-podzolic and podzolic, alluvial meadow-bog, alluvial bog, clay-humus bog, peat-bog, and peat bog soils. By hydrological regime (the most influential factor of radionuclide migration), these soils are pooled into two basic groups: automorphic soils (not connected to the capillary fringe and not exposed to lateral addition of water, i.e., developed under atmospheric moistening only: watersheds, upper terraces, etc.) and hydromorphic soils (formed under conditions of additional lateral or capillary moistening: local depressions, hollows, bogs, floodplains, etc.) [8, 133, 171, 233].

Characteristics of Forest Litters

Although forest litter is believed to be a part of the soil profile, it is a very specific and, to a large extent, independent component of biogeocenosis and is different from the "classical" soil in many respects [90, 103, 229]. It consists mainly of organic matter, and its thickness and mass undergo considerable seasonal and long-term variations [103]. On the one hand, forest litter is a sink for nutrients, and, on the other hand, it often determines the amount and composition of the organic matter in deeper horizon, which influences the radionuclide mobility [2, 105, 106]. The role of forest litter is especially important in sandy soils, where it is the only obstacle (biogeochemical barrier) to fast vertical and horizontal radionuclide migration in the soils and landscapes. Most of the researchers share the opinion that forest litter serves as a long-term sink for a considerable portion of radionuclide deposition [241, 242, 277, 290]. The contribution of forest litter to the radionuclide retention depends primarily on its thickness and composition [250, 280].

Forest litters in the investigated regions represent virtually the entire spectrum of its possible types: from raw, peat-like *mor* to almost completely humified *forest mull* containing a large proportion of the humus-mineral and humus-clay complex compounds [197]. A well-manifested forest litter is characteristic of most of the investigated forest ecosystems, which is an evidence of the undisturbed soil surface for at least the recent 10–20 years (the time needed for its formation).

The average thickness of forest litter in various ecosystems is 4–5 cm (Table 10). Variation of this index depends on ecosystem type: the maximum value of the variation coefficient (V) is typical of the broad-leaved and mixed, coniferous-broad-leaved forests. This index is relatively low for pine stand (plantations and natural forests). At the same time, variation in the thickness of forest litter reflects regional differences in the genesis and composition of litter

Таблица 10. Variation of the forest litter inventory and thickness in the investigated key sites (numerator is the inventory g/m^2; denominator is the thickness, cm)

Type of biogeocenosis, (composition of the forest litter)	Statistical indices (see Footnote)						
	n	M	±m	max	min	G	V, %
Bryansk region (Russia)							
Pine forest with moss cover (moss and needles)	16	4.91 5.02	0.27 0.34	7.5 8.03	3.00 3.61	1.08 1.35	21.9 26.8
Pine plantation (moss and needles)	16	4.29 2.97	0.23 0.18	6.00 4.48	2.7 1.3	0.93 0.74	21.6 24.9
Mixed, broad-leaved-pine forest (needles and leaves)	16	4.93 4.57	0.35 0.41	8.00 7.54	3.00 2.15	1.41 1.63	28.6 35.7
Pine forest with mixed cereal/sedge cover (moss, needle, and grass)	16	4.21 4.78	0.29 0.34	6.5 6.79	2.00 2.37	1.16 1.37	27.5 28.7
Pine plantation (grass and needles)	16	4.25 4.29	0.24 0.37	6.00 7.88	2.00 1.61	0.95 1.48	22.3 34.6
Kaluga region (Russia)							
Mixed, broad-leaved-pine forest (needles and leaves)	24	4.87 3.96	0.34 0.41	7.50 7.22	2.50 0.42	1.68 1.64	34.4 41.5
Deciduous forest (leaves)	24	4.04 4.19	0.19 0.16	6.70 5.71	3.00 2.88	0.93 0.79	23.0 18.8
Mixed, broad-leaved-pine forest (needles and leaves)	24	3.86 28.5	0.25 0.21	7.0 6.3	1.50 1.33	1.25 1.05	32.5 36.8
Open pine forest on a sphagnum bog (sphagnum debris)	24	11.31 6.56	0.65 0.60	18.5 9.85	5.50 2.93	3.17 1.98	28 33
Pine forest with grass cover (needles and grass)	5	4.30 nd*	nd	5 nd	3.5 nd	nd	nd
Spruce forest with green moss cover (moss-needles)	5	7.3 nd	nd	7.0 nd	6.5 nd	nd	nd
Alder forest (leaves)	5	3.1 nd	nd	4.0 nd	2.0 nd	nd	nd

* no data

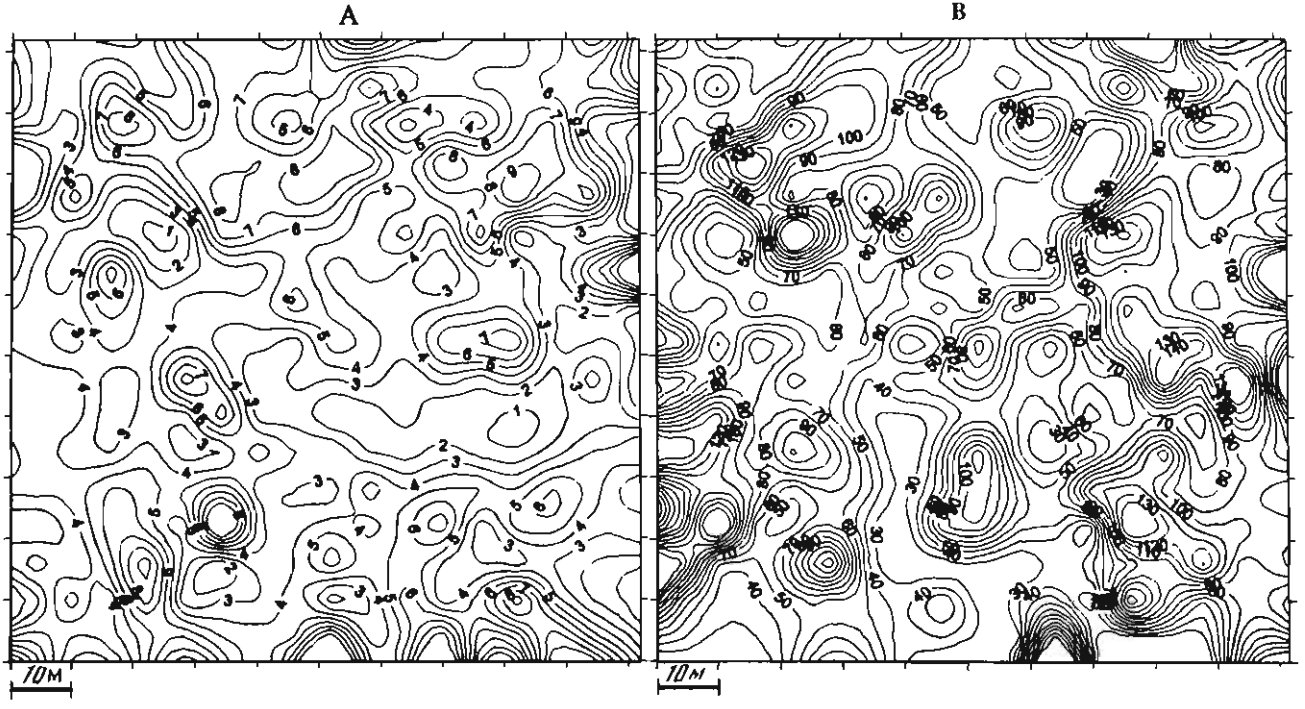

Fig. 4. Spatial distribution of the forest litter (A) thickness (cm) and (B) inventory (g/dm^2, dry weight) in a pine forest (Ukraine)

horizons anthropogenic load over the given territory. For example, the variation coefficient of the forest litter thickness in pine forests of Ukrainian Polessie with well-manifested mesotopography is higher than that in pine forests of Bryansk region by a factor of 1.5. In forest plantations of Kaluga region, forest litters usually have an incomplete profile with the weakly manifested, fragmentary A0h horizon. The variation in the thickness in such forest litters also exceeds the average value.

Thus, spatial variability of the forest litter thickness exceeds 48% even within the relatively small areas [102]. Spatial heterogeneity causes the corresponding variation in the intensity of radionuclide migration in the soil-forest litter system.

By our data, the reserves of organic matter in forest litters reach 3–5 kg/cm^2 in Bryansk region, 2 to 4 kg/cm^2 in Kaluga region, and about 7 kg/cm^2 in pine forests of Ukrainian Polesie (Table 10). These values are in a good agreement with the data by other authors (2 to 10 kg/cm^2) [115]. Variation of the reserves of forest litter is roughly the same as the variation in its thickness (Table 10).

At the same time, isolines characterising the forest litter thickness are in a closer correlation with the microtopography of the investigated site than the corresponding isolines of forest litter reserves (Fig. 4). In other words, spatial variability in the reserves of forest litter is higher than spatial variability in the forest litter thickness. A schematic map of the forest litter pattern is, therefore, the most reliable tool to characterise the distribution and intensity of the processes running in the forest litter-mineral soil and soil-plant systems, and to assess the efficiency of possible mitigation measures.

Particle-Size Composition of Soils

The forests in Bryansk and Kaluga regions and in Ukrainian Polessie commonly occupy the areas with coarse (sandy and sand-loamy) soils. The only exception is podzolized chernozems of Tula region that have a silty clay loamy consistence (Table 11) with the dominance of coarse silt and fine clay (<0.001 mm) fractions that, taken together, constitute about 70% of the soil mass.

The mass proportion of the integral silt plus clay fraction in chernozem is about 70%. The percentage of sand fraction in less than 1%. The expected transfer of chemical elements, including radionuclides, together with infiltrating water in these soils is expected to be lower than in sandy soils of the other investigated regions. In addition, a high content of clay minerals makes the investigated podzolized chernozems highly capable for intensive irreversible adsorption of caesium isotopes.

The content of the so-called physical clay (< 0.01 mm) in the upper horizons of soddy-podzolic soils of the southern taiga zone varies from 4.6 to 11.4% (Table 11). The total content of fine clay (< 0.001 mm) and clay (0.005–0.001 mm) is 1.9 to 4.1% and 0.1–4.7%, respectively. Therefore, the investigated soils are poor in the fractions capable for irreversible radionuclide sorption, and relatively high soil-to-plant transfer factors (TF) are expected.

The soils of Ukrainian Polessie (represented by the exclusion zone) possess, as a rule, a somewhat coarser consistence. The adsorption capacity of these soils is not high, and some fixation of the chemical element is manifested only in the upper 2–5 cm of the soil profile with the maximum clay content. The

Table 11. Vertical distribution of the particle-size composition of the investigated soils

Depth, cm	Particle size, mm							Classification by texture
	1–0.25	0.25–0.05	0.05–0.01	0.01–0.005	0.005–0.001	<0.001	< 0.01	
				Tula region Russia, (*podzolic, clayey chernozem*)				
0–10	0.4	5.5	42.1	13.3	14.5	24.2	52	Silty clay loam
20–30	0.4	5.4	43.7	11.1	14.4	25	50.5	Silty clay loam
40–50	0.4	4.4	43.6	10.2	12.4	29.1	51.6	Silty clay loam
60–70	0.5	6.5	39.5	10.6	11.8	31.1	53.5	Silty clay loam
80–90	0.5	5.9	40.8	8.6	12.3	31.9	52.8	Silty clay loam
100–110	0.5	41.2	7.9	11.1	33.5	33.5	52.6	Silty clay loam
120–130	1	4.6	39.1	10.6	12.2	32.5	55.3	Silty clay loam
140–150	1	5.1	43.6	10.7	10.5	29.2	50.4	Silty clay loam
				Bryansk and Kaluga region,Russia, (*podozolic sandy soils*)				
5–15	29.4	37.2	15.2	2.1	0.7	3	5.8	Cohesive sand
6–15	30	50	14.8	0.7	1.2	3.3	5.2	Loamy sand
4–18	21.8	57.6	13.1	0.4	3.6	3.5	7.5	Cohesive sand
6–15	34	45.4	13.2	0.9	4.2	2.3	7.4	Cohesive sand
5–22	36.6	38.4	19.4	0.7	1.6	2.3	4.6	Cohesive sand
4–19	25.6	54.2	12.9	0.8	2.9	2.9	6.6	Cohesive sand
120–130	17.5	75.6	1.9	2.2	0.8	2	5	Cohesive sand
130–140	12.3	82.1	2.1	0.3	0.3	2.9	3.5	Loose sand

Kiev region, Ukraine

Podzolic sandy soils (1)

0–10	48.2	48.2	2	0.1	0.3	1.2	1.6	Loose sand
10–17	53.2	42.7	2	0.3	0.7	1.1	2.1	Loose sand
17–24	45.3	49.4	2.4	0.2	0.8	1.9	2.9	Loose sand
24–32	44.2	51.8	1.7	0.1	0.8	1.4	2.3	Loose sand
32–40	54.2	42.2	1.6	0.3	0.6	1.1	2	Loose sand
40–50	38.5	57.2	2.6	0.2	0.4	1.1	1.7	Loose sand
50–60	54.2	38.3	5.1	0.5	0.5	1.4	2.4	Loose sand
60–70	47.3	50.3	1.4	0	0.5	0.5	1	Loose sand
70–80	44.5	53.3	1.3	0	0.3	0.6	0.9	Loose sand
90–100	52	45.8	1.7	0.1	0.3	0.1	0.5	Loose sand

Podzolic sandy soils (2)

0–5	42.6	33.4	19.4	0.7	1.6	2.3	4.6	Loose sand
5–15	47.9	38.7	10	0.4	1.3	1.7	3.4	
15–20	47	41.1	0	0.3	1.6	2	3.9	Loose sand
20–30	43.4	40.7	10.7	0.6	1.7	2.9	5.2	Cohesive sand.
30–45	51.2	38.5	6.1	0.2	1.1	1.9	4.2	
55–65	60.7	36.5	1.6	0.1	0.2	0.9	1.2	Loose sand

Podzolic sandy soils (2)

65–75	59.5	37.3	2	0	0.2	1	1.2	Loose sand
75–85	44.2	48.3	6	0.1	0.5	0.9	1.5	Loose sand
85–95	47.4	40.9	9.3	0.1	0.6	1.7	2.4	Loose sand
110–120	39.5	40.7	17.6	0.2	0.7	1.3	2.2	Loose sand

Table 11. (continuation)

Depth, cm	Particle size, mm							Classification by texture
	1–0.25	0.25–0.05	0.05–0.01	0.01–0.005	0.005–0.001	<0.001	< 0.01	
				Podzolic sandy soils (2)				
0–6	26.3	54.2	12.9	0.8	2.9	2.9	6.6	Cohesive sand
6–8	37.4	48.2	10.8	0.3	1.6	1.7	3.6	Sand
8–10	38.9	51.9	6.7	0.1	1	1.4	2.5	Loose sans
10–14	34.3	55.7	7	0.2	1.3	1.5	3	Loose sand
14–23	40	49.9	7.4	0.3	1.2	1.2	2.7	Loose sand
23–31	44	48.9	4.8	0.1	1.1	1.1	2.3	Loose sand
31–40	31.5	59.4	6.2	0.2	1.2	1.5	2.9	Loose sand
40–47	31.1	58.7	7.7	0.2	0.9	1.4	2.5	Loose sand
60–72	45.6	48.8	3.9	0	0.6	1.1	1.7	Loose sand
100–110	24.4	46.4	26.2	0.5	1.3	1.2	3	Loose sand
				Alluvial, meadow-bog soils				
0–5	0	4	55.3	9.9	15.6	15.2	40.7	Clay loam
5–10	0	5.5	51	7	15.5	21	43.5	Clay loam
10–15	0	3	55	8	15	19	42	Clay loam
15–25	0	11.1	61.1	6.7	10	11.1	27.8	Sand loam
40–50	0	14.4	57.7	0	5	22.9	27.9	Sand loam
60–70	0	34.9	50.5	0	1.9	14.1	14.6	Loamy Band

physical properties of the soils are characterised by low bulk and specific densities, low water capacity, and high water permeability. The soil structure is not pronounced and the factor of potential aggregation does not exceed 3%. All these factors make sand soils very vulnerable to erosion.

Alluvial soils of the investigated territory have a loamy and clay-loam consistence (Table 11). Down the soil profile, the relative proportion of sandy fractions increases, so that lower horizons (deeper than 70 cm) have a loam- sandy consistence. The soils are formed on alluvial sediments with numerous thin clayey laminae. A high content of fine particles ensures the high adsorption and water-holding capacities of alluvial soils, as well as their good aggregation. These features determine the high retention capacity of these soils for radionuclides.

Mineralogical Composition

The mineralogical composition of the investigated soils varies considerably and depends on the genetic type of soils. In chernozems, the main components of the clay fraction (< 0.005 mm) are dioctahedral illites (with iron admixture in the octahedral layers) and labile silicates. The latter are represented by irregular mixed-layered silicates with some smectite admixture. The proportion of labile minerals increases, and the proportion of illite decreases with depth. There are also some amounts of quartz, kaolinite, and chlorite in the soil profile as well. The chlorite content increases downward the soil profile. In chernozems, labile minerals predominate in the mineralogical composition, which results in 10–100 times more effective adsorption of caesium by chernozems compared to other investigated soils.

Chloritized structures, dioctahedral illites, quartz, and feldspars predominate in the mineralogical composition of soddy-podzolic soils. Chlorites proper are found in the soil profile below 45–50 cm; their content increases gradually with depth. The content of minerals with labile structures gains its maximum in upper horizons. Labile silicates are represented by the mixed-layered illite-smectite and illite-vermiculite. Dioctahedral illite and chlorite predominate in the deep soil overlaying the moraine loam. Some amounts of quartz and chlorite are also present. Labile silicates are represented by mixed-layered, irregular illite-smectite and illite-vermiculite.

Chloritized structures, dioctahedral illites, and quartz dominate also in podzolic soils. Some proportion of chlorite is found in the soils under pine forests. All soils contain some admixture of labile minerals represented by vermiculite and mixed-layered illite-smectite. A very specific mineralogical composition was found at a depth of 40–55 cm in the near (10 km) zone of ChNPP: well-crystallised montmorillonite with quartz admixture.

Quartz is the dominant component of the clay fraction in hydromorphic soils. It is accompanied with a small proportion of stratified silicate-chloritized structures and dioctahedral illite (in deeper horizons). The illite content increases in the middle of the soil profile. Gley horizons contain well-crystallised montmorillonite with an admixture of kaolinite, quartz, and illite.

Thus, the total adsorption capacity of the investigated soddy-podzolic, podzolic, and hydromorphic soils on glaciofluvial deposits is expected to be quite low because of a small proportion of clay minerals, predominance of the minerals with a regular stiff structures (quartz, chlorites, illites, etc.) and iron coat-

ing over mineral grains, which prevents cations from penetration into the interlayer spaces of labile minerals. More intensive radionuclide adsorption is expected to be pronounced in thin upper mineral layers of acid soils with more labile chloritized structures.

Chernozems and alluvial meadow-bog soils possess one to two orders of magnitude higher adsorption capacity for ^{137}Cs compared to the other investigated soils, because of the clayey consistence and higher proportion of labile structures.

Chemical and Physicochemical Soil Properties

The investigated soils are very different by their chemical and physicochemical properties (Table 12).

Podzolized chernozems of the contaminated part of the northern forest-steppe zone (Tula region) are characterised by a well pronounced (though rather thin) humus horizon with an average humus content in the upper layer about 5%. The cation exchange capacity in horizon reaches 22–23 mg-equiv/100 g. The main part (73%) of exchangeable cations is represented by Ca^{2+}; the shares of Mg^{2+}, K^{+}, and H^{+} constitute 14, 1, and 11%, respectively. In the podzolized horizon, the content of exchangeable hydrogen increases to 4 mg-equiv/100 g and then quickly decreases with depth. The base saturation degree changes down the soil profile in agreement with these regularities.

Podzolized chernozems have a slightly acid reaction and weak alkaline reaction in upper horizons and a slightly alkaline reaction in deep layers. Soil supply with nutrients is not high: the contents of available nutrients are as follows: phosphorus, 3–4 mg/100 g; potassium, 10–11 mg/100 g; ammonium and nitrate nitrogen, 1–3 and 0.4–0.8 mg/100 g, respectively (average for the 0 to 20 cm layer). In general, the phosphorus content decreases down the soil profile, which is likely due to intensive root uptake of this nutrient. Profile distribution of potassium is rather uniform, whereas nitrogen tends to accumulate in the upper part of the profile.

The investigated soils of Kaluga and Bryansk regions may be pooled into two main groups by their humus status: organic and mineral. The first group includes alluvial mucky bog soils and peat gley soils. Upper layers of these soils consist of peat and organomineral horizons (7 to 90% of organic matter, Table 12). Gleyed horizons of the organomineral soils contain less organic matter (7–9%). Peat horizons contain 18 to 32% of organic matter depending on the degree of mineralization and a mass proportion of mineral particles in the organic horizon. The humus content depletes below 1% in the layers deeper than 1 m.

Peat-gley bog soils differ from the alluvial soils in a thinner organic layer (50–60 cm). Upper horizons (T1 and T2) are represented by low-ash peat with the average organic matter content of 85–90%. A low mineral content in these horizons determines their loose consistence, good water permeability, and relatively low adsorption capacity, which may lead to intensive radionuclide leaching. Deeper horizons TG (peat-gley) and G (gley) contain 20–25 and 1–3% of organic matter, respectively, but the organic matter (particularly in TG) is considerably different from peat: it consists of dark black and dark brown humus compounds tightly bound with mineral soil compo-

nents. Principally, these particular layers are likely to be the most manifested geochemical barrier preventing vertical and lateral migration of soluble matter. The effectiveness of the barrier depends, however, on local physicochemical and moisture regimes.

The investigated mineral soils in the taiga zone are represented by podzolic and soddy-podzolic sandy soils with various types of organic profile.

(1) *Humus-iron-illuvial podzolic soils* exhibit a complex humus distribution down the profile. The organomineral compounds are leached from upper horizons and accumulate in iron-illuvial (B_{hf}) horizons in the form of several laminae (iron pans) of a higher density. These pan retard water migration, which results in temporarily stagnant water regime, the development of reducing regime (gley process), and mobilisation of most of the metal elements, including radionuclides. Since B_{hf} horizons are heterogeneous and contain water permeable sand fragments reaching the confining bed (moraine), these horizons are unlikely to retain considerable amounts of mobile compounds. Parent rocks in the region are represented primarily by clayey, impermeable moraine that serves as a geochemical barrier for vertical migration of liquids and may promote lateral migration.

(2) *Soddy-podzolic and podzolic weakly differentiated sandy soils* are very different from the humus-iron-illuvial podzols. They develop from glaciofluvial sandy rocks. Their profile is weakly differentiated, devoid of a local confining bed, and highly permeable. The humus horizon is represented by a lower (A0h) layer of forest litter or/and a thin organomineral horizon (A1) that lies immediately beneath the forest litter layer. The rest of the soil profile is virtually humusless; it is rather permeable for all potentially mobile compounds. Some of the soils (see key site L-1) possess a somewhat thicker humus horizon. Low but stable humus content (0.1–0.2%) in the middle and deep layers of these soils is likely to be an inherited feature and has nothing to do with current processes of humus redistribution.

In general, the investigated soils in the southern taiga zone belong to the group of acid unsaturated low-buffer soils. Their adsorption capacity is usually less than 8 mg-equiv/100 g in upper horizons and somewhat higher in the parent rocks (ash-rich peat in alluvial and bog soils or moraine clay in mineral soils). The proportion of bivalent cations Ca^{2+} and Mg^{2+} in the cation exchange complex (CEC) is maximum in upper soil layers and constitutes 15 to 40% of the CEC (Table 12). This suggests that the behaviour of these elements should be significantly dependent on their biological cycle. The soil solution has an acid reaction due to a high concentration of soluble organic compounds (mainly fulvic and low-molecular organic acids). Minimum pH is characteristic of upper soil horizons.

Peat gley bog soils have a strongly acid reaction: pH of water and salt (KCl) extracts is 3.6–4.4 and 3.1–3.3, respectively (Table 12). Our particular interest was focused on T2/G horizon, which is a transitional horizon from the peat to mineral gleyed layers. This acid (pH 4.1) horizon is characterised by a high content of exchangeable Al and H, i.e., it is rich of soil colloids of variable charge. Such horizons usually possess high buffer capacity and stable acidity; they provide conditions for the mobilisation and migration of various elements and compounds.

Table 12. Chemical and physicochemical soil properties in the territory of Russian Federation (remote zone), means of n = 3

Soil horizon	Horizon thickness (cm)	pH		Humus content, %	Hydrolytic acidity mg-equiv/100 g
		water	KCl		
Tula region					
Podzolic clayey chernozem on loess loam (key site Pl-1)					
A	1–19	6.2	5.3	5.2	2.4
AB	19–40	6.1	4.9	2.4	3.9
B1	40–58	6.4	5	1.5	2.7
B2	58–90	6.6	5	0.8	1.8
BC	95–120	7.5	6.1	0.5	0.8
C	125–200	8.1	7.2	0.5	nd*
Kaluga region					
Podzolic, weakly stratified, sandy soil on fluvioglacial depositions (key site L-1)					
AE	5–11	4	3.2	1.2	4.4
EB	11–20	4.5	3.9	0.8	3.4
B	20–40	4.7	4.3	0.3	2.5
BC	40–62	4.8	4.4	0.2	1.9
C	62–180	5.4	4.3	0.1	2.5
Soddy-podzolic soil on sandstone eluvium (key site Kh-2)					
A	5–11	4.7	3.6	1.4	4.2
EB	11–34	5.3	4.3	0.1	0.9
B1	34–54	5.5	4.5	0	0.8
BC/C	54–110	5.7	4.4	0.1	1
D	110–120	5.9	4.3	0.1	1.3
Podzolic, iron-illuvial, gleyic, sandy soil on double-layered deposition (key site E-1)					
AE	5–18	4.2	3.7	0.6	1.6
Eg	18–47	4.4	4.1	0.1	1.7
Blh, f	53–86	4.6	4.3	0.4	1.9
BC	86–96	5.1	4.6	0.0	1.3
Dg	100–160	5.1	3.6	0.1	7.9
Bog-peat soil on alluvial deposition (key site Kh-3)					
T1	7–15	4.9	4.2	26.9***	14.2
T1/G	15–25	5.5	4.3	8.0***	6.2
T2	25–47	5.4	4.3	18.6***	10.1
T2/G	47–70	5.5	4.5	9.3***	8.7
T3	70–93	5.4	4.5	31.8***	16.5
G	93–120	5.9	4.5	7.1***	5.6
CG	120–160	5.7	4.6	0.3	1.2

Exchange cations				Total exchange capacity	Cation exchange capacity %	Available nutrient, mg/100 g			
Ca^{2+}	Mg^{2+}	K^{+}	Al^{3+}			P_2O_5	K_2O	NH_4	NO_3
mg-equiv/100 g									
15.7	3.1	0.2	nd	19	88.8	3.6	10.6	2.5	0.6
16.3	2.5	0.1	–	18.9	83	6.7	4.9	1.7	0.1
15.4	2.4	0.1	–	17.9	86.9	10.3	4.9	1	0.1
14.3	2.4	0.1	–	16.8	90.3	14.7	5	0.7	0.1.
14.4	2.5	0.1	–	17	95.5	12.1	4	0.7	0.2
nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
1	0.2	0.1	0.8	2.1	22.8	2.2	2.1	0.3	0.1
0.6	0.1	bd**	0.9	1.6	17.1	2.9	1.1	0.1	0.1
0.5	0.1	bd	0.7	1.3	19.3	6.1	1	bd	0.1
0.5	0.1	bd	0.6	1.2	24	8.3	1	bd	0.1
1.5	0.3	bd	0.8	1.6	41.9	7.2	2.1	bd	0.1
2.3	0.5	0.1	0.4	3.3	40.8	1.4	4.2	0.1	0.1
0.7	0.1	0	0.1	0.9	47.1	2.5	1.2	bd	0.1
0.6	0.1	0	0.1	0.8	46.7	6.9	1.2	bd	0.1
0.6	0.2	0.1	0.2	1.1	47.4	9.6	3.5	bd	0.1
3.3	0.5	0.1	0.1	4	75	nd	4.3	bd	0.1
0.9	0.1	0.4	0.1	1.5	46.7	0.9	1.5	0.1	0
1.3	0.3	0.2	0.2	2.0	51.4	0.8	0.7	bd	0
0.7	0.1	0.3	0.7	1.8	36.7	2.9	1.0	bd	0
0.5	0.6	0.2	0.4	1.7	50.0	2.2	0.9	bd	0
6.4	2.2	0.2	1.0	9.8	52.7	1.8	7.5	bd	0
11.7	0.6	0.2	0.4	12.9	46.8	17.3	8.6	6.0	0
10.8	0.4	0.1	0.3	11.6	64.6	12.4	2.1	1.8	0
11.4	0.8	0.1	0.3	12.6	54.9	11.0	2.5	2.3	0
14.2	0.9	0.1	0.1	15.3	63.6	5.4	2.7	2.0	0
24.1	1.7	0.1	0.1	26.0	61.1	2.8	2.2	2.8	0
16.3	1.8	0.1	0.1	18.3	76.5	3.1	2.4	1.8	0
3.7	0.5	0.0	0.0	4.2	77.8	1.6	1.4	1.4	0

Table 12. (continuation)

Soil horizon	Horizon thickness (cm)	pH		Humus content, %	Hydrolytic acidity mg-equiv/100 g
		water	KCl		
Sphagnum-peat, gleyic soil, on lake deposition (key site L-2)					
Sphagnum debris	0–13	3.6	3.1	90.6	2.3
T1	13–19	4.1	3.3	85.2***	2.4
T2	19–37	4.4	3.3	89.9***	2.4
T2/G	37–52	4.1	3.2	23.8***	7.3
G	52–73	4.6	3.7	2.0***	6.0
BC/G	73.–96	5.0	3.9	0.9	4.6
C/G	96–180	5.1	4.1	0.2	2.7
Bryansk Region					
Podzolic, iron-illuvial, sandy, deep gleyic soil, on fluvioglacial deposition (key site Kl-1)					
OE	4–6	4.6	3.7	0.65	6.5
EB	6–19	4.8	4.0	0.51	3.6
Blf,h	19–34	5.1	4.2	0.27	2.9
B2	34–54	5.7	4.7	0.02	1.4
B3	54–73	5.9	4.8	bd	0.9
BCg	73–150	5.6	4.6	0.05	1.0
Cg	150–210	5.1	4.3	1.49	1.6
Podzolic, weakly stratified, sandy soil on fluvioglacial deposition (key site Zl-1)					
AE	6–23	5.2	4.2	0.41	3.2
B1	23–45	5.6	4.5	0.36	2.1
B2	45–71	5.8	4.8	bd	1.6
BC	71–110	6.1	5.0	0.06	0.8
C	110–200	5.8	4.7	0.34	1.4
Podzolic, iron-illuvial, sandy soil on double-layered deposition (key site NZ-1)					
AE	4–10	4.7	3.9	0.83	4.6
B1	10–18	4.7	4.1	0.37	3.8
B2f	18–38	5.0	4.5	0.1	1.7
B3	38–62	5.4	4.6	bd	1.1
BC	62–91	6.1	5.0	bd	0.7
Cg	91–130	6.1	4.9	bd	0.8
D	130–190	5.4	4.2	0.4	2.2

*No data
**Below detectable
***Loss at ignition (total organic carbon content)

Exchange cations				Total exchange capacity	Cation exchange capacity %	Available nutrient, mg/100 g			
Ca^{2+}	Mg^{2+}	K^{+}	Al^{3+}			P_2O_5	K_2O	NH_4	NO_3
mg-equiv/100 g									
2.4	0.3	0.2	0.5	3.4	55.8	2.8	6.5	bd	0
2.5	0.3	0.1	0.6	3.5	54.7	2.6	3.7	0.1	0
2.2	0.3	0.1	0.6	3.2	52.0	2.1	2.6	0.5	0
3.4	0.3	bd	1.0	4.7	33.6	7.3	1.8	1.0	0
1.3	0.2	bd	0.9	2.4	20.0	25.4	1.4	bd	0
1.7	0.2	bd	0.9	2.8	29.2	31.4	1.9	bd	0
2.1	0.3	0.1	0.6	3.1	48.1	11.6	2.6	bd	0
1.7	0.2	0.9	0.7	3.5	22.6	4.0	4.5	1.5	0.1
0.6	0.1	0.9	0,7	2.3	16.3	8.2	1.3	0.8	0.1
0,8	0.1	0.6	0.5	2.0	23.7	11.0	0.8	0.8	0.1
0.8	0.1	0.1	bd	1.0	39.1	12.0	0.7	0.8	0.1
0.8	0.1	0.1	bd	1.0	50.0	14.1	0.7	0.8	bd
0.8	0.1	0.2	0.1	1.2	47.4	10.5	1.0	0.8	bd
0.9	0.2	0.6	0.5	2.2	40.7	8.8	1.6	0.7	bd
1.2	0.1	0.5	0.4	2.2	28.9	8.7	2.6	0.7	bd
0.9	0.1	0.2	0.1	1.3	32.3	12.1	1.6	0.7	bd
0.8	0.1	0.1	0.1	1.1	36.0	9.3	1.1	0.6	bd
1.0	0.1	0.1	bd	1.2	57.9	8.9	1.1	0.8	bd
2.3	0.2	0.3	0.2	3.0	64.1	4.6	1.7	0.6	bd
0.6	0.1	1.1	1.0	2.8	13.2	11.7	2.6	0.8	0.1
0.4	0.1	0.9	0.0	2.3	11.6	11.8	1.6	0.5	0.1
0.3	0.1	0.5	0.4	1.3	19.0	11.3	0.9	0.5	0.1
0.5	0.1	0.3	0.2	1.1	35.3	8.7	1.3	0.6	0.1
0.7	0.2	0.1	bd	1.0	56.2	4.8	2.1	0.7	bd
1.2	0.2	0.1	bd	1.5	63.6	4.2	2.9	0.7	bd
3.2	0.8	0.9	0..7	5.6	64.5	2.6	4.9	0.9	bd

The properties of soil solutions in soddy-podzolic and podzolic soils are similar to those in peat-gley bog soils; however, podzolic soils have a much lower cation exchange capacity, as well as total and exchangeable acidity. The eluvial-illuvial vertical distribution pattern of CEC, nutrient content, and other physicochemical parameters (normally typical of podzolic soils) are weakly manifested in the case. Commonly, the soil profiles are characterised by a significant increase in the cation exchange capacity in the deep layers (below 100 cm), which is likely due to inherited lithological features.

Alluvial meadow-bog peat-gley soils are very different from all other soils in their physicochemical properties. A high humus content and clayey texture of alluvial parent rock predetermine their high adsorption capacity. These soils develop in backswamp areas of floodplains. They are rich in bases and nutrients. The proportion of Ca^{2+} and Mg^{2+} in the exchange complex exceeds 45%, and the cation exchange capacity varies from 17–26 mg-equiv/100 g (in upper layers) to 32 mg-equiv/100 g (in deeper layers). Both exchange and actual acidity are not high, and pH of soil solution increases with depth from 4.9 to 6.0. The total acidity is particularly high within the low-ash peat fragments. Unlike the mineral-peat horizon of bog soils T2G that also has a high total acidity, the investigated alluvial soils are not rich in exchangeable Al^{3+} and H^{+}. Their total acidity is almost completely determined by the dissociation of humus and other organic acids. Thus, these soils have a high buffer capacity, though they are less acid compared to bog soils. These features should be taken into account in the predictions of the behaviour of potentially mobile elements.

All investigated soils are poor in nutrients (Table 12): the phosphorus, potassium and nitrogen (nitrate/ammonium) content is of 1–9, 1.5–9, and 0.1–02/0.1–08 mg-equiv/100 g, respectively. The only exception is the upper horizon of alluvial meadow-bog soils, in which the nutrient content (especially, P and N) is an order of magnitude higher compared to the corresponding horizons of other soils. This is likely due to the addition of nutrients with alluvial sediments and an intensive biological cycle: available nutrients are intercepted by the roots and remain in the upper (root mat and humus) horizons. A similar, although not so well manifested, picture takes place in the bog soil where the accumulative horizons lie in the middle and lower parts of the soil profile.

Nutrient distribution down the soil profile in the investigated soils has its own features. Phosphorus tends to accumulate in the illuvial horizons of automorphic podzolic and soddy-podzolic soils, and in gley horizons of hydromorphic soils. Since phosphorus tends to be bound with sesquioxides, its accumulation may serve as an indirect index of the redistribution of sesquioxides down the soil profile.

Nitrogen is rather uniformly distributed down the soil profiles and tends to accumulate in organic, especially, peat horizons. This is likely due to a high intensity of microbiological processes (ammonification and nitrification) in these horizons. The potassium content is in close correlation with the content of fine mineral fractions. In terms of this study, potassium accumulation indicates the areas of potentially active irreversible caesium adsorp-

tion. High potassium content in forest litters and humus-accumulative horizons suggest that these horizons play an important geochemical role in the migration of both K and Cs.

Soils of Ukrainian Polesie (including the exclusion zone of ChNPP) are studied by the example of several soils that are most widespread in this region. Almost all of them develop on glaciofluvial deposits and are characterised by two characteristic features: (i) the low content of nutrients and bases, and (ii) low fertility. These soils may be pooled into two basic groups: automorphic and hydromorphic. The first group represented by soddy-podzolic soils; the second group includes peat-gley bog, meadow-bog, and alluvial-bog mucky gley soils.

The humus content in the upper horizons of automorphic soils does not exceed 5.2% (usually 1.5–2%, Table 13). In the undisturbed soils, the humus horizon lies immediately under the forest litter and is not deeper than 10–15 cm. The lower boundary of the humus horizon is not well manifested, and measurable amount of humus is detected in other soil horizons as well, which suggests a rather intensive vertical migration of the organic compounds.

Thus, the investigated automorphic forest soils below 10–15 cm are devoid of geochemical barrier preventing the potential vertical migration of radionuclides. Radionuclide replacement beyond this zone with time may intensify the radionuclide vertical migration and increase the soil-to-plant transfer.

In hydromorphic soils, the organic matter content may be as high as 15%, which is considerably higher than in the automorphic soils (Table 13). Peat formation is always accompanied with some release of mobile humus compounds that migrate deeper into the soil profile. That is why, the humus content declines very gradually with depth and is quite high down to the depth of the water table. A significant variation in the humus content down the soil profile is observed only in the alluvial soils, which is due to buried humus strata.

In fact, both hydromorphic and automorphic soil groups (except for the alluvial soils) possess only two geochemical barriers: forest litter and humus horizons. The latter is very thick in hydromorphic soils, where it may serve as an important obstacle for the radionuclide migration to deeper soil layers and (potentially) ground water. Abundant humus and clayey laminae in the profile of alluvial soils serve as an additional barriers for the radionuclide vertical migration.

Automorphic soils of the region have generally low cation exchange capacity (CEC) even in upper horizons (about 4–13 mg-equiv/100 g) (Table 13). Soil adsorption complex is not rich is bases: the content of exchangeable alkali and alkali earth metals is 14–32% (0.6–3.0 mg-equiv/100 g); the share of exchangeable aluminium is rather high. The pH of water and salt extracts is low, and the total acidity (particularly in upper horizons) is high (2.8–9.8 mg-equiv/100 g). All these indices are combined with low clay content, which results in the extremely low cation exchange capacity of automorphic soils, and promote radionuclide mobility in the soil and the soil-plant system [8, 133, 211].

Both CEC and exchangeable Ca content are much higher in the hydromorphic soils (20–30 and 8–16 mg-equiv/100 g, respectively). This is due to accumulative geochemical processes in hydromorphic soils. The content of alkali

Table 13. Chemical and physicochemical soil properties in the territory of Ukraine (30 km exclusion zone of CHNPP), mean values at n = 3

Soil horizon	Horizon thickness, cm	pH		Humus content, %	Hydrolytic acidity, mg-equiv/100 g
		water	KCl		
Kiev region (Ukraine)					
Soddy-podzolic, sandy, old-tilled soil on fluvioglacial deposition and brown-red moraine bedrock (key site D-5)					
A_{tilled}	0–35	4.5	4.1	1.71	4.1
B1	35–55	5.5	4.9	0.71	1.4
B2	55–69	5.6	5.1	0.53	0.6
BC	69–100	5.1	4.8	0.53	0.5
C	110–120	5.3	4.8	0.68	0.8
Secondary-podzolic, sandy soil on fluvioglacial deposition (key site K-2)					
AE	5–8	5.1	4.2	1.71	3.7
B1	8–14	5.1	4.3	0.81	2.4
B2	14–25	4.9	4.3	0.78	2.3
B_{relict}	25–42	4.8	4.5	0.43	1.3
BC	45–75	5.1	4.7	0.38	0.8
C	75–125	5.6	5.0	0.28	0.5
Soddy-podzolic, illuvial-humus, sandy soil on fluvioglacial deposition (key site D-1)					
A	6–11	4.6	4.7	1.29	2.8
B1h	11–24	4.8	4.4	0.91	2.6
B2	24–40	5.2	4.6	0.75	2.3
BC	40–70	5.2	4.7	0.75	1.1
C	70–150	5.8	5.2	0.46	0.7
Soddy-podzolic, weakly stratified sandy soil on fluvioglacial deposition (key site Sh-1)					
OA	4–6	4.4	3.4	5.21	9.8
AE	6–10	4.4	3.5	2.21	5.2
B1	10–23	5.4	4.5	0.81	2.2
B2	23–40	4.8	4.5	0.72	1.9
BC	40–70	4.8	4.6	0.46	1.5
C	70–110	4.7	4.9	0.53	1.0

Exchange cations			Total exchange capacity	Cation exchange capacity, %	Available nutrient, mg/100 g		
Ca^{2+}	Mg^{2+}	Al^{3+}			P_2O_5	K_2O	$NH_4 + NO_3$
mg-equiv/100 g							
1.6	0.4	0.4	2.4	32.8	6.9	5.8	7.9
0.8	0.3	0.1	1.2	44.0	1.8	2.3	1.6
0.6	0.3	0.0	0.9	60.0	0.6	1.7	0.5
0.6	0.3	0.0	0.9	64.3	0.4	1.2	0.6
0.7	0.3	0.1	1.1	55.6	0.4	1.3	0.6
0.4	0.2	0.4	1.0	13.9	3.7	4.1	3.5
0.5	0.1	0.4	1.0	20.0	3.9	2.2	0.9
0.4	0.1	0.4	0.9	17.9	3.7	1.5	0.7
0.3	0.1	0.3	0.7	23.5	3.2	1.0	0.4
0.3	0.1	0.2	0.6	33.3	2.2	0.9	0.3
0.6	0.2	bd*	0.8	61.5	3.1	1.4	0.4
0.5	0.2	0.3	1.0	20.0	1.3	3.1	6.8
0.4	0.2	0.5	1.1	18.7	8.8	2.3	2.5
0.3	0.1	0.2	0.6	14.8	7.5	2.7	1.2
0.4	0.1	0.1	0.6	31.2	2.9	1.7	0.7
0.3	0.1	0.0	0.4	36.4	2.4	1.3	0.5
2.5	0.5	0.8	3.8	23.4	2.4	6.9	4.9
0.9	0.3	1.0	2.2	18.7	1.7	4.1	2.4
1.0	0.2	0.3	1.5	35.3	2.9	2.0	0.9
0.5	0.1	0.3	0.9	24.0	3.3	1.2	0.6
0.1	0.1	0.2	0.4	11.8	4.3	1.1	0.3
0.2	0.1	0.2	0.5	23.1	2.0	0.9	0.3

Table 13. (continuation)

Soil horizon	Horizon thickness, cm	pH		Humus content, %	Hydrolytic acidity, mg-equiv/100 g
		water	KCl		
Meadow-bog, peat-gley sandy soil on fluvioglacial deposition (key site D-4)					
T	7–13	5.3	4.4	9.3	12.2
Ag	13–23	5.5	5.0	3.0	3.8
B1g	23–46	5.7	5.1	1.09	1.5
B2g	46–60	5.6	5.3	0.8	1.1
BCg	60–90	5.5	4.9	0.57	0.8
G	90–115	5.2	4.5	0.45	0.7
Alluvial-bog, clay-humus, gley soil on floodplain deposition (key site K-4)					
Ad	0–5	4.9	4.0	13.1	15.5
AT	5–25	4.6	3.9	11.73	16.4
Bg	25–60	5.3	4.2	1.98	3.9
G	60–70	5.7	4.8	1.4	1.0
Valley bog, peat-gley soil on fluvioglacial deposition (key site D-3)					
T	5–20	5.6	5.2	15.24	9.4
Ag	20–35	6.1	5.8	4.11	2.2
Bg	35–60	6.2	6.0	0.85	0.5
G	60–90	6.2	6.1	0.63	0.5

*Below detectable
**No data

and alkali-earth elements in the exchange complex is high (40–60% in the upper horizons and up to 80% in deep horizons).

The nitrogen content in most of the automorphic soils is low and correlates with the humus content (Table 13).

Soil provision with phosphorus and potassium is not very high (3–7 mg/100 g), which is normal for the investigated territory [95]. A low potassium and calcium content is a precondition for high transfer factors of ^{137}Cs and ^{90}Sr to plants.

On the contrary, the investigated hydromorphic soils have a rather high content of basic nutrients.

Exchange cations			Total exchange capacity	Cation exchange capacity, %	Available nutrient, mg/100 g		
Ca^{2+}	Mg^{2+}	Al^{3+}			P_2O_5	K_2O	$NH_4 + NO_3$
mg-equiv/100 g							
7.8	0.6	0.2	8.6	40.8	nd**	23.0	42.8
2.8	0.2	0.1	3.1	44.1	40.0	4.0	12.6
1.6	0.1	0.0	1.7	53.1	14.7	1.3	4.4
1.4	0.2	0.0	1.6	59.2	12.0	2.0	4.8
1.4	0.1	0.0	1.5	65.2	17.0	4.3	3.2
1.2	0.1	0.0	1.3	65.0	15.5	5.0	3.9
16.8	1.8	0.3	18.9	54.5	1.2	11.3	6.2
14.2	1.2	0.6	16.0	48.4	1.8	7.0	15.5
9.8	2.3	0.2	12.3	75.6	1.6	4.6	0.8
3.1	0.8	0.0	3.9	79.6	2.7	3.0	0.3
14.7	3.0	0.4	18.1	65.3	18.2	30.7	18.3
6.8	1.3	0.0	8.1	78.6	18.0	4.7	4.9
1.7	0.4	0.0	2.1	80.8	11.5	2.2	4.7
1.4	0.3	0.0	1.7	77.3	9.7	3.7	4.7

The potential impact of the humus state and physicochemical soil properties on strontium and caesium migration depends primarily on a high soil acidity, low content of exchangeable cations, and good drainage. The ability of these soils to absorb cations, including radionuclides, is low.

Soddy-podzolic and podzolic sandy automorphic soils are poor in fine particles and have very low reserves of humus. Thus, forest litter is the only geochemical barrier capable of accumulation and preventing intensive vertical and lateral migration of radionuclides. At the same time, forest litter provides all conditions for caesium mobilisation and its uptake by roots because of low pH and adsorption capacity of the organic matter for caesium.

2. IMPACT OF FOREST BIOGEOCENOSES ON THE INITIAL DISTRIBUTION OF RADIOACTIVE FALLOUT

2.1. INTERCEPTION OF RADIOACTIVE FALLOUT BY ARBOREAL VEGETATION

Arboreal vegetation is the first barrier on the way of radionuclide expansion in the fallout-polluted forest ecosystems. As the fallout occurs, the trees retain a significant proportion of the fallout particles, involve the radionuclides into the biological cycle, and prevent them from further migration [246]. Initial radionuclide pattern determines to a large extent the dose burden on the biological objects [119, 239]. Thus, determination of quantitative parameters of the initial distribution of radionuclides at the initial stages of migration is of top priority for radioecology.

A considerable portion of the radionuclides is reported to be intercepted by the tree crowns and retained for a while by the tree canopy. The retention coefficient varies from 20 to 100% depending on a wide range of factors, such as the species composition of the stand, relative crown projection, weather conditions and stand age (Table 14). The highest interception is observed in the coniferous forests upon normal (typical) weather conditions; in the case of deciduous forest, the interception of radionuclides gains maximum during the stage of maximum leaf development. On the average, the fallout proportion intercepted by the tree canopy is roughly the same as the relative crown projection cover. The only exception is deciduous forests during the leafless period, when the interception rate is about 3 times lower than the projective cover of tree crowns [8, 66, 235].

The second barrier on the way of the radioactive fallout is the understorey, including shrubs, herbaceous vegetation, mosses, and lichens. The retention capacity of the understorey depends also on the above-mentioned factors, but it is more species-dependent. In general, the interception capacity increases in the following sequence: herbaceous vegetation < lichens < mosses [8].

Another specific feature of the initial distribution of the fallout depending on forest ecosystems is the so-called "marginal effect": a higher contamination of the tree crowns adjacent to the forest margin at the windward side relative to the source of contamination. This phenomenon was first described in the forests contaminated due to the Kyshtym accident [234]. Thus, the clearances in the windless "shadow" are shielded from the fallout.

The Chernobyl-derived fallout was considerably different from the fallout generated by any other radiological accident known. The continuous release (about 11 days) occurred in the middle spring (April 26) and coincided with active growth processes. This caused the maximum unfavourable direct effect of irradiation on the stand [101, 119, 224]. The trees retained 60 to 90% of the fallout. The retention was most manifested

Table 14. The coefficients of radionuclide retention in the tree canopy depending on the climatic zone, age, and type of fallout [8]

Object	Type of fallout	Coefficient of retention, %
Young pine forest (6–10 years old), crown projection = 1.0	Experimental spraying of the tree crowns by water solution of ^{89}Sr	90–100
Old pine forest (60 years old), crown projection = 0.9	Fallout particles < 50 μm	80–100
Mature pine forest (25 years old), crown projection = 0.8	Fallout particles < 100 μm	70–90
Mature pine forest (30 years old), crown projection = 0.8	Secondary fallout (soil/fallout particles resuspended from the soil by wind)	40–60
Winter birch forest (40 years old), crown projection = 0.8	Secondary fallout (soil/fallout particles resuspended from the soil by wind)	20–25
Summer birch forest (35–40 years old), crown projection = 0.8	Global (weapon) fallout	20–60
Old pine forest (50–60 years old), crown projection = 1.0	Global (weapon) fallout	50–90
Tropical rainforest (Puerto-Rico, Central America)	Global (weapon) fallout	100

in the coniferous stand, while the deciduous species exhibited 1.5–3 times lower retention capacity because they were leafless at the time of intensive fallout [283].

Within the tree crowns, the maximum radionuclide content was found in the leaves; a much lower content of radionuclides was found in the bark and branches, and only a negligible amount directly penetrated into the wood through the leaf and bark surface (Table 15).

Immediately after the fallout, the contaminated tree crowns became a source of secondary contamination for the neighbouring territories due to the radionuclide transport by wind. This is confirmed by the extreme variation of radionuclide concentration in the atmosphere of the exclusion zone during several weeks after the accident [34, 165, 218].

The marginal effect is irregularly manifested in the exclusion zone. A considerable increase in the deposition was observed in the forest margins on the southern radioactive trace (Table 16); however, it was less characteristic of the western trace. This effect is practically insignificant in the remote territories (tens and hundreds of kilometres from ChNPP) where the fallout was mainly composed of very fine particles, and the precipitation depended on the atmospheric turbulence and rains rather than on the wind

Table 15. Radionuclide distribution in the tree canopy, %
(the exclusion zone, June of 1986, average of n = 12)

Needles (leaves)	Barked wood	Bark total (cork + alive bark "bast")	Branches		Total, %
			small	large	
49.3	1.8	29.7	11.9	7.3	100

Table 16. Spatial distribution of the deposition at the forest margin
(κBq/m^2, 1987)

Location	Observation point		
	Meadow	Forest margin	Forest
6 km to the south of ChNPP	14,210	19,910	17,200
25 km to the south of ChNPP	1,070	No data	1,300

direction [348]. As a rule, the marginal effect is most pronounced in the 20 to 50 m wide (sometimes, up to 200–330 m) shelter belts of small forest groves faced toward the accidental unit (Fig. 5) [177]. Somewhat higher deposition was observed around single trees standing in the treeless territories [228].

In general, the forests received at least 20–30% more radioactive fallout compared to the treeless areas, although some researchers believe this difference to be even higher [26, 45, 123, 214, 295, 296, 341]. This phenomenon is in a good agreement with the features of the air mass transfer and rainfall distribution over the forested areas in the warm period. According to long-term observations, the

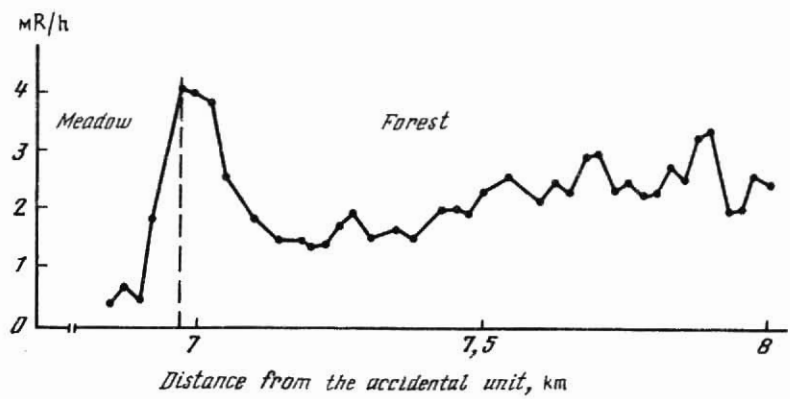

Fig. 5. Variation of the dose rate in the forest-meadow edge

forest areas receive 15–30% more rainfall compared to the treeless areas in the same region [253]. Some published data do not confirm the phenomenon of a higher deposition under the forest cover [49], but the increase in the rainfall is a statistically reliable fact, as well as a close correlation between the rainfall and deposition.

2.2. EARLY STAGES OF THE RADIONUCLIDE MIGRATION IN FOREST ECOSYSTEMS

The adhesion of the fallout particles to the plant surface immediately after the precipitation is very weak. They are easily blown away by wind and washed off by rains [8]. The residence time of radionuclides in tree crowns was shown to be much shorter than that in the other components of the trees [291].

Since only surface components of the tree were exposed to the initial fallout, the physicochemical forms of the fallout were the most influential factor of the radionuclide migration in the tree canopy. The maximum intensity of the natural decontamination processes running in the forest was observed in the near zone (less than 10 km from ChNPP) where the fallout particles were the largest. This fact gives us grounds to state that the particle size is a factor determining the radionuclide half-stay in the tree crown, which was shown in the earlier studies [243, 354]. The actual mechanism of this phenomenon is not completely clear, however.

The active growth processes during and after the accident were the factors of relatively fast natural decontamination of the tree crowns from the radioactive particles. Radionuclides were actively removed in the course of bark desquamation; buds, leaf, and twig development, etc. The dynamics of the radionuclide removal from the tree crowns in the "near zone" was complicated by the continuous character of the fallout (11 days) [99, 210]. According to our estimations, when the stand is not affected by acute irradiation (i.e., when direct irradiation effect is negligible), the effective half-stay of the radionuclides in the contaminated stand varies from 3 weeks to one month [234, 239].

The natural decontamination processes were most intensive during the first months after the accident and depended primarily on the season, particle size, weather conditions, and type of biogeocenosis. By August of 1986, the tree crowns contained no more than 10–15% of the initial deposition (Fig. 6). Variation in this value depended also on the species composition and stand parameters. For example, birch dominance in the species composition increased the radionuclide content in the arboreal vegetation up to 18% of the total deposition (data of August, 1986, Table 17).

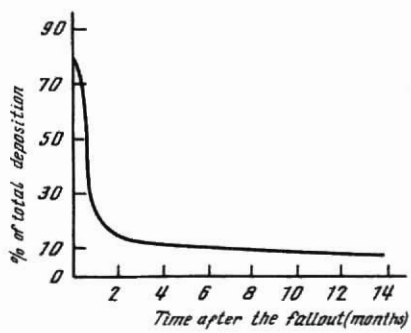

Fig. 6. The dynamics of natural decontamination processes in the tree canopy during the first year after the accident

Table 17. Radionuclide distribution among basic components of forest biogeocenoses in the exclusion zone, % (August of 1986)

Wood	Bark	Needles (leaves)	Branches		Total in vegetation	Forest litter	Mineral layers	Total in soil
			small	large				
Mixed birch/pine/oak forest								
0.1	3.3	1.6	6.5	1.1	12.7	86.4	0.9	87.3
Mixed oak/birch (+pine/aspen) forest								
0.2	3.7	9.5	3.2	1.2	17.6	80.5	1.9	82.4
Pine/birch/alder (+oak) forest								
0.1	2.1	7.2	1.4	0.9	11.7	87.3	1.0	88.3

Table 18. Radionuclide content in the components of dominant tree species in the exclusion zone (kBq/kg, August of 1986)

Species	Total contamination, kBq/kg (fresh weight)				
	Needles (leaves)	Barked wood	Bark (cork + + bast)	Branches	
				small	large
Pine	185.0	0.37	129.5	225.7	29.6
Birch	392.2	0.74	37.0	66.6	25.9
Oak	48.1	1.48	66.6	155.4	22.2
Aspen	11.1	0.37	103.6	14.8	7.4
Alder	296.0	0.74	29.6	48.1	18.5

The rate of natural decontamination may be ranked depending on tree species as follows: aspen > oak > pine > alder > birch. At the same time, the contaminant dynamics was different for various structural components, such as leaves (needles) or bark. (Table 18).

The differences between the contamination dynamics in the assimilative organs and bark are due to (i) species-dependent peculiarities (leaf surface and its adhesive properties, microstructure of the leaf and bark surface, etc.) [98, 243]; (ii) the intensity of the radionuclide redistribution among the crown and other tree components; and (iii) the intensity of the radionuclide redistribution within the inner tree tissues.

The radionuclide redistribution down the tree stem results in a somewhat higher contamination of the stem middle part and, particularly, tree butt compared to its top. The rate of the redistribution depends, apparently, on the bark features and is most manifested in the pine stand. Our data suggest that pine

Table 19. Radionuclide distribution among the components of dominant tree species in the exclusion zone, % (August of 1986)

Species	Needles (leaves)	Barked wood	Bark (cork + bast)	Branches		Total
				small	large	
Pine	27.7	0.9	46.6	15.4	9.4	100
Birch	59.5	1.3	12.6	15.3	11.3	100
Oak	9.3	5.6	60.1	6.2	18.8	100
Aspen	3.7	1.1	86.2	3	6	100
Alder	66.5	1.9	17.2	6.1	8.3	100

bark has the least retention capacity of all investigated species. On the contrary, the least radionuclide redistribution (and the highest retention capacity) was observed in the alder and oak stands [45].

During the first months after the accident, the tree parts and organs directly exposed to the fallout (bark, needles, and leaves) remained the most contaminated components of the stand. The contribution of debarked wood did not exceed 1% of the total radionuclide pool in tree canopies, in spite of a high wood biomass (Table 19).

In brief, intensive mechanical replacement of the fallout particles from the stand canopy to the understorey and soil surface took place during the first months after the accident (until August of 1986). At this stage, the radionuclide content in the tree canopy was fully determined by the surface contamination, and the radionuclide composition of the stand components was the same as the fallout (decay corrected). The contamination dynamics in different BGC was generally the same. We consider this period as the first stage of the long-term radionuclide migration in the ecosystem (Figs. 6, 7).

The second stage (from autumn of 1986 to 1988/89) is characterised by active natural decontamination and initial increase in the radionuclide root uptake (Fig. 7). The natural decontamination in this period is represented by a biologically-induced replacement of tree structures (annual litterfall, bark desquamation, etc.) Gradual increase in the root uptake is indicated by some increase in the proportion of ^{137}Cs and ^{90}Sr in the decay-corrected radionuclide composition of the vegetation [277].

The third stage (from 1989 up to now) is characterised by a clear dominance of the root uptake and gradual approximation to a quasi equilibrium in the soil-plant system. The radionuclide fate during this period depends to a large extent on the ecosystem and landscape features. In the automorphic soils of eluvial landscapes, the radionuclide availability for plants decreases, which results in a gradual decrease in the share of biota-fixed radionuclides in the total deposition.

In the hydromorphic soils of accumulative landscapes, radiocaesium tends to accumulate in the vegetation due to active root uptake, which results in a gradual increase in the share of biota-fixed radionuclides.

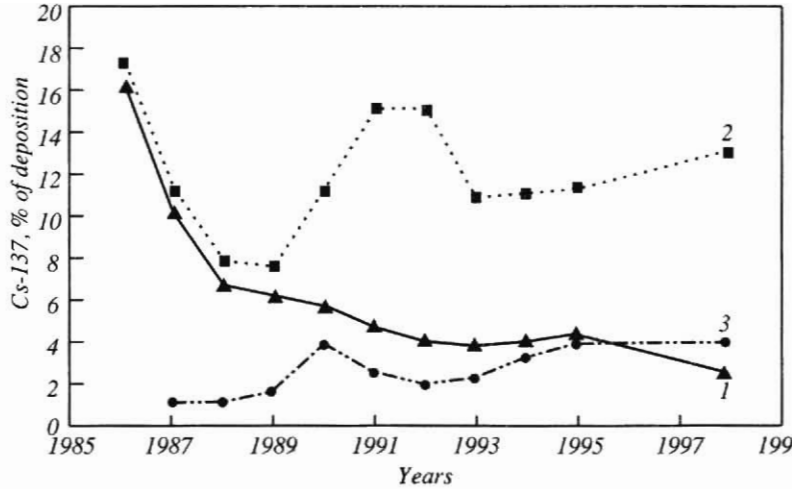

Fig. 7. The dynamics of ^{137}Cs content in the tree canopy:
(1) mixed, broad-leaved-pine forests, eluvial landscape; (2) alder forest, accumulative landscape; (3) mixed, broad-leaved-pine forests, eluvial landscape, "near zone"

A more complex radionuclide dynamics is characteristic of the territory adjacent to ChNPP that has been exposed to the low-dispersed fallout enriched with the insoluble fuel component ("near zone"). The radionuclide fate in this zone depends on the processes of particle disintegration, dissolution, and radionuclide release and interaction with soil.

3. RADIONUCLIDE UPTAKE BY FOREST VEGETATION AND FUNGI

There is a wide range of publications presenting an extended information on radionuclide accumulation by various structural tree organs due to global (weapon) fallout and nuclear accidents [8, 132, 145, 159, 201, 232, 235, 237, 248, 339, 350,]. The published data are extremely variable and sometimes even contradictory because of numerous factors affecting the processes of radionuclide uptake, such as the character of fallout, deposition, growth conditions, species, etc. [98, 128, 146, 150, 161, 175, 219, 232, 241, 247, 249, 274, 277, 280]. The data on "weapon" fallout are not fully applicable to the analysis and forecast of the radiological situation in forest ecosystems contaminated due to the Chernobyl accident. The most comprehensive and detailed data on radionuclide behaviour in forest environments are dedicated primarily to ^{90}Sr, whereas the Chernobyl-derived fallout is mainly represented by ^{137}Cs. In addition, the physicochemical properties of the Chernobyl-born fallout are very different from those of the Kyshtym and "weapon" fallouts, and the environmental conditions in the forest area severely contaminated due to the Chernobyl accident (the exclusion zone) are very specific. A range of papers and monographs regarding the fate of the Chernobyl-derived radionuclides in the environment (including natural ecosystems and forests) were published in the recent decade [69, 98, 119, 120, 175, 267]. Nevertheless, data on the quantitative parameters of radionuclide biogeochemical migration and cycling in various forest environments are still far incomplete and insufficient to perform a reliable forecast of the radioecological situation in the contaminated territories.

3.1 RADIONUCLIDES IN THE ARBOREAL VEGETATION

3.1.1 Radionuclide Composition and Its Dynamics

The radionuclide composition and its dynamics in various tree organs and structural parts depend on numerous factors: physical half-life of the radionuclide, initial composition of the fallout, species composition of the vegetation, and type of a structural component of the contaminated tree.

At the initial stage of aerial contamination, the radionuclide composition of external (exposed to the fallout) tree components and organs is identical to the decay-corrected composition of the initial fallout (Table 20).

The latter was represented by a wide spectrum of fission and activation products, such as ^{137}Cs, ^{90}Sr ^{106}Ru, ^{95}Nb, ^{95}Zr, etc. In 1986, inner structural

Table 20. Average radionuclide composition of forest vegetation in the European part of CIS (June, 1986)

Radionuclide	Radionuclide composition, %		Radionuclide	Radionuclide composition, %	
	30 km exclusion zone (average) [16]	Russian Federation [167]		30 km exclusion zone (average) [16]	Russian Federation [167]
^{95}Zr, ^{95}Nb	28.8	2	^{137}Ca, ^{137}Ba	3.3	25
^{103}Ru, ^{103}Rh	17	No data	^{140}Ba, ^{140}La	2	7.5
^{106}Ru, ^{106}Rh	9.2	27.0	^{140}Ce	2	7.5
^{131}J	<1.8	23.3	^{141}Ce	8	No data
^{134}Cs	0.9	13	^{144}Ce, ^{144}Pr	30	2.2

components of the vegetation (not directly exposed to the fallout) contained a negligible amount of Chernobyl-derived caesium isotopes only. We believe that this early contamination of the inner tissues was due to direct adsorption of caesium by leaf surfaces [277].

The further dynamics of the radionuclide composition varied depending on the tree component (Fig. 8–10). The external organs were characterised by a monotonous decrease in the proportion of short-lived radionuclides and the corresponding increase in the proportion of long-lived isotopes, such as ^{90}Sr and $^{134,137}Cs$. As a result, such radionuclides as ^{95}Nb, and ^{95}Zr (half-life period of 35 and 64 days) were practically undetectable in the territory of the Russian Federation and in the exclusion zone since 1987 and 1989, respectively. The corresponding "expiring time" for ^{144}Ce and ^{106}Ru (half-life period of 284 and 367 days) is 2–3 and 5–6 years, respectively. The changes in the radionuclide composition run particularly fast in the intensively growing tree components, such as assimilative organs and small branches. In a very short period, the contamination of these organs became almost exclusively dependent on caesium and strontium isotopes. The longest presence of the short-lived radionuclides (5–6 years) was registered in the external bark (cork). The inner tree components (wood and internal bark), from the very beginning, contained exclusively ^{137}Cs, ^{134}Cs, and ^{90}Sr .

Thus, 2–3 years after the fallout, ^{137}Cs (half-life of 30.2 years) and ^{90}Sr (half-life of 27.7 years) became the only significant radionuclides in the tree canopy.

The composition of radionuclides retained in the tree canopy depends, to a large extent, on the dispersion of the fallout particles. Highly dispersed aerosols are better sorbed by the needles and internal bark, which prolongs the stay period of the initial fallout in the external components of the stand. The "external" origin of some portion of the contamination is indicated by the presence of ^{144}Ce in the radionuclide composition. In the territories exposed to the low-dispersed fallout (near zone), large particles are less retained in the tree crowns, which

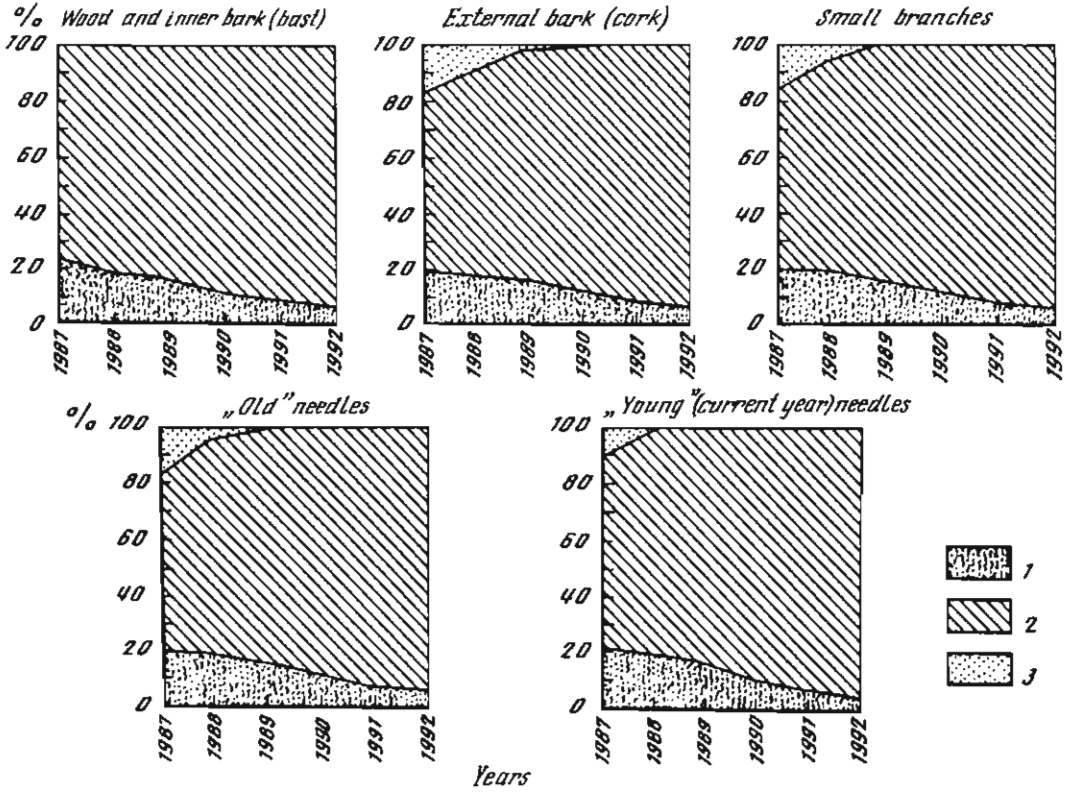

Fig. 8. The dynamics of the radionuclide composition of various tree organs and components (Tula Region, Russian Federation):

(1) ^{134}Cs; (2) ^{137}Cs; (3) ^{106}Ru

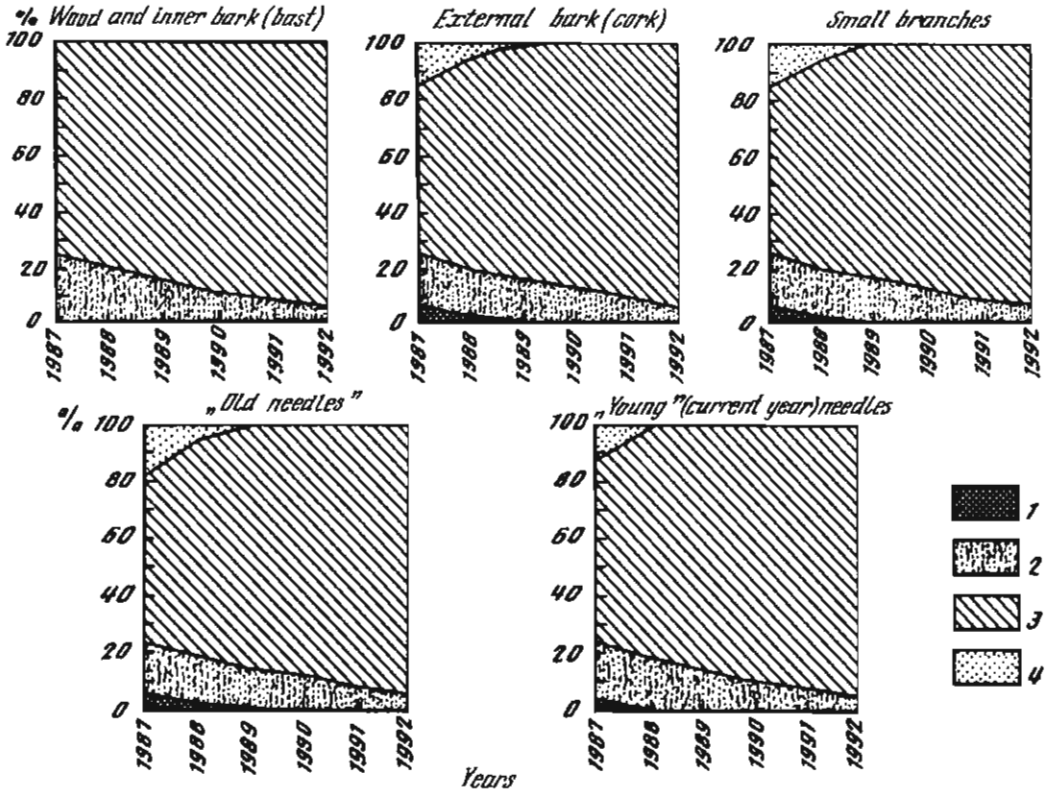

Fig. 9. The dynamics of the radionuclide composition of various tree organs and components (Bryansk Region, Russian Federation):

(1) ^{144}Ce; (2) ^{134}Cs; (3) ^{137}Cs ; (4) $10^{6}Ru$

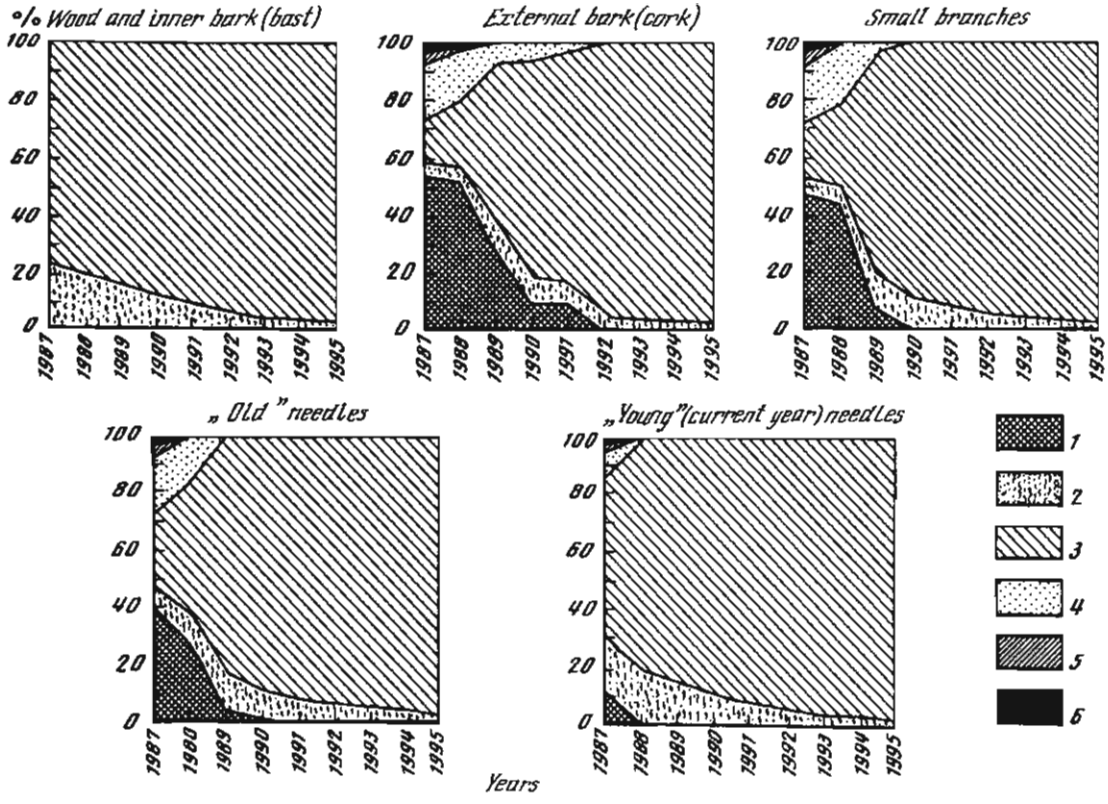

Fig. 10. The dynamics of the radionuclide composition of various tree organs and components:

(1) ^{144}Ce; (2) ^{134}Cs; (3) ^{137}Cs; (4) ^{106}Ru; (5) ^{95}Nb; (6) ^{95}Zr (30 km exclusion zone)

results in a relatively fast surface decontamination and enrichment of the radionuclide composition by ^{137}Cs . The same processes of surface decontamination determine the effect of tree species on the radionuclide composition dynamics. The longest stay of ^{144}Ce as an indicator of the external contamination is typical of oak and alder bark. Pine and birch bark sampled from a tree butt also contained this radionuclide. Pine needles hold the fallout for 2–3 years, i.e., up to their death and removal.

The dynamics of radionuclide composition depends also on the peculiarities of ^{137}Cs and ^{90}Sr root uptake, on one hand, and natural surface decontamination (mechanical and biological), on the other hand. The decontamination processes run faster in the near zone (see above), while the rate of the root uptake in this zone increases in time monotonously. Therefore, the radionuclide composition of the external organs in the near zone is enriched in ^{137}Cs and ^{90}Sr faster compared to the remote contaminated territories.

Thus, the dynamics of the radionuclide composition in the tree canopy depends on (i) radionuclide half-life, (ii) rate of the natural decontamination, and (iii) rate of the root uptake. In the case of the Chernobyl fallout, ^{137}Cs and ^{90}Sr become the most significant radionuclides in the tree canopy 2–3 years after the fallout (in the near zone) or 1–2 years after the fallout (in the remote contaminated territories).

3.1.2. Distribution of ^{137}Cs and ^{90}Sr Among Tree Organs and Structural Parts

The absolute radionuclide content (Bq/kg) and deposition (Bq/m^2) in the tree components vary by 3–4 orders of magnitude in the contaminated area. Therefore, absolute units are inapplicable to analyse ^{137}Cs and ^{90}Sr distribution among the stand components. The use of the so-called relative distribution ranges is more convenient and makes it possible to compare large samples over the entire contaminated territory (Table 21).

In general, ^{137}Cs content in the tree components sampled in the same site decreases in the range as follows: external bark > assimilative organs > branches > wood. However, the radionuclide content in the same components sampled in various sites depends on a set of factors, such as tree species, type of fallout, soil-ecological (environmental) conditions, and time after the accident.

At the stage of direct aerial contamination (1986), the maximum concentration of ^{137}Cs in all tree species was attributed to assimilative organs and small branches (Table 21, Fig. 11). The minimum specific activity was characteristic of the inner (alive) bark and wood [226].

The radionuclide content in the assimilative organs of various species increases in the following sequence: birch > alder > aspen > pine > oak, which is determined by (i) the specific shape and microtopography of leaves or adhesive charges on the needle surface [98, 243], and (ii) continuous processes of the leaf growth at the time of fallout (the leaf area of oak and aspen was not completely developed at the time of the Chernobyl accident, which reduced the initial contamination and particle stay). Lesser contamination of leaves has resulted in a higher contamination of other structures of these species: the maximum

Table 21. Comparative series of ^{137}Cs content in various tree organs and components (eluvial, automorphic landscapes, 1986, 1992)

Area	Species	Comparative series (see footnote)
	By the data of 1986	
30 km exclusion zone	*Pine*	SB> N> EB> LB> IB> W
	Birch	L> SB> EB> LB> IB> W
	Oak	SB> EB> LB> L> IB> W
	Aspen	L> SB> EB> LB> IB> W
	Alder	L> SB> EB> LB> IB> W
	By the data of 1992	
30 km exclusion zone		EB > N> IB> SB> LB> W
Bryansk region, RF	*Pine*	N > EB> EB> SB> LB> W
Kaluga region, RF		N > EB> IB> SB> LB> W
Tula region, RF		EB > N> IB> SB> LB> W
30 km exclusion zone		EB > L> SB> LB> IB> W
Bryansk region, RF	*Birch*	EB > L> SB> LB> IB> W
Kaluga region, RF		EB > L> SB> LB> IB> W
Tula region, RF		EB > L> SB> LB> IB> W
30 km exclusion zone		EB > L> LB> IB> SB> W
Bryansk region, RF	*Oak*	EB > L> LB> IB> SB> W
Kaluga region, RF		EB > LB> L> IB> SB> W
Tula region, RF		EB > LB> L> IB> SB> W
30-km exclusion zone		EB> L> IB> SB> LB> W
Bryansk region, RF	*Aspen*	EB> L> IB> SB> LB> W
Kaluga region, RF		EB> L> IB> SB> LB> W
30 km exclusion zone	*Alder*	EB> L> SB> IB> LB> W
Kaluga region, RF		EB> L> SB> IB> LB> W

Footnote: N – needles of current year; L – leaves; EB – external bark (cork); IB – inner bark (bast); LB – large branches; SB – small branches; W – wood

radionuclide concentration in oak and aspen was attributed to the external bark and small branches. The same components were the least contaminated in the alder and birch stands in 1996

Two months after the fallout, leaves were the most contaminated components in the birch and alder stand. Small branches and the external bark during this period served as the main sinks for radionuclides in the pine and oak (and aspen) stands, respectively. The character of the initial radionuclide retention, apparently, affects the intensity of radionuclide penetration into the plant tissues (surface uptake), i.e., may cause the early contamination of wood and other tissues that have not directly been exposed to the fallout.

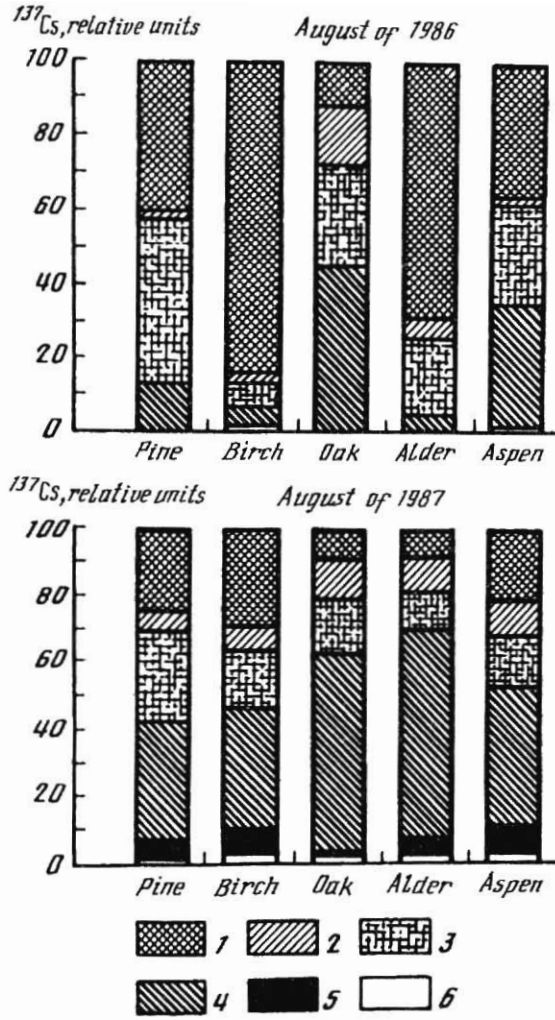

Fig. 11. Relative content of ^{137}Cs in the organs and components of various tree species (30 km exclusion zone, Ukraine):

(1) leaves and needles; (2) large branches; (3) small branches; (4) external bark (cork); (5) inner bark (bast); (6) wood

Morphological and physiological features of tree species affect, though to a smaller extent, the radionuclide content in the stand component at later stages as well. For example, one year after the fallout, the external bark becomes the most contaminated tree component (Fig. 11). Since the radionuclide concentration in the bark depends on its surface structure, the tree species with a rough or adhesive bark, such as oak, aspen, alder, and some others species, tend to accumulate more radionuclides [8, 306]. Smooth (birch) or exfoliating (spruce, pine) bark is characterised by a lower content of radionuclides.

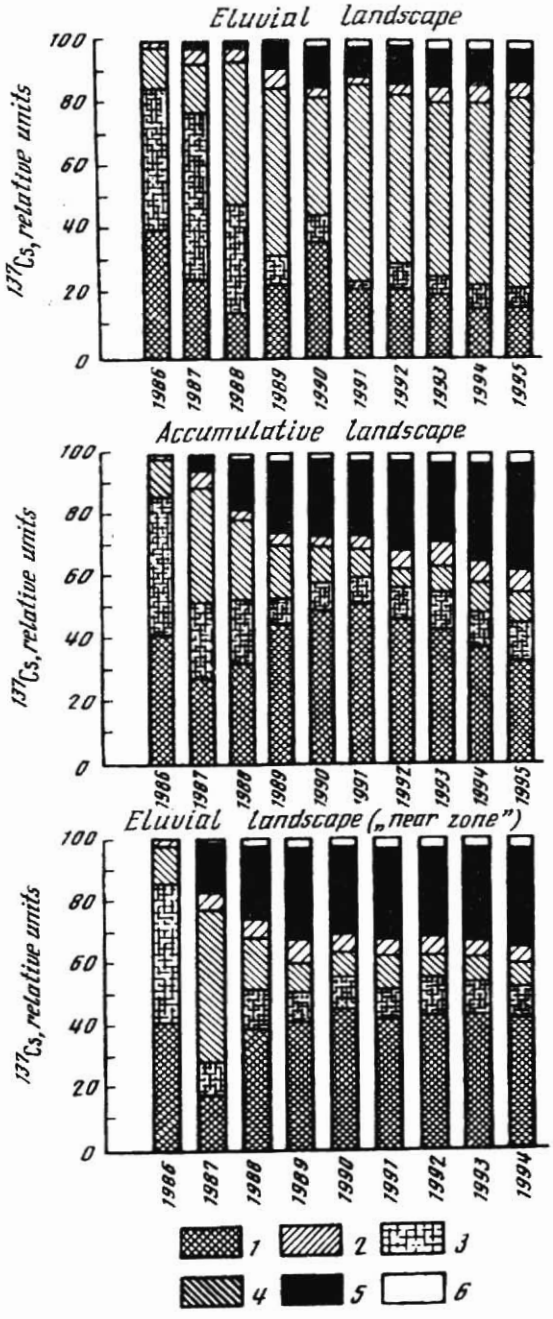

Fig. 12. ^{137}Cs dynamics in various pine organs and components (30 km exclusion zone):

(1) leaves and needles; (2) large branches; (3) small branches; (4) external bark (cork); (5) inner bark (bast); (6) wood

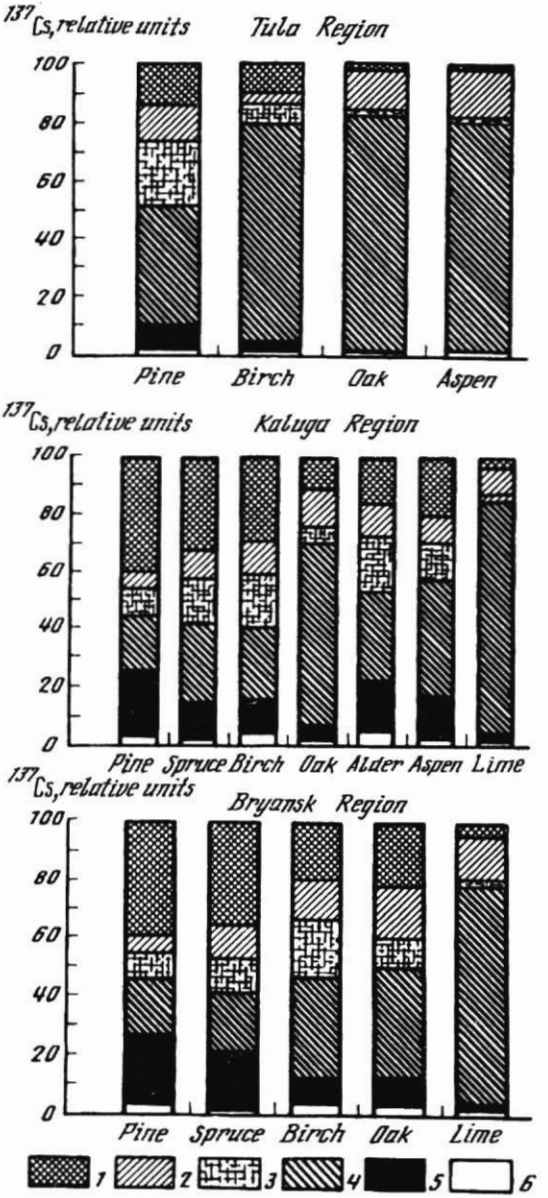

Fig. 13. ^{137}Cs content in the organs and components of various tree species (Russian Federation, 1992):

(1) leaves and needles; (2) large branches; (3) small branches; (4) external bark (cork); (5) inner bark (bast); (6) wood

Three years after the fallout, the root uptake becomes considerably more manifested. In this period, ^{137}Cs distribution among the stand components depends on the competitive processes of (i) root uptake and (ii) external natural decontamination. Both processes depend more or less on a number of environmental parameters, soil properties, physicochemical properties of the initial fallout, and species.

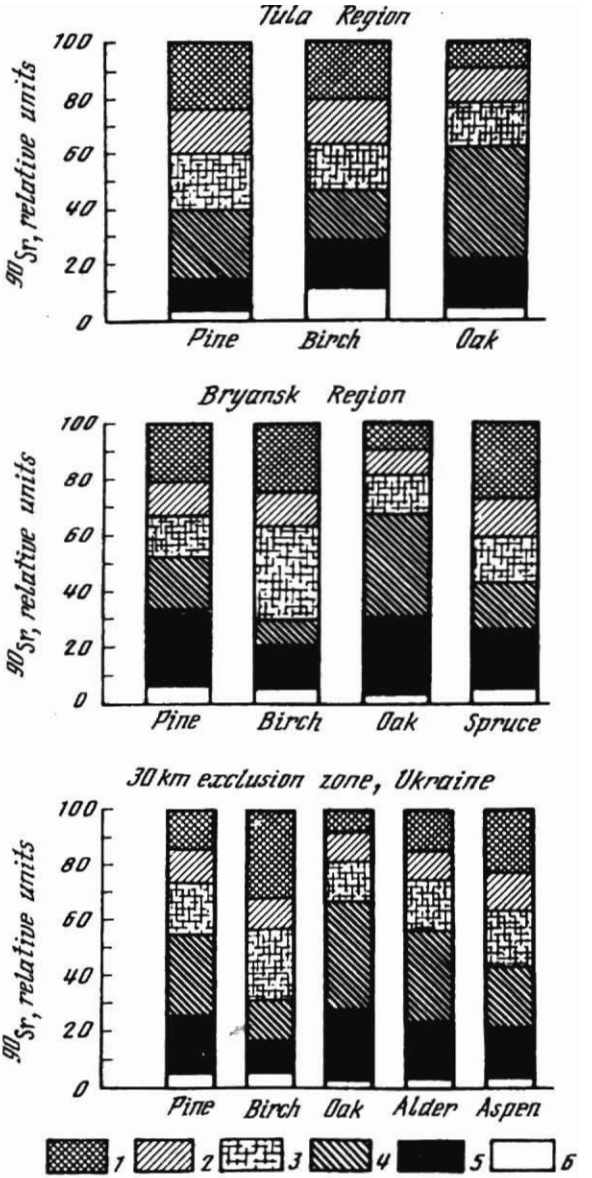

Fig. 14. ^{90}Sr content in the organs and components of various tree species (Russian Federation and Ukraine, 1992):

(1) leaves and needles; (2) large branches; (3) small branches; (4) external bark (cork); (5) inner bark (bast); (6) wood

In some territories, the radionuclides in the root-abundant zone remain available for quite a long time. These are either the areas covered by hydromorphic and semi-hydromorphic soils or the so-called "near zone" exposed to the low-soluble fallout enriched with the fuel component. The assimilative organs and inner bark are the most contaminated tree components in these territories (Fig. 12). On the contrary, the external bark remains the most contaminated tree component in automorphic soils and in the areas with lower radionuclide availability and root

Table 22. Comparative series of ^{90}Sr content in the tree organs and components (eluvial, automorphic landscapes, 1992)

Region	Species	Comparative series (see footnote)
30 km exclusion zone		IB> EB> SB> LB> ON > YN> W> C
Bryansk region, RF	Pine	IB> EB> SB> LB> ON> YN> W> C
Tula region, RF		EB> SB> LB> ON> IB> YN> W> C
30 km exclusion zone		L> SB> EB> LB> IB> W
Bryansk region, RF	Birch	L> SB> IB> LB> EB> W
Tula region, RF		L> EB> IB> SB> LB> W
30 km exclusion zone		EB> IB> SB> LB> L> W
Bryansk region, RF	Oak	EB> IB> SB> LB> L> W
Tula region, RF		EB> IB> SB> LB> L> W
Bryansk region, RF	Spruce	IB> EB> SB> LB> ON> YN> W> C
30 km exclusion zone	Aspen	EB> L> SB> LB> IB> W
30 km exclusion zone	Alder	EB> IB> SB> L> LB> W

Footnote: YN – "young" needles (of current year); ON – "old" needles; L – leaves; EB – external bark (cork); IB – inner bark (bast); LB – large branches; SB – small branches; W – wood; C – cones

uptake (Fig. 12). This phenomenon is manifested mostly on clayey chernozems (Tula region, the Russian Federation) where ^{137}Cs is the least available [133, 233]. The radionuclide content in the external bark of all tree species on chernozems is several times higher than in the other organs and tree components (Fig. 13). This is likely due to slower natural decontamination of the stand exposed to the fine aerosol fallout in the "remote area" compared to the "near zone".

At the stage of the dominant root uptake, the investigated tree species may be pooled into two main groups by the features of ^{137}Cs pattern in the tree components and organs. The first group includes coniferous species (pine and spruce) with the maximum radionuclide content attributed to the assimilative organs and internal bark, and the minimum radionuclide content attributed to the wood and external bark (Fig. 13). On the opposite, deciduous species tend to "conserve" more ^{137}Cs in the external bark and branches. Lime is a very specific species that does not belong to any of the above-mentioned groups: ^{137}Cs is virtually absent in the wood and internal bark and concentrated in the external bark of lime trees. This is likely due to the combined effect of (1) extreme ability of lime bark for fallout interception [134] and (2) low root uptake, since this species grows usually on automorphic soils with high fertility.

The process of ^{137}Cs redistribution among the stand components reaches its quasi-equilibrium (steady state) by the 5–6th year after the fallout. The radiocaesium pattern in the stand is expected to remain more or less stable for quite a while, although the absolute radionuclide content in the components varies in time and space depending on the long-term accumulation of ^{137}Cs and ^{90}Sr by plants.

The accumulation of another biologically important radionuclide ^{90}Sr in the stand shortly after the fallout is much similar to ^{137}Cs and determined by the same factors. However, ^{90}Sr distribution among the tree components becomes more specific in the course of time (Fig. 14, Table 22).

In general, ^{90}Sr is more uniformly distributed among the components and, unlike ^{137}Cs, ^{90}Sr does not exhibit the absolute dominance in any organ, although some tree components are enriched with this radionuclide. The latter is well illustrated by ^{90}Sr behaviour in the external bark. A number of species, such as birch, pine, and spruce, have a rather moderate ^{90}Sr proportion in the external bark compared to other tree organs and components, irrespective of landscape or soil conditions. A specific feature of ^{90}Sr distribution in a tree at the third stage (dominant root uptake) is its higher proportion in the wood and internal bark than in the external bark. This is particularly typical of chernozem soils in the remote contaminated territory (300–400 km from ChNPP). The root uptake of ^{90}Sr is apparently higher than that of ^{137}Cs and, on the contrary, the soil factors are less influential on ^{90}Sr availability compared to ^{137}Cs.

Radial Distribution of ^{137}Cs and ^{90}Sr in the Stem Wood

Using the radiocarbon and tritium methods, we have determined that these radionuclides and their stable isotopic carriers are incorporated into the tree tissues and insignificantly depend on the metabolic and exchange processes in the tree [35]. ^{90}Sr and ^{137}Cs are also incorporated into the tree tissues, along with their non-isotopic carriers Ca and K. ^{137}Cs and ^{90}Sr content in the wood is believed to vary by both stem radius (tree rings) and the height of a tree (from tree butt toward the top) [8, 98, 251, 347], but the data are often contradictory*.

The tree ring analysis has revealed that the radionuclide content in the stem wood varies as follows: (i) the highest concentration of radionuclides is observed in the external (current year) ring, which is apparently due to the most intensive growth processes and active radionuclide and nutrient transport to these tissues. Every next year, the peak of radionuclide concentration is replaced to the next current ring (Fig. 15). The "current year ring" is most contaminated in the tree butt where the alive bark is also most contaminated.

The Chernobyl-derived radionuclides present, though in much lower concentration, in the other tree rings, including those formed before the accident. This may be a side effect of the so-called "acropetal migration" (nutrient flow from the leaves toward roots) or diffusion-like processes through resin canals (in pine wood) following the radial (vertical) radionuclide gradient in the stem. The most significant increase in the concentration of ^{137}Cs and, especially, ^{90}Sr is attributed to the rings adjacent to the central (nucleus) stem wood. This phenomenon is manifested in the tree butt as well (Fig. 15, 16).

Radial distribution of radionuclides in the stem wood depends primarily on the wood structure. The nucleus bearing species (pine and particularly oak) exhibit more manifested differences between the radionuclide concentration in central and marginal wood. For other species (birch, alder, etc.), the decrease in radionuclide concentration in the pre-accidental wood is also observable, though less manifested compared to pine and oak [98].

* See also: Thyry, Y., The true distribution and accumulation of radiocesium in stems of Scots pine, J Environ. Radioactiviy (2001, in press)

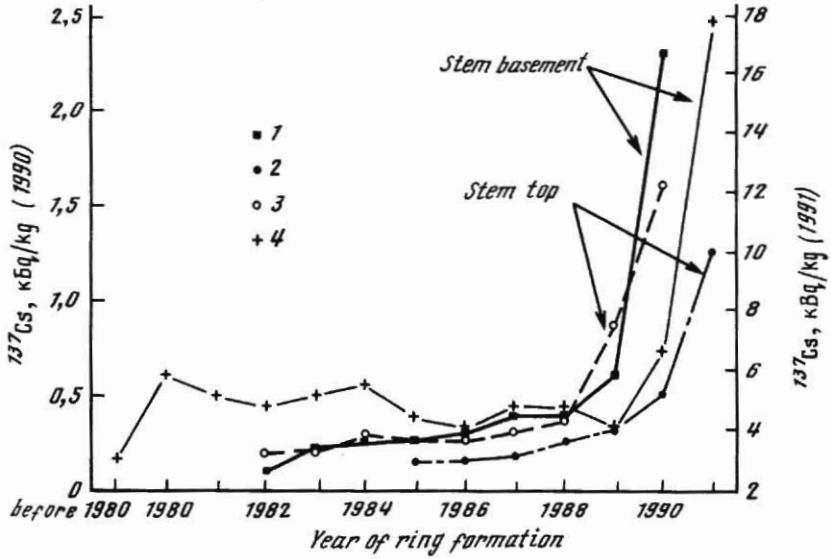

Fig. 15. ^{137}Cs distribution in the annual tree rings:
(1, 2) 1990; (3, 4) 1991

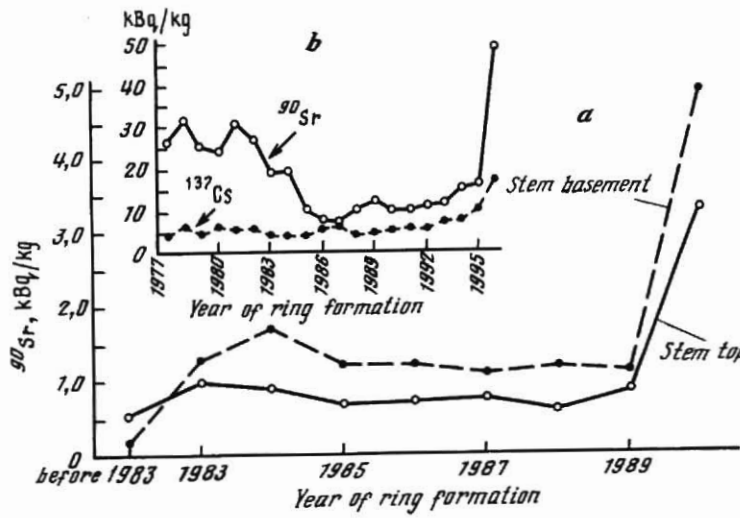

Fig. 16. ^{90}Sr distribution in the annual tree rings:
(a) original data; (b) data by [35]

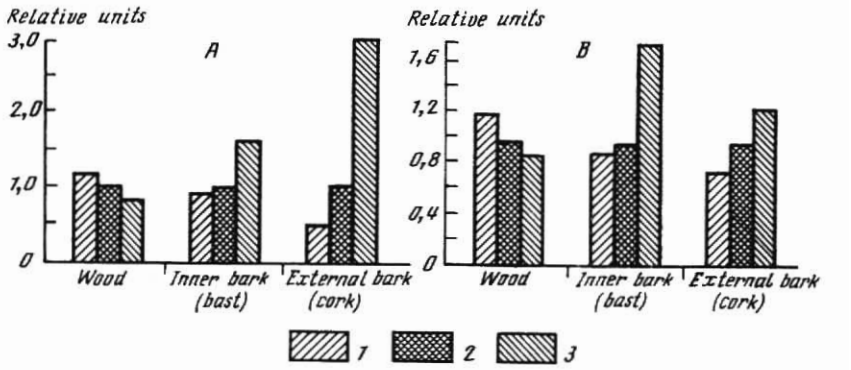

Fig. 17. ^{137}Cs dynamics in the stem components under (a) automorphic and (b) hydromorphic conditions:
(1) top of the tree; (2) middle of the tree; (3) basement of the tree

The concentration of both ^{137}Cs and, particularly, ^{90}Sr within the same ring decreases from the tree butt to the top (Fig. 15, 16). On the contrary, the weighted average concentration of ^{137}Cs in the stem wood increases from the butt to the top for all species, i.e. the barked wood of younger stem parts contains more radionuclides compared to the older parts (Fig. 17). The extent of these differences depends on variation in the stem thickness, species-dependent features of the wood structure, and contribution of the tree rings formed after 1986 to the total wood biomass, since these rings are considerably more contaminated compared to the pre-accidental rings (Table 23) [98, 347]. Such a height-dependent distribution of radionuclides in the wood is most manifested for pine and oak, and least manifested for alder.

Table 23. Distribution of the wood mass and ^{137}Cs across the tree stem (*Pinus sylvestris*, 70 years old)

Stem component	Tree rings, (years of formation)	Wood (mass %, dry weight)	^{137}Cs , Bq/kg
Tree top	1983–85*	11.8	2.92
	1986–91**	88.2	5.5
	Total	100	5.2
Stem basement	1922–79*	51.7	3.07
	1980–84**	21.3	4.81
	1985–91**	27	5.95
	Total	100	4.22

* sapwood
** heartwood

Table 24. Relative content of ^{137}Cs in the bark and various parts of stem wood (1993)

Species	Tree components*								
	Stem wood			Inner bark			External bark		
	1	2	3	1	2	3	1	2	3
Spruce	1.45	1**	1	1.11	1	1.24	1.91	1	1.06
Birch	1.32	1	1.02	0.96	1	0.85	0.68	1	1.15
Oak	1.37	1	0.91	0.83	1	1.1	1.08	1	0.58
Aspen	1.35	1	0.56	1.22	1	1.12	0.4	1	0.35
Alder	1.17	1	1.16	0.76	1	0.96	0.51	1	0.41

* 1 – top of the tree ; 2 – middle of the tree ; 3 – bottom of the tree; ** ^{137}Cs content in the middle of the tree is taken as 1

The height-dependent distribution of radionuclides in the inner, alive bark is opposite to that in the wood (see Fig. 15, 17): the highest and the lowest ^{137}Cs contents are attributed to the butt and top, respectively. The cause of this difference is not clear yet. It may only be said that the contamination of alive bark depends primarily on the root uptake, although it may be affected by the contaminated external bark (cork) as well. The differences between the radionuclide content in the external bark sampled from the tree top and butt are especially typical of the trees growing in hydromorphic accumulative landscapes or in the areas with the most pronounced root uptake of radionuclides (see Fig. 17) [280].

The behaviour of ^{137}Cs in the external bark depending on the stem part (Fig. 17, Table 24) is rather ambiguous. The species with smooth or scaly bark (birch, pine) accumulate more ^{137}Cs in the butt end, which may be explained by a higher sorption capacity of thick rough and fractured cork covering the tree butt and/or by the enrichment of the tree butt with radionuclides owing to the downward migration from the upper parts of the tree (througfall, stemflow, etc.). The species with a rough bark possess, apparently, a higher capacity for particle retention, and tend to accumulate more radionuclides in the middle stem.

Distribution of ^{137}Cs in Roots

It is reported that the radionuclides tend to accumulate in the tree root systems [146, 207]. The weighted average concentration of ^{137}Cs was shown to be 1.5–2 times higher in the pine underground organs (roots) compared to the aboveground organs [207] (Table 25). ^{137}Cs is more uniformly distributed among the root components (root wood and root covers) compared to the wood and bark of branches and stem, although the main trends are the same: minimum and maximum radionuclide concentrations are characteristic of internal (wood) and external (bark, root covers) parts, respectively. ^{137}Cs concentration in the pine roots depends on their depth, diameter, and structure: it is maximal in the roots attributed to the upper soil layers and thin sucking roots of 1–2 mm in diameter. In deeper soil layers and in the large roots, ^{137}Cs content is several times lower because of a higher proportion of wood (the least contaminated component) and a lower radionuclide content in the adjacent soil. Interestingly,

Table 25. ^{137}Cs content and distribution in the pine roots (*Pinus sylvestris*, kBq/kg dry mass, 1994)

Depth, cm	Root diameter, mm				
	0–1	2–3	3–10	10–20	20–30
	Entire roots				
0–10	6.9	3	nd*	nd	nd
10–20	5	2	nd	nd	nd
20–30	2.6	2.3	nd	nd	nd
30–70	2.2	1.6	nd	nd	nd
10–70	nd	nd	1.4	0.8	0.7
	Root components (layer 10–70 cm)				
Component					
Wood	nd	nd	nd	0.5	0.3
Inner skin	nd	nd	nd	1.5	1.8
External skin	nd	nd	nd	1.4	1.4

* No data

^{137}Cs concentration in the deep roots is higher than in the adjacent soil by a factor of 2–100 depending on the depth (soil layers 0–10 and 30–70 cm, respectively). The deep soil layers may therefore be enriched with radionuclides via root exudates (at least theoretically).

Thus, during the first months after the accident, ^{137}Cs and ^{90}Sr are rather uniformly distributed among the tree components over the entire contaminated territory, being controlled by the same factors (weather conditions, parameters of the fallout, etc.). The radionuclide concentration is maximal in those tree components that were directly exposed to the fallout (leaves, needles, twigs, etc.). The radionuclide content in the tree components depends on the vegetative stage and character of the surface of the organs exposed to the fallout, presence of liquid exudates on their surface, etc. One year after the accident, ^{137}Cs and ^{90}Sr distribution in the tree components is still quite uniform, but external bark becomes the most contaminated organ. Tree species with a rough and fractured bark (oak, alder, lime tree) are usually more contaminated than others. Later on, 2–3 years after the fallout, the distribution of radionuclides among the structural components depends on a wide range of factors. ^{90}Sr concentration in the tree organs unexposed to the initial fallout starts to increase. At the same time, ^{137}Cs redistribution among the tree components depends primarily on the soil conditions and distance from ChNPP (physicochemical properties of the initial fallout). On automorphic soils, particularly chernozems and other clayey soils, the contribution of external bark to the total stand contamination remains the same or even increases with time. On hydromorphic soils and in the near zone enriched with the fuel component, the contribution of inner components

Table 26. Statistical indices of ^{137}Cs and ^{90}Sr content in the organs and components of *Pinus sylvestris* (30 km exclusion zone, 1992, kBq/kg)

Tree components	Statistical indices						
	n	m	±m	max	min	G	V, %
			^{137}Cs				
Wood	42	5.01	0.47	10.36	0.52	3.08	61.5
Inner bark (bast)	42	53.09	5.2	152.93	8.88	33.76	63.6
External bark (cork)	42	38.6	4	114.7	18.87	25.94	67.2
Large branches	41	10.92	1.14	35.52	2.22	7.3	66.9
Small branches	41	19.53	1.8	44.4	3.15	11.51	58.9
"Old" needles	42	17.71	1.77	48.1	2.63	11.49	64.9
Needles of current year	42	64.74	5.76	144.3	10.73	35.35	57.7
Cones	31	46.07	6.47	136.9	8.14	36.01	78.2
			^{90}Sr				
Wood	42	5.91	1.17	35.52	0.67	7.56	127.8
Inner bark (bast)	42	29.35	4.64	155.4	4.11	30.1	102.6
External bark (cork)	42	21.31	3.98	99.9	3.03	25.81	121.1
Large branches	42	12.25	2	51.8	1.71	12.95	107.5
Small branches	42	15.73	3.18	92.5	0.59	20.58	130.8
"Old" needles	42	15.15	3.4	88.47	1.04	22.03	145.5
Needles of current year	39	12.14	2.91	77.7	0.67	18.17	149.7
Cones	26	0.6	0.08	1.67	0.08	0.39	65.8

Footnote: m – mean; G – standard deviation; ±m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

(internal bark, wood, young assimilative organs) contaminated due to the root uptake increases in time, and the contribution of the tree components exposed to the initial fallout decreases drastically.

The root uptake is, therefore, the leading process determining ^{90}Sr distribution among tree organs and components in the entire investigated territory. For ^{137}Cs, the leading process depends on a wider range of factors: in automorphic areas it is a combination of mechanical and biological natural decontamination, while the root uptake is the most influential process in hydromorphic areas.

Both ^{137}Cs and ^{90}Sr undergo vertical and lateral redistribution within the stem wood. ^{90}Sr is characterised by a higher migration capacity and a more manifested accumulation in the pre-accidental tree rings compared to ^{137}Cs.

Table 27. Statistical indices of ^{137}Cs content in the wood and inner bark of *Pinus sylvestris* in the middle, top and bottom of the stem (kBq/kg, 1992)

Tree components	Statistical indices						
	n	m	±m	max	min	G	V, %
Wood							
Top of the tree	38	5.55	0.54	11.84	0.74	3.36	60.5
Middle of the tree	42	4.83	0.47	10.36	0.48	3.07	63.5
Bottom of the tree	40	4.17	0.45	9.62	0.35	2.87	68.7
Inner (alive) bark							
Top of the tree	38	44.49	4.55	114.7	7.77	28.02	63
Middle of the tree	42	40.15	3.89	114.7	7.03	25.22	62.8
Bottom of the tree	40	75.75	8.08	229.4	11.84	51.08	67.4

Footnote: m - mean; G - standard deviation; ±m - mean error; V, % - variation coefficient; max, min - maximal and minimal values, respectively

General features of the distribution of radionuclides in the aboveground and underground components of trees are rather similar: bark (root covers) and wood are the most and the least contaminated components, respectively. The weighted-average concentration of radiocaesium in the roots is 1.5–2 times higher than in the aboveground vegetative organs. The most contaminated are thin roots of 1–2 mm in diameter located in the soil upper layers. ^{137}Cs content in the underground roots decreases as the depth of the roots increases, though the specific radioactivity in roots increases with the depth of these roots.

Spatial Variability of the Radionuclide Accumulation by Plants

To predict correctly the behaviour of radionuclides in arboreal vegetation and forest produce it is necessary to estimate the peculiarities of spatial variability of the radionuclides in the plant components. In general, the variation coefficient of specific activity in the stand is 2–3 times higher than in the soil, where it is about 30% [344]. Thus, radionuclide accumulation by trees depends, as a rule, on the variation of the soil and other environmental conditions rather than on the initial deposition (even within a limited area). Spatial variation of ^{137}Cs in the investigated tree components may be arranged in the following sequence: cones > external bark > internal bark > needles > wood > large branches > small branches > twigs of the current year.

The ^{90}Sr content in the pine organs is more variable than ^{137}Cs (Table 26). This is likely due to a higher biological availability of ^{90}Sr (increase in the uptake rate leads to a corresponding increase in the variation). Interestingly, the range of ^{90}Sr variation depending on tree component is almost opposite to the corresponding range for ^{137}Cs.

The variation of the radionuclide content increases from the tree top to the bottom (Table 27), which is apparently due to the change in the proportion of the core of wood (heart-wood) in the stem wood. The core of wood is the least contaminated tree component, and its contribution to the total stem contamination depends on both stand age and diameter.

Accumulation ^{137}Cs and ^{90}Sr by the Tree Organs and Components

High variability in the radionuclide accumulation by different tree organs calls for a high-replication sampling even within a limited key site to obtain reliable data. This increases the research cost manifold, and is practically impossible in the case of long-term monitoring (extended sampling of arboreal vegetation implies the intensive tree felling, which will inevitably interfere with natural ecological processes in the key site).

This problem may be solved by establishment of a correlation between the radionuclide content in the tree components to reveal the tissues and organs most representative for the radionuclide accumulation in the other tree organs and tissues.

Based on the extended field data, we calculated correlation coefficients for the radionuclide accumulation capacity between various pairs of pine components. The general regression equation is as follows:

$$Y = a_0 + a_1 \cdot x,$$

Where a_0 and a_1 are empirical coefficients shown in Table 28. The ^{137}Cs content in both twigs of the current year and internal bark is in the closest correlation with the radionuclide content in the other organs ($r > 0.9$). It means that the long-term monitoring of ^{137}Cs dynamics in the pine stand may be performed by annual (seasonal) sampling of young needles. Such sampling is convenient for the purpose of dynamic research because it provides a large sample without expensive and labour-consuming procedures and any adverse effect to the investigated ecosystem.

The correlation between ^{90}Sr content in the pine tissues is less than ^{137}Cs ($r_{max} = 0.8–0.9$). External bark and aged needles are the most representative tissues for this radionuclide. Unlike ^{137}Cs, ^{90}Sr is retained firmly by the "aged" organs and tissues, which may serve as a good indicator of the radionuclide content in other organs and tissues, except for such intensively growing components as leaves, needles, and cones. ^{90}Sr content in the latter is least agreed with its content in the above-mentioned indicative organs.

Thus, the indicative organs are radionuclide-specific. For ^{137}Cs, the best indicative organs are the most physiologically active ones, such as leaves or needles of the current year. In contrast with it, the most representative for ^{90}Sr are dead or aged components (cork and aged needles). That is why we classify ^{90}Sr and ^{137}Cs as "residually accumulated" and "actually accumulated" radionuclides, respectively. On the multiyear scale, it is reasonable to expect more manifested accumulation of ^{90}Sr, and less manifested accumulation of ^{137}Cs in pine wood.

Transfer Factors of ^{137}Cs and ^{90}Sr to the Components of Arboreal Vegetation: Variation and Dynamics

It is impossible to use the absolute radionuclide content (Bq/kg or Bq/m^2) to characterise a spatial variation of the radionuclide accumulation by plants under a wide range of deposition conditions (in this case, almost four orders of magnitude for ^{137}Cs and three orders of magnitude for ^{90}Sr). A direct effect of deposition on radionuclide accumulation by the tree components is obvious and has nothing to do with scientific analysis. Much more interesting are the data on the so-called normalised concentrations or transfer factors (TF) of radionuclides

Table 28. The coefficients of correlation and regression for ^{137}Cs and ^{90}Sr content in the tree components (n = 53)

Tree components		Coefficient of correlation	Coefficients of regression $y = a_0 + a_1 \cdot x$	
y	X	r	a_0	a_1
	^{137}Cs			
	Stem wood:			
	Top	0.87	9.8	0.08
	Middle	0.81	7.15	0.06
	Bottom	0.89	–7.08	0.07
	Entire stem	0.84	17.14	0.07
	Inner bark:			
	Top	0.9	–16.13	0.75
	Middle	0.9	–25.35	0.69
	Bottom	0.86	–86.26	1.37
Needles of	Entire stem	0.78	176.4	0.73
current year	External bark:			
	Top	0.9	25.41	0.25
	Middle	0.58	259.6	0.15
	Bottom	0.24	NS	NS*
	Entire stem	0.71	652.5	0.25
	Old needles	0.93	–15.14	0.29
	Cones	0.88	–185.6	0.91
	Small branches	0.89	23.75	0.31
	Large branches	0.36	90.56	0.15
	^{90}Sr			
	Needles of current year	0.69	0.73	0.13
	Wood	0.93	2.67	0.18
	External bark	0.77	0.66	0.99
Old needles	Inner bark	0.69	0.51	0.08
	Small branches	0.77	0.72	0.09
	Large branches	0.88	0.52	0.04
	Cones	0.44	0.738	3.1
	Wood	0.95	2.84	0.5
External bark	Large branches	0.83	1.65	0.18
	Inner bark	0.67	0.58	0.1

* No significant correlation

Table 29. The range of variation of TF^{137}Cs and TF^{90}Sr from the soil to various pine organs and components,
(TF = kBq/m^2_{soil}:Bq/kg_{plant} = n 10^{-3} m^2/kg, dry weight,1992)

Region	TF	
	^{137}Cs	^{90}Sr
Wood		
Tula region (RF)	0.05–0.1	0.6
Kaluga region (RF)	0.05–7.4	No data
Bryansk region (RF)	0.15–4.4	1.5–12.1
Kiev region (Ukraine)	0.05–6.1	0.1–3.7
Entire territory	0.05–7.4	0.1–12.1
Inner bark		
Tula region (RF)	0.1–0.4	2.5
Kaluga region (RF)	0.4–44	No data
Bryansk region (RF)	0.7–22	8.6–43.7
Kiev region (Ukraine)	0.1–87	1.6–18.4
Entire territory	0.1–87	1.6–43.7
External bark		
Tula region (RF)	1.9–13.2	5.2
Kaluga region (RF)	1.6–15.6	No data
Bryansk region (RF)	1.2–21.6	4.9–52.6
Kiev region (Ukraine)	0.8–30.6	0.4–36.7
Entire territory	0.8–30.6	0.4–52.6
Large branches		
Tula region (RF)	0.2–2.3	3.3
Kaluga region (RF)	0.2–10.1	No data
Bryansk region (RF)	0.3–11.7	2.3–31.4
Kiev region (Ukraine)	0.3–13	0.3–20.2
Entire territory	0.2–13	0.3–31.4
Small branches		
Tula region (RF)	0.06–0.4	4.3
Kaluga region (RF)	0.3–28.8	No data
Bryansk region (RF)	0.2–17.7	3.6–79.2
Kiev region (Ukraine)	0.3–34.2	0.3–25.1
Entire territory	0.06–34.2	0.3–79.2

Region	TF	
	^{137}Cs	^{90}Sr
"Old" needles		
Tula region (RF)	0.2	3.3
Kaluga region (RF)	0.2–14.3	No data
Bryansk region (RF)	0.2–32.2	1.9–8.1
Kiev region (Ukraine)	0.2–21.9	0.1–5.7
Entire territory	0.2–32.2	0.1–8.1
"Young" needles (of current year) and leaves		
Tula region (RF)	0.1–0.6	1.5
Kaluga region (RF)	0.5–48.6	No data
Bryansk region (RF)	0.5–37.4	2.3–67.6
Kiev region (Ukraine)	0.3–111.0	0.2–32.2
Entire territory	0.1–111.0	0.2–67.6
Cones		
Tula region (RF)	0.5–1.7	0.6
Kaluga region (RF)	1.2–37.8	No data
Bryansk region (RF)	0.9–39.4	0.5–2.2
Kiev region (Ukraine)	1.6–88.5	0.02–0.6
Entire territory	0.5–88.8	0.02–2.2

from various environments to the plants. In our study, TF serves as an integral index of biological availability of the radionuclide, and is calculated as follows:

TF = [concentration in the plant (Bq/kg)] : [deposition (Bq/m^2)].

Normalisation of the radionuclide content against deposition (and not against concentration in the soil) makes it possible to compare the features of radionuclide accumulation by a plant irrespective of the vertical distribution of radionuclides in the soil profile, i.e. depending on ecological factors, soil properties (regime), species, etc.

Table 29 shows that TF of ^{137}Cs and ^{90}Sr to arboreal vegetation vary in a wide range depending on the spatial scale (ecotope, climatic zone, or contaminated territory as a whole), species, and structural component. The variation range of TF in all tree components tends to increase as the observed territory increases (e.g., TF variability over a single experimental site is 10 times less than the corresponding value over a climatic zone). There is a general descending trend of TF variation from the 30 km exclusion zone (Kiev region, Ukraine) toward the forest-steppe belt north-east of ChNPP (Tula region, the Russian Federation). The minimum variation range is observed in the chernozem zone,

Table 30. The range of variation of $TF^{137}Cs$ and $TF^{90}Sr$ to different species and tree components ($TF = Bq/kg_{plant} : kBq/m^2_{soil} = n \cdot 10^{-3}\ m^2/kg$, dry weight, 1992)

Radionuclide	Species						
	Pine	Spruce	Birch	Oak	Aspen	Alder	Teil
			Wood				
^{137}Cs	0.04–6.1	0.05–1.5	0.05–7.4	0.1–4.7	0.02–1.9	0.5–2.7	0.2
^{90}Sr	0.03–3.3	No data	0.3–12.1	0.02–2.8	0.2–1.9	0.2–1.0	No data
			Inner bark (bast)				
^{137}Cs	0.4–87.0	2.2–24.8	0.3–23.0	0.2–19.8	0.09–13.2	5.4–10.4	0.5
^{90}Sr	0.2–18.2	No data	0.4–36.6	2.6–43.7	2.2–18.4	1.8–7.6	0.5
			External bark (cork)				
^{137}Cs	1.0–18.1	3.4–20.0	0.8–11.8	12.5–28.6	5.7–30.6	9.2–37.3	9.9–11.6
^{90}Sr	0.4–37.3	No data	0.5–24.8	1.2–20.3	1.8–33.3	4.0–12.4	No data
			Large branches				
^{137}Cs	0.2–8	0.7–11.7	0.2–10.3	0.6–10.3	0.3–3.8	3.5–8.9	1.2–2.1
^{90}Sr	0.2–8.8	No data	0.6–31.4	1.0–4.7	1.3–20.2	1.7–2.8	No data
			Small branches				
^{137}Cs	0.2–28.8	1.0–12.5	0.4–34.2	0.2–17.6	0.06–6.5	5.8–13.5	0.3
^{90}Sr	0.3–98.0	No data	1.2–79.2	1.2–8.5	2.6–25.1	2.6–5.9	No data
			Needles of current year and leaves				
^{137}Cs	0.4–111.0	1.6–25.7	0.5–55.3	0.3–37.5	0.1–24.3	4.9–13.1	0.5–0.6
^{90}Sr	0.08–15.5	2.5–67.6	0.8–2.4	No data	2.4–32.2	1.7–5.7	No data
			Generative organs (cones and birch aglet)				
^{137}Cs	0.9–88.5	1.2–2.4	No data	No data	No data	No data	No data
^{90}Sr	0.02–2.2	No data	0–0.9	No data	No data	No data	No data

which is due to a higher clay content and less variable moisture regime of the soil cover in the forest-steppe and steppe natural zones compared to Ukrainian Polesie (south taiga zone). The TF variation is, therefore, determined primarily by soil-ecological factors. As to the variation range of $TF^{137}Cs$ to different tree components (organs), the highest values are typical of the components contaminated primarily via the root uptake (assimilative organs, internal bark and wood). This indicates high sensitivity of these organs to the radionuclide availability in the soils.

External bark (cork) is characterised by minimum variation of TF, and is less dependent on a climatic zone. This reflects its lower dependence on the root uptake and higher dependence on external contamination compared to the other organs and tissues.

^{90}Sr is characterised by a lower TF variability, but higher limiting (max, min) and average TF values compared to ^{137}Cs. The latter reflects a higher mobility of ^{90}Sr and lower dependence of its availability on the soil-ecological conditions. The ^{90}Sr availability for plants increases as the distance from the accidental unit increases, which is undoubtedly due to different physicochemical forms of the initial fallout: unlike ^{137}Cs, ^{90}Sr was incorporated mainly in the low-soluble fuel particles. This factor still affects the behaviour of strontium in the environment up to now, more than 10 years after the accident [111]. Thus, the physicochemical properties of the initial fallout are a more influential and more long-term factor of ^{90}Sr than ^{137}Cs transfer to the tree plants. The highest variation of $TF^{90}Sr$ was observed in leaves and branches, and the lowest variation was typical of the internal bark. This is an example of high sensitivity of assimilating organs to biological availability of strontium in the soil.

The range of TF variation of both ^{90}Sr and ^{137}Cs depends also on tree species (Table 30). There is a positive correlation between TF of these radionuclides and species habitat (area). The highest variation of TF is characteristic of the species with a wide natural area, such as birch and pine. In contrast with it, minimum variation of TF is typical of the species with limited area, such as bass-wood, alder, and oak. The latter are known to grow on fertile soils rich of nutrients, and bass-wood gravitates toward moist habitats. ^{90}Sr and ^{137}Cs are differently accumulated by tree species: birch exhibits the highest $TF^{90}Sr$ of all species that is twice as high as $TF^{137}Cs$. Alder exhibits a completely opposite feature: the lowest $TF^{90}Sr$ of all species that is twice as low as $TF^{137}Cs$. These peculiarities may depend on both species physiology and different relative availability of ^{137}Cs and ^{90}Sr in different habitat (for example, ^{90}Sr is known to be less mobile than ^{137}Cs in peat soil which is the habitat of alder) [135].

Seasonal dynamics. The radionuclide content in trees exhibits a pronounced seasonal dynamics. For ^{137}Cs, seasonal dynamics is expressed as a monotonous decrease during the growing season (from spring to autumn, Fig. 18). This is manifested mostly for young trees due to a higher intensity of metabolism and root uptake. Maximum differences in the radionuclide uptake between mature and younger trees take place in the beginning of a growing season, during spring months. In other words, the intensity of basic biogeochemical processes influencing the radionuclide migration in the soil-plant system varies by seasons.

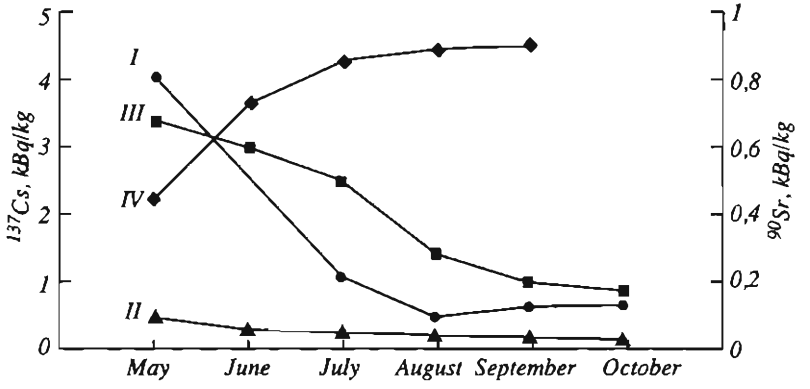

Fig. 18. Seasonal dynamics of ^{137}Cs and ^{90}Sr in the pine needles of a current year (young needles) depending on the tree age:

I - ^{137}Cs, 20 years old (site D-1); II - ^{137}Cs, 60 years old (site D-1); III - ^{137}Cs, 60 years old (site K-2); IV - ^{90}Sr, 60 years old (site K-2)

Unlike ^{137}Cs, seasonal dynamics of ^{90}Sr in the assimilative organs of various tree species reaches its maximum by early autumn. This is likely due to the fact that ^{90}Sr (like its non-isotopic carrier Ca) tends to accumulate in older tissues with less intensive metabolism. Calcium is known to control the water balance regulation in the plant tissues. In particular, the increase in Ca (Sr) concentration leads to decrease in the water content in the plant tissues by the end of the vegetative season, which is one of the plant adaptation to low winter temperatures [8].

Long-term dynamics. The long-term (multiyear) dynamics of TF to vegetation is most manifested during the first months and years after the accident. For example, in the first 3–5 years after the accident, ^{137}Cs content in the plants decreases by several orders of magnitude [255, 256]. In the course of time, this process slows down and the TF dynamics becomes more complex. This is due to radionuclide redistribution in the "solid-liquid phase" system, i.e. radionuclide interaction with soil adsorption complex. In the case of ^{137}Cs, the interaction leads to irreversible fixation of a considerable amount of this radionuclide on clay minerals [29]. At this stage, the long-term caesium dynamics in the arboreal vegetation is determined by two competitive processes: (i) natural surface decontamination and (ii) root uptake. The intensity of both processes depends on a wide range of biotic and abiotic factors: the physicochemical properties and particle size of the initial fallout, weather and climate conditions, species composition of the stand, the soil properties and regimes, etc. That is why the long-term dynamics of TF^{137}Cs to arboreal vegetation varies and is very different over the contaminated zone. In general, we distinguish three basic types of the long-term dynamics of TF^{137}Cs to arboreal vegetation (Fig. 19, 20).

1. Pronounced decrease in TF. This type of dynamics is observed in automorphic soils of eluvial landscapes. The monotonous decreasing trend of TF depends on the synergetic effect of (i) continuous natural decontamination and (ii) continuous decrease in ^{137}Cs biological availability in the soil because of its irreversible adsorption by the soil solid phase. This is confirmed by a particu-

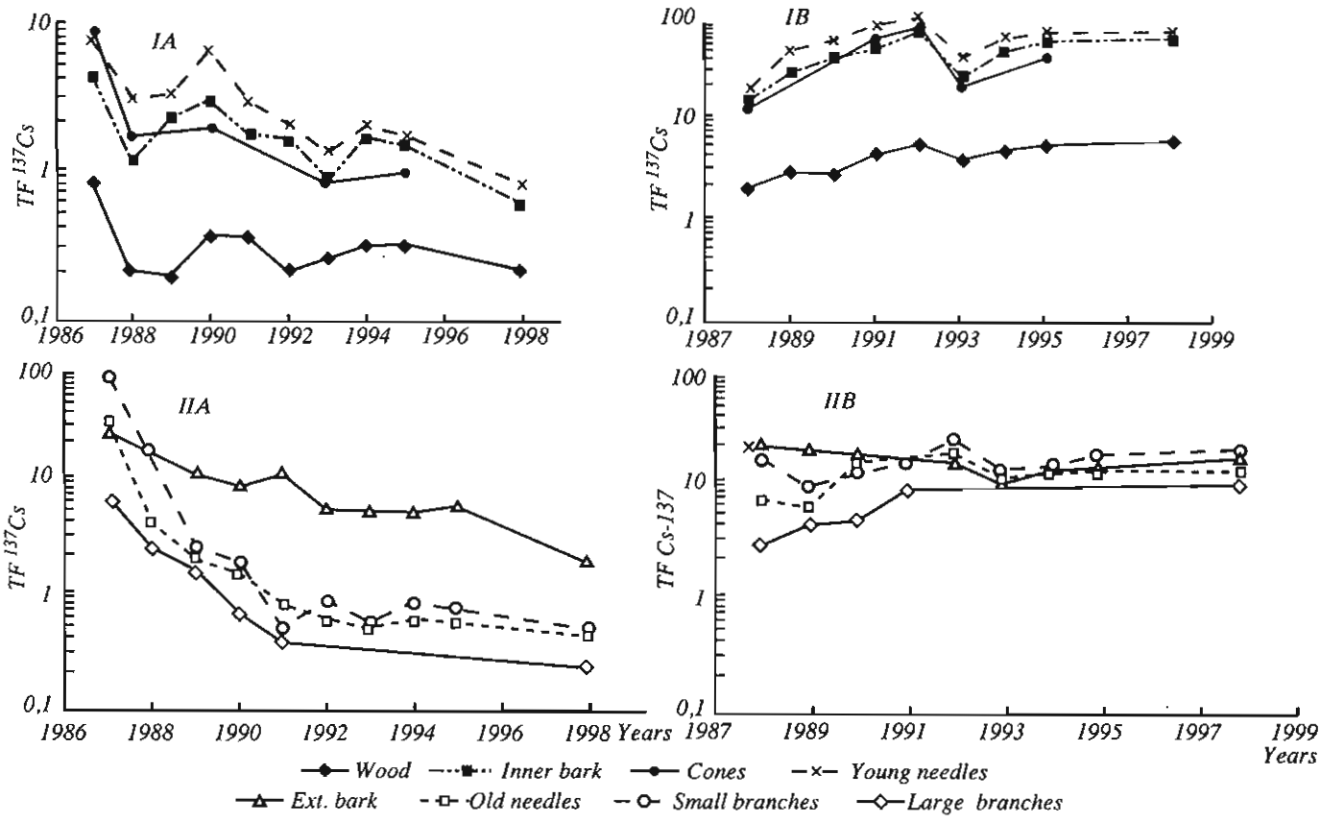

Fig. 19. The dynamics of TF^{137}Cs from the soil to various pine organs and components:
(I) not exposed and (II) exposed to the surface contamination, on (A) automorphic and (B) hydromorphic soils

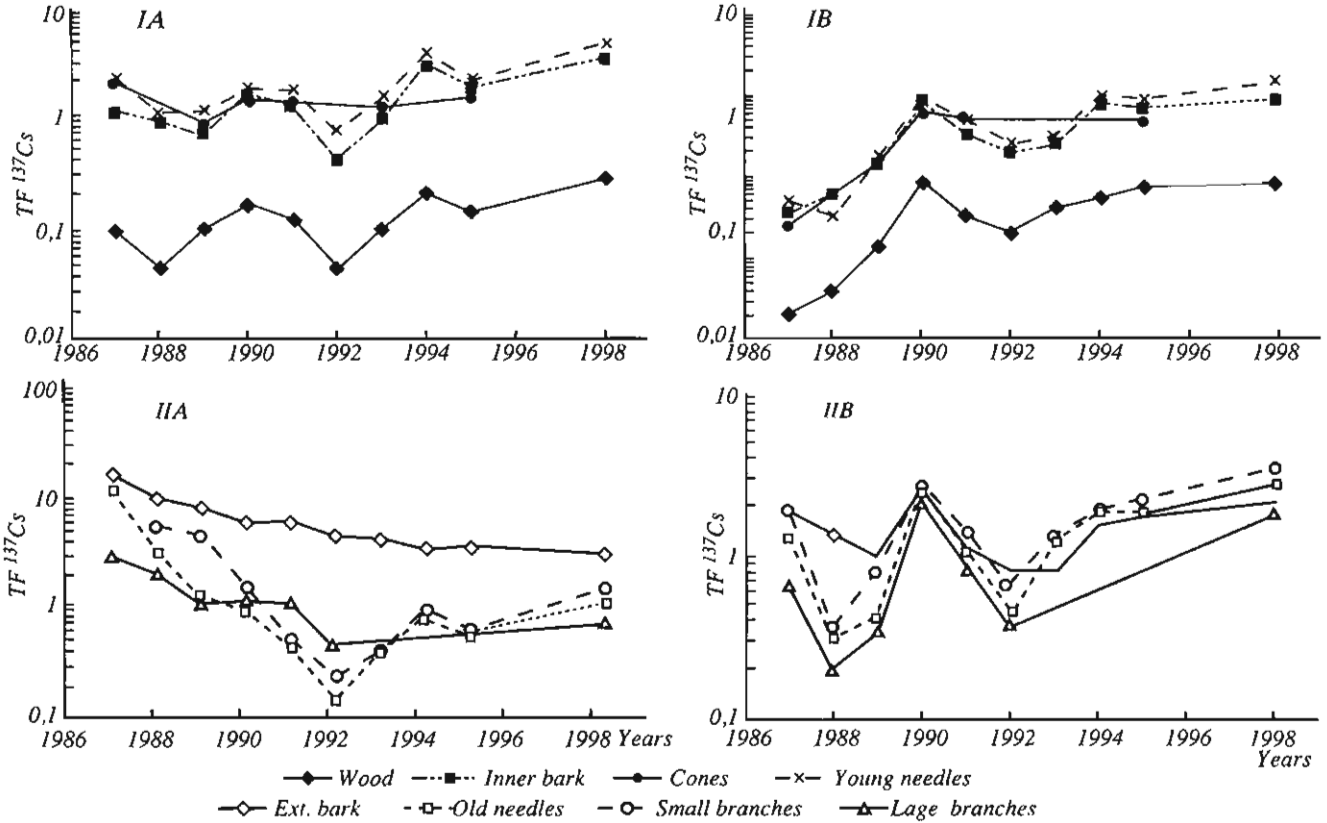

Fig. 20. The dynamics of $TF^{137}Cs$ from the soil to various pine organs and components:
(I) not exposed and(II) exposed to the surface contamination, on the automorphic soils in (A) the middle of exclusion zone (5–10 km from ChNPP) and (B) in the "near zone"

larly pronounced decrease in TF on chernozems where the irreversible fixation of ^{137}Cs is known to be very intensive [122]. Variation in the TF dynamics by years is likely to be due to weather conditions and corresponding changes in the soil moisture and caesium equilibrium in the "solid-liquid" system.

2. Pronounced increase in TF. This type of dynamics is characteristic of (i) hydromorphic areas and (ii) territories adjacent to the accidental unit (5–10 km of the near zone). In both cases, the increase in TF is due to dominance of the root uptake over the decontamination processes. The reasons for the dominance are, however, case-specific. In the accumulative landscapes, the increase in TF is determined by (i) radionuclide input from neighbouring eluvial territories, (ii) intensive radionuclide redistribution within the root-abundant layer, and (iii) absence of the irreversible adsorption in the organic (peat) soils typical of the accumulative landscapes. In the near zone, TF increases at the expense of continuous release of mobile ^{137}Cs to the soil solution from the fuel particles. Annual variation of TF is likely to be due to different weather conditions affecting the rate of radionuclide release from the fallout particles.

3. No manifested trend of TF. This type of dynamics is characterised by a high annual variability of TF without a pronounced descending or ascending trend. Such dynamics is observed in semi-hydromorphic (trans-accumulative) landscapes and the central part of the exclusion zone (10–15 km from ChNPP). The absence of a manifested trend is apparently due to the cancellation effect of the above-mentioned competitive processes (radionuclide mobilisation, immobilisation, lateral migration, sorption-desorption, irreversible adsorption, etc.).

A general accumulative character of the radionuclide dynamics in the tree phytomass may, however, result in a weak ascending trend of $TF^{137}Cs$ on a long-term scale (assuming that all other above-discussed factors are stable). We would like to emphasise that such an increase in TF is due to the long-term phytomass accumulation in the forest ecosystem (and the corresponding caesium accumulation in the plant tissues) rather than the changes in the biological availability of caesium in the soil.

The tree components may be pooled into two groups by the long-term dynamics of $TF^{137}Cs$. The first group includes inner (covered) components and organs contaminated almost completely via the root uptake: alive bark, cambium, assimilative organs of the current year, generative organs, and wood. The dynamics of TF for this group obeys the above-mentioned three basic types depending on the ecosystem and geographical position of the territory. The second group includes (i) tree components and organs exposed to the primary fallout and resuspended fallout particles, and (ii) surface decontamination (mechanical or/and biological) of external bark, large branches, and aged needles (2–3 years old). The TF trend in these components is characterised as an exponential decrease in the first 5–6 years after the accident, followed by an ambiguous trend in the ensuing years. The ecosystems with the conditions for a monotonous increase in the root uptake (like those in the near zone) are the only exception. In the latter case, the late stages of the multiyear TF dynamics to the components of the second group may (1) be similar to the first group (dependent on the root uptake), or (2) remain more or less stable, with annual variation. The latter takes place in the case of quasi-equilibrium when gradually damping processes of natural decontamination are generally compensated by the increasing root uptake.

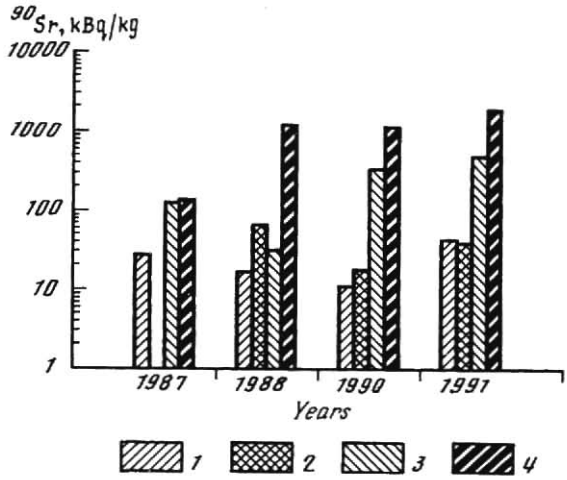

Fig. 21. The dynamics of ^{90}Sr content in the pine wood in various key sites: (1) D-1; (2) D-3; (3) K-2; (4) Sh-1

The long-term dynamics of TF^{90}Sr to arboreal vegetation is different from TF^{137}Cs (Fig. 21), and the differences increase with the passage of time. This is due to the fact that biological availability of ^{137}Cs in the first years after the accident was higher than that for ^{90}Sr, and TF^{137}Cs was higher at that time. In the subsequent years, the availability of ^{137}Cs for tree plants decreased and the availability of ^{90}Sr increased in all investigated key sites. This is likely due to the fact that the accumulative character of the multiyear dynamics of tree phytomass has a higher effect on the dynamics of ^{90}Sr compared to ^{137}Cs. We have already mentioned that ^{90}Sr tends to accumulate in older tissues with a less intensive metabolism.

The effect of soil properties on the biological availability of ^{90}Sr is much less than that of ^{137}Cs, and sometimes the effect of the same soil factor on TF of these radionuclides is even contradictory [8]. In general, the ^{90}Sr content in the pine wood increases in time on automorphic soils and decreases on peat soils, which is likely due to a different pH and Ca content in the soil adsorption complex in different soil types. Another possible reason of the above-mentioned differences is that, unlike ^{137}Cs, ^{90}Sr is able to bind firmly with soil organic compounds, which makes it low mobile in the organic soils. In contrast with it, organic soils promote high mobility of ^{137}Cs.

Dynamics of the Variation Range of TF^{137}Cs to Arboreal Vegetation.

The long-term dynamics of TF^{137}Cs to arboreal vegetation is also expressed in the dynamics of limits (max, min) and a variation range ($Vr = TF_{max} - TF_{min}$) of the radionuclide content (Fig. 22). The dynamics of these indices is not the same for different tree components. In the components exposed to surface contamination (external bark, small branches, and "old" needles), the variation range of TF^{137}Cs is maximum in the first years after the accident. By the third or fourth year after the accident, this index drops drastically, and remains stable (with some annual variation about the average level) during the subsequent years. Such dynamics is apparently dependent on the individual contribution of two competitive processes: surface decontamination and root uptake. In the first

2–3 years after the accident, the first process was absolutely dominant. In the ensued years, natural decontamination is superposed with the increasing root uptake, followed by physiological redistribution of the radionuclide among the tree components.

The components contaminated primarily due to the root uptake (internal bark and needles of the current year) are characterised by a 3 to 5 times higher variation range of $TF^{137}Cs$. Wood has the least *Vr* of all components, but the long-term dynamics of this index is similar to that in the second group. All these components exhibit a dramatic increase in the variation range of $TF^{137}Cs$ during the first post-accidental years (with maximum in 1992), followed by some decrease and stabilisation. In some cases, *Vr* increases for a while, which is likely due to an annual increase in the root uptake rate.

$TF^{90}Sr$ is characterised by less *Vr* at any spatial scale (from an individual key site to a climatic zone). This is likely due to a less manifested effect of the soil properties and regimes on ^{90}Sr accumulation. In this case, the differences in its root uptake depend primarily on spatial heterogeneity of the initial fallout.

The Effect of Various Factors on ^{137}Cs and ^{90}Sr Transfer to Arboreal Vegetation

Physicochemical properties of the initial fallout. The physicochemical properties of the fallout, including particle size and solubility, are the most influential factor of the initial distribution of the fallout among the components of arboreal vegetation and radionuclide behaviour in the ensuing years. Since the size of the fallout particles was the largest in the near zone, TF of all investigated radionuclides to arboreal vegetation in this zone was considerably lower than in the remote territories (50–500 km and more from ChNPP). The maximum factor of variation of this index is 25 for pine and 10 for deciduous species, which is due to the differences in the initial biological availability of the radionuclide.

Such a great difference in the radionuclide availability is caused by a number of factors. In the closest vicinity of ChNPP (particularly, within the so-called western plume trace of the initial radioactive plume), the fallout particles reached 100 μm, whereas the particle size in the remote territories normally did not exceed 0.1–0.2 μm. Thus, natural decontamination of the tree crowns in the vicinity of ChNPP runs much faster than in the remote territories, which has resulted in a shorter contact time of the radioactive particles with leaves (needles) and less intensive period of direct radionuclide transport via the plant surface. Besides, large particles precipitated in the near zone had low solubility because of their lower specific area and chemical properties [91], which also reduced the radionuclide surface uptake. In addition, high sensitivity of pine to ionising irradiation could affect the physiological processes responsible for ion transport from the assimilative organs to the wood, which is likely to be one of the reasons of a higher TF variation in the pine wood compared to deciduous species.

In fact, the effect of the physicochemical properties of the fallout on radionuclide availability is limited by the boundaries of the 30 km exclusion zone. The radionuclide availability increases by a factor of 5–10 as the distance from the accidental unit increases from 5 to 30 km, whereas a farther increase in distance has much lower effect on the TF.

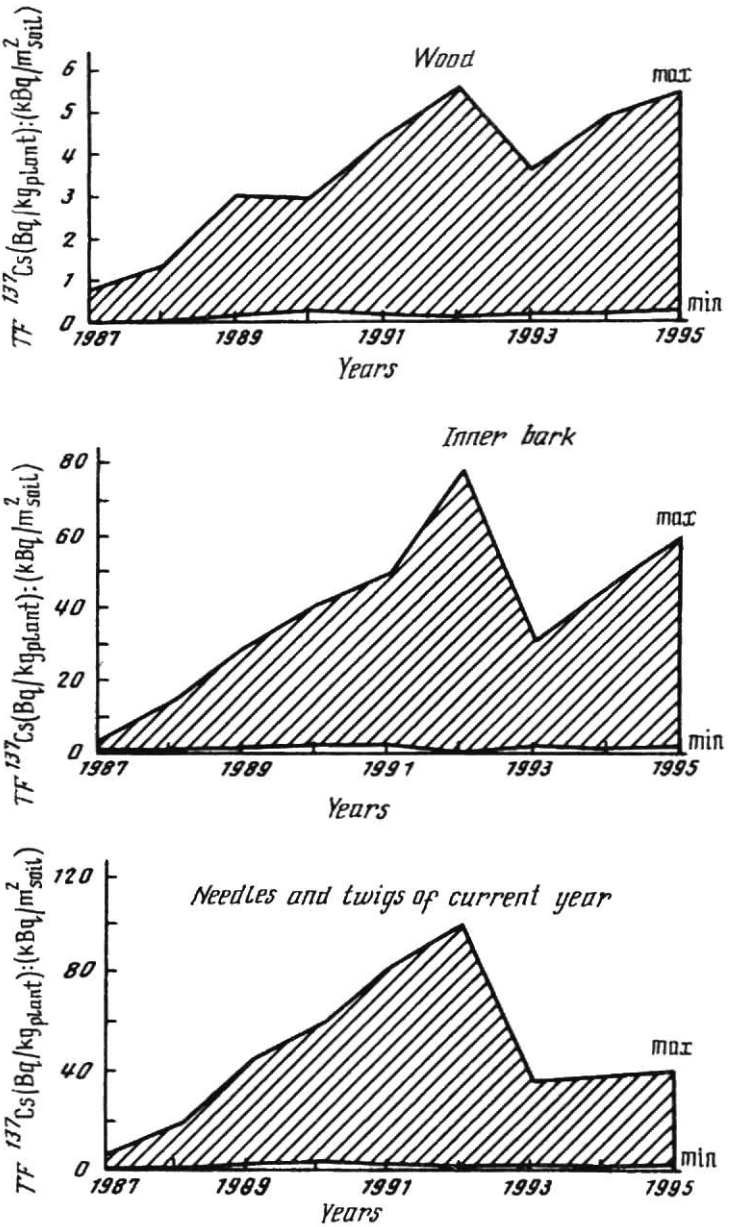

Fig. 22. The range of variation and limits of TF^{137}Cs from the soil to various pine organs and components (the exclusion zone)

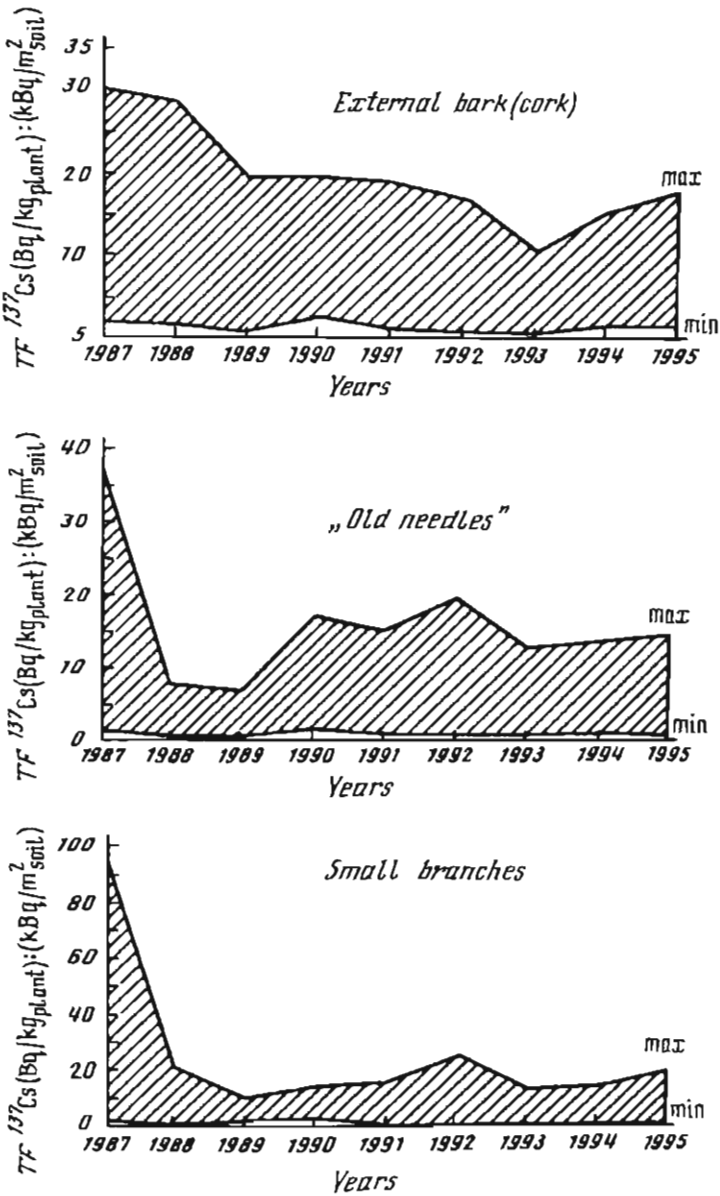

Fig. 22. (continuation)

The differences in TF due to the physicochemical properties of the fallout are going smoother with time because of the particle disintegration and more intensive radionuclide leaching, adsorption, and uptake. For example, in the first year after the accident (1986), the differences between $TF^{137}Cs$ to the same components of the same species reached one order of magnitude, two years

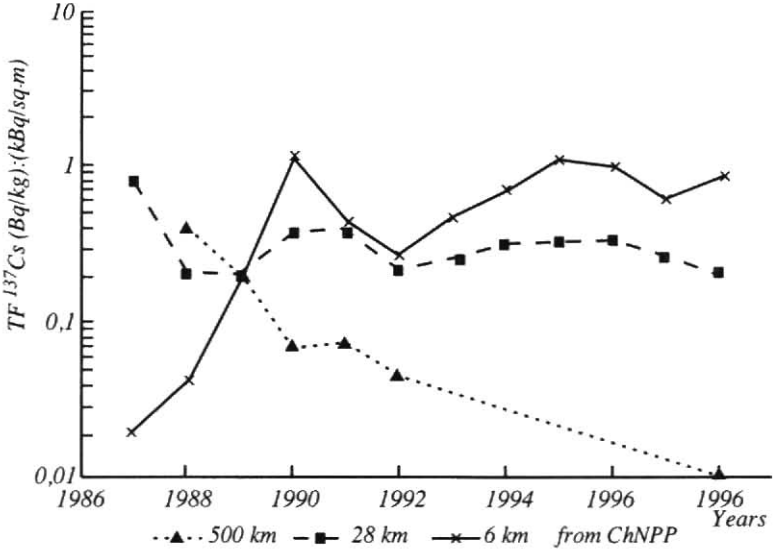

Fig. 23. The dynamics of TF^{137}Cs to the pine wood in automorphic areas at different distances from ChNPP

after, however, the factor of variation did not exceed 2–3. Four years after the accident, the biological availability of ^{137}Cs becomes almost uniform, though highly variable over the entire contaminated territory except for the near zone. Reliable differences in the ^{137}Cs content remained in the external bark only, whose contamination was higher in the marginal part of the 30 km exclusion zone compared to its inner part. 10 years after the accident, TF^{137}Cs to the tree plants in the "near zone" was still higher compared to the remote territories (Fig. 23). This gives us grounds to conclude that the physicochemical factor of the initial fallout loses its significance, although its effect may be observable for a long time within some limited areas. The present higher availability of ^{137}Cs in the near zone may exist only because of continuous leaching of the radionuclides from the "hot" particles. The latter is confirmed by the long-term dynamics of TF^{90}Sr to the overstorey. Unlike ^{137}Cs, ^{90}Sr is known to be incorporated in the fuel component [92, 111]. That is why, TF^{90}Sr to the wood in the first years after the accident was lower than TF^{137}Cs. By 1990, TF of both radionuclides increased, and the dynamics of TF^{137}Cs reached its absolute maximum, while TF^{90}Sr still continued to rise. The dynamics of TF of ^{90}Sr and ^{137}Cs reflects their leaching from the fallout particles, on the one hand, and irreversible adsorption in the soil (manifested chiefly for ^{137}Cs), on the other hand. The above-mentioned decrease in TF^{137}Cs since 1990 reflects irreversible adsorption of ^{137}Cs in the soil. The dynamics of TF^{90}Sr depends mainly on the rate of its leaching from the "hot" particles, since this radionuclide is not adsorbed by soil at the same high rate as ^{137}Cs. We believe that radionuclide leaching from the fuel particles in the near zone will remain an important factor of radionuclide transfer to plants, at least, in the near-term perspective.

Table 31. The effect of soil moisture regime on the $TF^{137}Cs$ and $TF^{90}Sr$ (Bq/kg_{plant} : kBq/m^2_{soil}) in Kaluga and Bryansk regions ($n \cdot 10^{-3}$ m^2/kg)

Area (moisture type)	Radionuclide	Organs and components						
		Wood	Bark		Branches		Leaves (needles)	Cones
			Inner	External	Large	Small		
				Pine				
Automorphic	^{137}Cs	0.07	0.56	1.14	0.26	0.4	1.11/0.17*	–
	^{90}Sr	0.61	3.64	2.17	3.8	6.1	1.6/2.3	–
Hydromorphic	^{137}Cs	3.96	44	10.9	7.97	17.5	48.6/14.5	37.8
	^{90}Sr	0.51	2.5	3.05	1.35	2.2	0.94/0.57	0.4
				Birch				
Automorphic	^{137}Cs	0.11	0.38	5.95	0.95	1.47	1.73	–
	^{90}Sr	2.25	6.36	9.1	6.42	13.4	19.8	–
Hydromorphic	^{137}Cs	7.39	23	8.53	10.1	34.2	67	–
	^{90}Sr	2.13	5.67	8.76	6.7	19.6	18.6	–

* Numerator: needles of the current year; denominator: old needles (2-3 years old)

Soil hydrology (moisture regime). Moisture regime is known to be one of the most influential factor of the element (nutrient and pollutant) transfer from soil to plants [8, 18, 31, 59, 82, 345].

The plants inhabiting moisture environments (topographical depressions, slope basements, river terraces, and other hydromorphic areas) accumulate considerably more radionuclides compared to the same species inhabiting relatively dry environments (convex forms of local topography, watershed ridges, and other automorphic areas). The differences between $TF^{137}Cs$ may reach a factor of 70 (Table 31, Fig. 24), which is due to a synergetic effect of several factors:

(1) Higher biological availability of ^{137}Cs in the hydromorphic soils of accumulative landscapes. Usually, the soils of such landscapes are enriched with organic matter or/and possess a peat layer where ^{137}Cs can hardly be adsorbed irreversibly. Thus, a considerable proportion of radiocaesium leached from the fallout particles remains potentially available for plants.

(2) Higher intensity of plant growth under a favourable moisture regime (Fig. 24). The plants under optimal conditions are known to accumulate more ^{137}Cs [89].

(3) Lateral inflow of mobile caesium to the accumulative areas or landscapes from adjacent eluvial areas, and development of the gley process. This provides a higher mobility of nutrients, most of trace elements, and ^{137}Cs [132, 133].

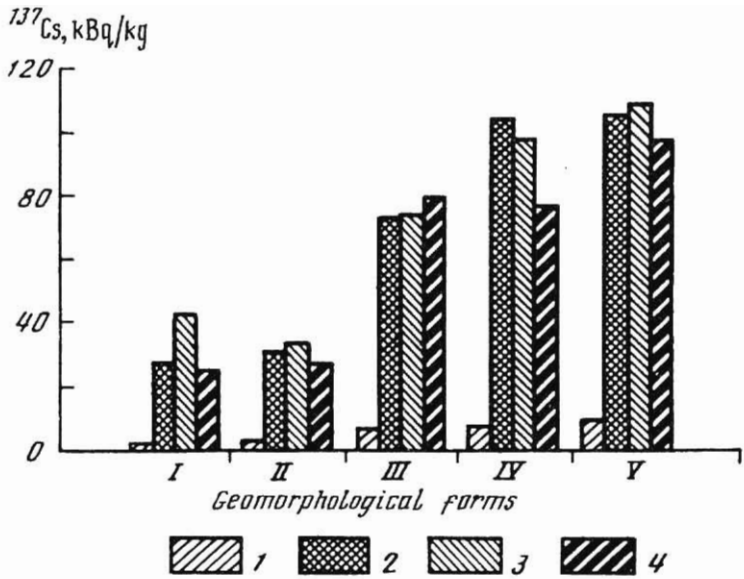

Fig. 24. ^{137}Cs in various pine organs and components depending on geomorphological forms:

(I) watershed, (II) upper part of the watershed slope, (III) terrace slope, (IV) terrace basement, (V) central area of the adjacent depression. Organs and components: (1) wood, (2) inner bark (bast), (3) needles and twigs of current year, (4) cones

$TF^{90}Sr$ to tree plants is 2–10 higher than $TF^{137}Cs$ in the automorphic areas and 4–40 times lower in the hydromorphic areas. The effect of landscape on ^{90}Sr accumulation is less significant and not as clear as on ^{137}Cs (Table 31). Our data suggest that the observed diapason of moisture conditions had no statistically significant effect on $TF^{90}Sr$ at the landscape or mesotopography scale, though some general trends may be outlined in the latter case (Fig. 25).

Soil type. The investigated soils may be ranked into the following series by $TF^{137}Cs$: peat-gley > peat-podzolic > soddy-podzolic > podzolized chernozems (Table 32). The explanation for the differences in TF between the soil types is in part discussed above.

In hydromorphic soils, ^{137}Cs availability increases in proportion with thickness and manifestation of the organic layer (carbon content). Alluvial soils are characterised by 2–3 times lower $TF^{137}Cs$ compared to other hydromorphic soils. This is due to lower acidity and higher content of competing nutrient (potassium) caused by specific pedogenesis of these soils, annual flooding, and higher clay content [76]. This results in higher irreversible adsorption and lower availability of ^{137}Cs in the alluvial soils.

Soddy-podzolic sand soils common in Bryansk and Kaluga regions exhibit the highest TF of all automorphic soils. Minimum TF is characteristic of chernozems. This is due to clayey consistence of these soils, and a high content of clay minerals capable of irreversible adsorption of caesium. In addition, abun-

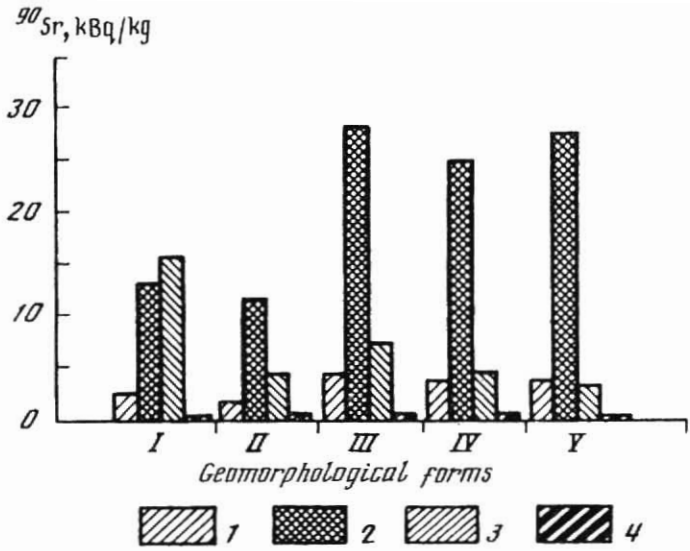

Fig. 25. ^{90}Sr content in various pine organs and components depending on geomorphological forms:

(I) watershed; (II) upper part of the watershed slope; (III) terrace slope; (IV) lower part of the terrace slope; (V) central area of the adjacent depression. Organs and components: (1) wood, (2) inner bark (bast), (3) needles and twigs of current year, (4) cones

dant soil fauna mixes the soil actively and promotes fast radionuclide migration down the soil profile, which reduces the radionuclide content in the root-abundant upper soil layers [280].

Type of vegetation is another important factor affecting $TF^{137}Cs$ (Table 33). In the mono-dominant stand (pure pine, spruce, etc.) $TF^{137}Cs$ to arboreal vegetation is usually 5–10 times higher than in mixed, coniferous-deciduous forests where TF values are minimal. This may be explained from a lower acidity and

Table 32. $TF^{137}Cs$ (Bq/kg_{plant} : kBq/m^2_{soil}) depending on the soil type ($n \cdot 10^{-3}$ m^2/kg, 1992)

Soil type	Organs and components						
	Wood	Bark		Branches		Leaves (needles)	Cones
		Inner	External	Large	Small		
Podzolized chernozem	0.02	0.32	0.95	0.17	0.31	0.47/0.14*	No data
Soddy-podzolized	0.53	6.81	2.69	1.5	2.24	7.83/2.01	4.73
Peat-podzolic, gleyic	3.4	22	9.5	6.1	14	39.0/9.5	No data
Peat-gley	3.96	44	10.9	7.97	17.5	48.6/14.3	37.84

* Numerator: needles of current year; denominator: old needles (2–3 years old)

Table 33. $TF^{137}Cs$ (Bq/kg_{plant} : kBq/m^2_{soil}) to the tree organs and components in various phytocenoses ($n \cdot 10^{-3}$ m^2/kg)

Soil type	Organs and components						
	Wood	Bark		Branches		Leaves (needles)	Cones
		Inner	External	Large	Small		
Mixed, broad-leaved-pine forest	0.07*	0.56	1.14	0.26	0.4	1.11/0.175**	–
	0.17	2.1	2.49	0.53	0.81	2.73/0.91	–
Pure pine forest	0.53	6.81	2.69	1.52	2.23	7.71/2.01	4.68
	0.9	5.99	9.36	3.75	4.45	11.4/2.3	–

* Upper line – min, lower line – max
** Numerator: needles of current year; denominator: old needles (2–3 years old)

Table 34. ^{137}Cs content in various organs and components of the investigated tree species. Mixed, broad-leaved-pine forest (kBq/kg, dry weight, automorphic areas, average deposition is 370–555 kBq/m^2)

Tree species	Organs and components						
	Wood	Bark		Branches		Leaves (needles)	Cones
		Inner	External	Large	Small		
Pine	0.05	0.51	0.65	0.18	0.25	0.8/0.11*	–
Spruce	0.03	0.63	2.76	2.16	2.79	1.04/0.46	0.77
Birch	0.06	0.21	2.56	0.41	0.62	0.74	0.80
Oak	0.18	0.69	7.77	1.54	0.68	1.48	–
Aspen	0.23	2.10	5.59	1.25	1.75	2.70	–
Teil	0.07	0.19	5.07	0.47	0.13	0.21	–
Alder	0.56	2.15	3.64	1.39	2.31	2.64	–

* Numerator: young needles of current year; denominator: old needles (2–3 years old)

ity and much higher content of Ca, K, and other nutrients in the soils of mixed forests compared to coniferous forests. Thinner forest litter of mixed forests promotes radionuclide migration to the mineral soil layers that are most capable of the irreversible adsorption of caesium and, correspondingly, its low availability and root uptake. On the other hand, the soils under pure coniferous (pine or spruce) cover are leached and depleted of potassium and Ca, and have much higher pH in the upper layers [33]. Higher radionuclides proportion in these ecosystems is retained in the forest litter, which promotes ^{137}Cs involvement in the biological cycle.

Table 35. The effect of the tree age on ^{137}Cs content in various tree organs and components (kBq/kg, dry weight, ^{137}Cs deposition is 370 kBq/m^2).

Age (years)	Organs and components						
	Wood	Bark		Branches		Leaves (needles)	Cones
		Inner	External	Large	Small		
				Pine			
45	0.04	0.20	0.39	0.03	0.09	0.29/0.06*	0.16
12	0.10	0.57	0.45	0.14	0.31	0.84/0.23	No data
				Birch			
45	0.12	0.30	0.22	0.56	0.43	2.75	No data
12	0.59	1.70	0.32	1.45	2.62	2.89	No data

* Numerator - young needles of current year, denominator - old needles (2-3 years old)

Plant species are known to have a very different ability for accumulation or discrimination of a wide range of chemical elements including radionuclide [10, 57, 161, 305]. However, species-dependent variation of $TF^{137}Cs$ to arboreal vegetation is often masked by spatial variability of this index due to initial distribution of the fallout, and heterogeneity of soil properties and regimes in the habitat. $TF^{137}Cs$ to the same tree species may vary by two orders of magnitude depending on moisture conditions, whereas the interspecies difference rarely exceeds the factor of 2–3. In general, species-dependent differences in the radionuclide accumulation by trees may be caused by a combined effect of specific physiological features (plant selectivity for several elements) and the bark surface morphology (Table 34). It may be supposed that the observed species-dependent difference in $TF^{137}Cs$ is caused by the individual factors in each case. For example, in the case of oak stand, it is likely caused by a higher requirements of oak for potassium (direct species effect), and in alder forest the most probable factor is specific moisture regime in the alder habitat (indirect species effect). The latter is confirmed by the fact that ^{137}Cs content in alder wood is low compared to its content in birch and pine growing in the same hydromorphic habitat.

Under the same conditions, the species possessing a rough or fractured bark surface (oak, aspen, and teil) tend to accumulate (retain) ^{137}Cs in the external bark [306]. Minimum relative content of the radionuclide in the external bark (apparent $TF^{137}Cs$) is characteristic of pine, which is likely due to intensive natural removal (desquamation) of its upper, most contaminated bark layers.

In general, the $TF^{137}Cs$ to the investigated tree species may be ranked as follows: aspen > oak > birch > pine > lime > spruce.

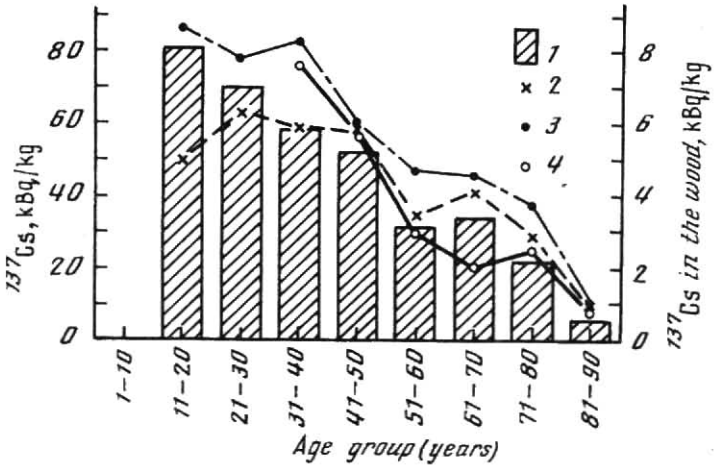

Fig. 26. ^{137}Cs content in the pine components and organs depending on the stand age: (1) wood, (2) inner bark, (3) needles and twigs of current year, (4) cones

Stand age. For almost all tree components (except for external bark) contamination and TF^{137}Cs to young trees are higher compared to the mature and aged trees (Table 35, Fig. 26). The differences may be as high as 3–8 times depending on the compared species and structural component (organ).

This is apparently due to higher intensity of physiological processes and higher weight contribution of actively growing tissues in the young trees compared to mature and old trees. Chemical elements (including radionuclides) are known to be absorbed primarily by meristem. In addition, young trees possess usually more superficial root systems, and a high proportion of sucking roots is attributed to the most contaminated top soil layers.

We calculated a reliable correlation between the age class of a pine stand (*Pinus sylvestris*) and the radionuclide content in various tree components and organs (Table 36, Fig. 27). The same dependence was calculated for the pair ^{137}Cs–stem diameter. In general, the effect of age and thickness on ^{137}Cs accumulation in the tree components is expressed by a general equation as follows:

$$y = e^{a-bx},$$

where a, b, and x are the empiric coefficients for pine different organs and components. The closest correlation between the above-mentioned valuation indices (age and thickness) and ^{137}Cs content is characteristic of wood, large branches, needles of the current year, and generative organs. The correlation is of less significance for such components as external bark, "old" needles, and internal bark (Table 36, Fig. 27).

^{90}Sr and ^{137}Cs content in the tree components varies depending on the stand age (Fig. 28). Usually, the dependence is well approximated by a general equation

$y = e^a \cdot x^b$, (Table 36 and Fig. 27)

Table 36. Regression equations for ^{137}Cs and ^{90}Sr content in various organs and components of *Pinus sylvestris* (Y[nCi/kg]) depending on the tree age (X[years])

Component	Regression	r	R^2	σ^2	m
	^{137}Cs				
Wood	$Y=e^{(6.16-0.032 \cdot X)}$	–0.90	80.69	5.48	0.42
Inner bark (bast)	$Y=e^{(7.96-0.021 \cdot X)}$	–0.80	63.40	2.88	0.42
External bark (cork)	$Y=e^{(7.30-0.0088 \cdot X)}$	–0.76	58.28	0.56	0.20
Large branches	$Y=e^{8.81} \cdot X^{-0.87}$	–0.95	90.66	2.05	0.18
Small branches	$Y=e^{(7.07-0.022 \cdot X)}$	–0.81	66.21	3.03	0.41
"Old" needles	$Y=e^{(6.99-0.023 \cdot X)}$	–0.82	67.70	3.41	0.43
Young (current year) needles	$Y=e^{(8.37-0.024 \cdot X)}$	–0.87	75.91	3.22	0.36
Cones	$Y=e^{(9.05-0.040 \cdot X)}$	–0.95	89.40	3.16	0.29
	^{90}Sr				
Wood	$Y=e^{11.39} \cdot X^{-1.76}$	–0.99	97.28	7.79	0.19
Inner bark (bast)	$Y=e^{12.13} \cdot X^{-1.50}$	–0.97	93.13	5.95	0.26
External bark (cork)	$Y=e^{11.74} \cdot X^{-1.49}$	–0.91	82.71	6.58	0.44
Large branches	$Y=e^{11.83} \cdot X^{-1.67}$	–0.97	93.77	7.32	0.28
Small branches	$Y=e^{11.81} \cdot X^{-1.58}$	–0.97	93.36	6.54	0.27
"Old" needles	$Y=e^{12.68} \cdot X^{-1.86}$	–0.94	87.65	9.70	0.45
Young (current year) needles	$Y=e^{9.62} \cdot X^{-1.03}$	–0.79	63.13	4.16	0.51
Cones	$Y=e^{(8.47-0.052 \cdot X)}$	–0.96	92.83	0.18	0.29

Footnote: r – correlation coefficient; R^2 – confidence level; σ^2 – standard square deviation; m – standard error of correlation

The correlation between the investigated parameters and ^{90}Sr content in the tree organs is even higher than ^{137}Cs. The only exclusion is generative organs (cones), which exhibit minimum correlation between the stand age and ^{90}Sr content. This is likely due to ^{90}Sr discrimination by these components.

Climate and weather conditions. The correlation analysis reveals a reliable correlation of $TF^{137}Cs$ with the rainfall during the growing season (Table 37). The closest correlation was observed in the coniferous forests with the unpronounced TF trend where the radionuclide leaching from the fallout particles was generally compensated by the irreversible adsorption by the soil solids. Somewhat lower, yet reliable correlation between the weather conditions and the radionuclide content was observed in the broad-leaved and mixed forests of the near zone. In the middle and marginal parts of the exclusion zone (15–30 km from ChNPP), the corresponding correlation is practically insignificant for all tree components but barked wood.

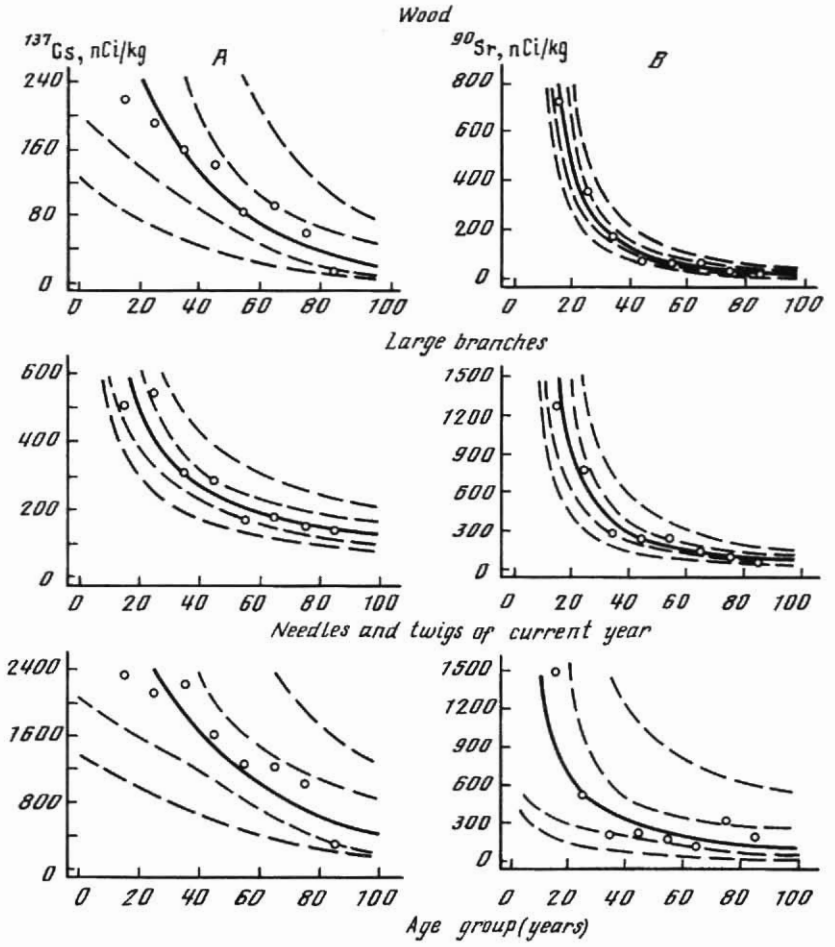

Fig. 27. The regression curves of (A) ^{137}Cs and (B) ^{90}Sr content in the pine organs and components depending on the tree age

The above-mentioned dependence is practically unpronounced in the hydromorphic areas, which is apparently due to optimal (sub-optimal) moisture regime during the complete vegetative season, independently on rainfall.

The closest correlation between a rainfall and $TF^{137}Cs$ was observed in those tree components contaminated via the root uptake (wood, alive bark, and needles of current year). On the contrary, minimum and practically insignificant correlation is typical of those components exposed to the external contamination. For the ecosystems in the near zone, however, the correlation in these components is also quite high.

Thus, biological availability of ^{137}Cs and ^{90}Sr and *ergo* their accumulation by the stand depend on a complex combination of factors. In our opinion, the following factors influence most accumulation of ^{137}Cs by arboreal vegetation:

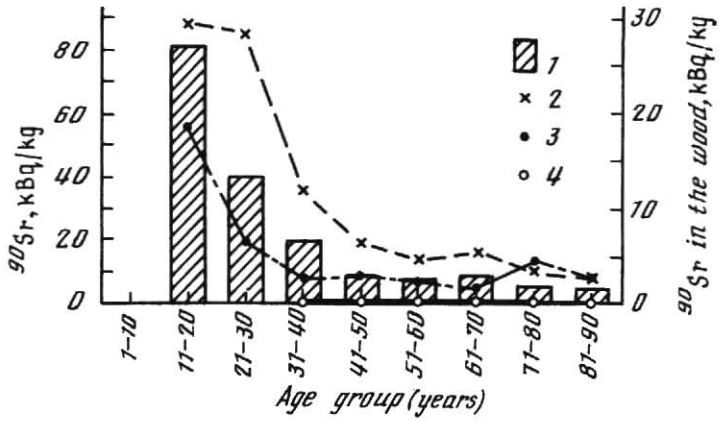

Fig. 28. ^{90}Sr content in the pine components depending on the tree age:
(1) wood; (2) inner bark (bast); (3) needles and twigs of current year; (4) cones

(i) soil type and (ii) its moisture regime. Less significant, though of importance are (iii) type of phytocenosis and stand age. Species composition (iv) is of least significance and difficult to trace on the background of other factors. The average accumulation of ^{137}Cs by trees depending on the above-mentioned factors varies by a factor of 100, 10, 4 and 1.5, respectively. The effect of all these factors but age on the ^{90}Sr accumulation in the tree canopy is of less significance.

The Contribution of Various Tree Organs and Components to Total Radionuclide Inventory in the Stand

As discussed above, the initial contribution of arboreal vegetation to the total contamination of the ecosystem was 60–90%, with the most contribution of those tree components directly exposed to the fallout and the secondary contamination (external bark, branches, and assimilative organs). In the subsequent 2–3 months, the intensive natural mechanical and biological decontamination resulted in replacement of 80% of the contaminants to the soil surface. As small as 5% of the initial radionuclide inventory remained in the tree canopy one year after the accident. In the course of time, the increasing root uptake of the radionuclides makes their accumulation in the plants progressively dependent on various physiological and the site-specific environmental parameters. Ten years after the accident, arboreal vegetation contained from 1–3% to 26% of the total deposition including soil. The average value of this index over the entire investigated territory varies from 2% to 4%. The maximum accumulation is characteristic of pine forests of 30–40 years old, growing on hydromorphic peat soils, in the remote zone (more than 50 km from ChNPP) [36, 73, 89, 135, 277–280, 337]. Almost all researchers agree that the highest radionuclide inventory in the contaminated stand shall be attributed to the wood. However, it should not be necessary true in all cases. The relative distribution of radionuclides among the tree components, and the radionuclide inventory in the stand depend both on the growing conditions and the species-dependent physiological features, and may vary both in time and space.

Table 37. Correlation between the rainfall during the vegetative period and $TF^{137}Cs$ from soil to various pine organs and components (average of 6 years)

Organs and components	Correlation coefficient *(r)*
Mixed, broad-leaved-pine forest, marginal part of the 30 km exclusion zone, eluvial landscape, automorphic area (key site D-1)	
Barked wood	0.6
Inner bark (bast)	0.16
External bark (cork)	0.03
Needles of current year	0.13
"Old" needles	0.02
Small branches	0.03
Alder forest, marginal part of the 30 km exclusion zone, accumulative landscape, hydromorphic area (key site D-3)	
Barked wood	0.06
Inner bark (bast)	0.06
External bark (cork)	0.04
Needles of current year	0.21
"Old" needles	0.04
Small branches	0.09
Pine forest with mixed herb/moss cover, middle part of the 30 km exclusion zone, transit-accumulative landscape, automorphic area (key site K-2)	
Barked wood	0.84
Inner bark (bast)	0.89
External bark (cork)	0.01
Needles of current year	0.76
"Old" needles	0.17
Small branches	0.03
Mixed, broad-leaved-pine forest, "near zone", eluvial landscape, automorphic area (key site Sh-1)	
Barked wood	0.61
Inner bark (bast)	0.56
External bark (cork)	0.52
Needles of current year	0.52
"Old" needles	0.44
Small branches	0.6

The most radionuclide inventory is attributed to the wood in the territories characterised by high TF, i.e. in the ecosystems on hydromorphic and semi-hydromorphic soils or in the "near zone" characterised by an extended period of radionuclide leaching from the fallout particles to the root-abundant soil layer. In the remote zone (particularly in the automorphic areas), the most portion of

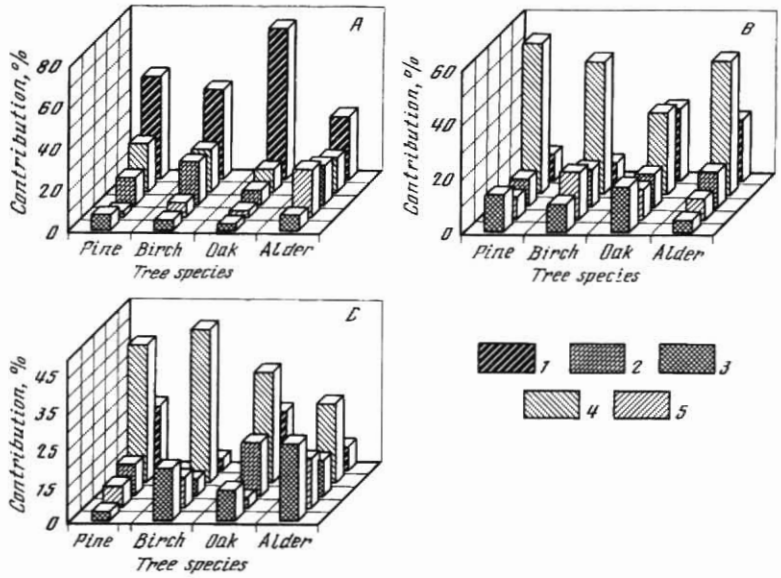

Fig. 29. Contribution of various tree organs and components to the radionuclide inventory in the tree canopy in (A) eluvial and (B) accumulative landscapes in the exclusion zone, and (C) in the "near zone" (average for 1988-1992):

(1) external bark (cork); (2) leaves and needles; (3) inner bark (bast); (4) wood; (5) branches

radionuclides was concentrated in the external bark until 1992 (Fig. 29). The contribution of this component increases as the clay content in the soil increases and reaches its maximum on loamy chernozems. The absolute value of this index depends also on the bark surface: the most contribution is characteristic of rough, deep-fractured oak bark (Fig. 29).

The dynamics of the contribution. The dynamics of individual contribution of each tree component to the total radionuclide inventory in the arboreal vegetation is shown in Fig. 30. It is clear that on hydromorphic and semi-hydromorphic soils and in the near zone, the wood contribution increases in time, which is undoubtedly due to higher $TF^{137}Cs$ and increasing role of the root uptake. On automorphic sand soils, the contribution of external components (exposed to the initial fallout) still continues to increase, in spite of insignificant rate of aerial contamination. The latter is explained by a monotonous decrease in the radionuclide root uptake in such territories because of active irreversible adsorption of ^{137}Cs in the automorphic soils. On this background, the annual variation of caesium dynamics is caused by the climatic features of individual years. In the extremely dry years, the contribution of external bark to the radionuclide inventory increases, and in wet years the contribution of the "root-dependent" components (primarily wood) increases and that of external bark decreases. In other words, the contribution of the "fallout-exposed" tree components increases as the ^{137}Cs root uptake decreases. This demonstrates high mobility and variability of ^{137}Cs inventory in the wood in spite of cumulative dynamics of the wood

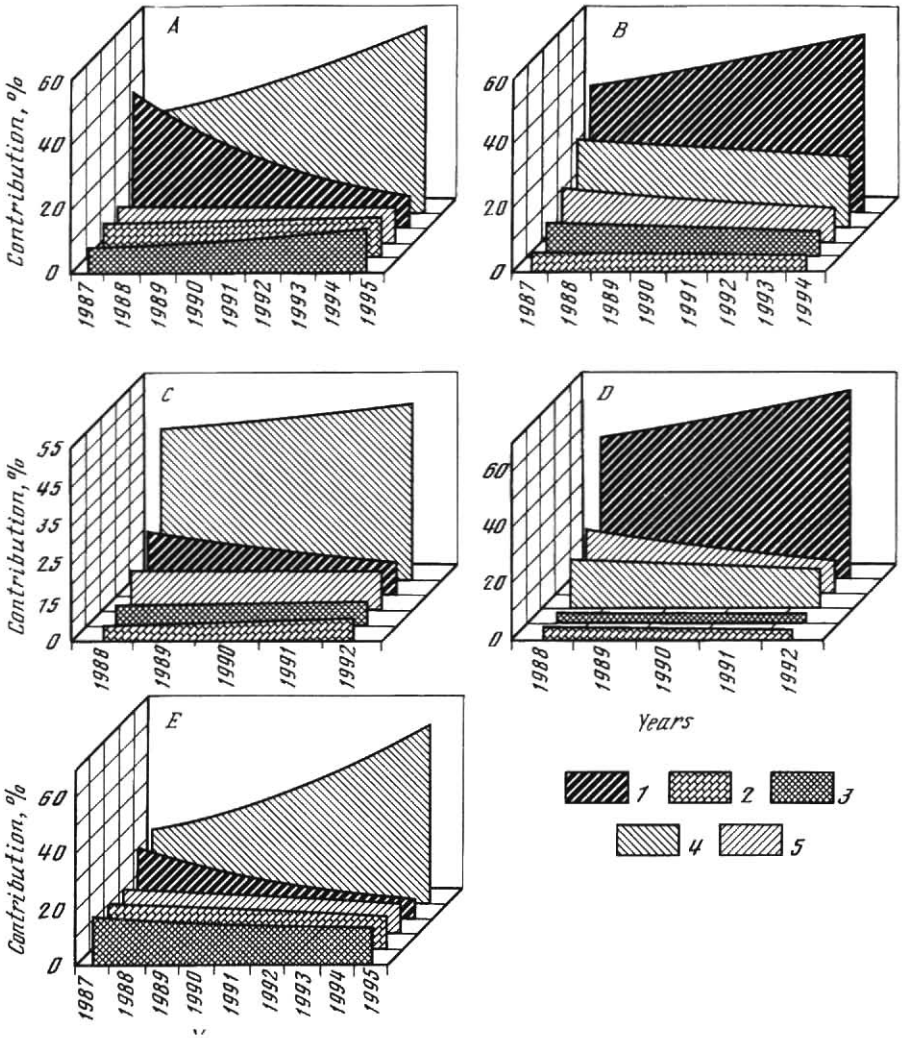

Fig. 30. The dynamics of the contribution of various tree organs and components to the ^{137}Cs inventory in the tree canopy (exponential approximation):

(A) on peat-gley soil (accumulative landscapes); (B) on podzolic, sandy soils (eluvial landscapes); (C) on soddy-podzolic, sandy, gleyic soils (transit-accumulative landscapes); (D) on podzolic clayey chernozems; (E) on weak-podzolic, sandy soils of eluvial landscapes in the exclusion zone. Components: (1) external bark (cork); (2) leaves and needles; (3) inner bark (bast); (4) wood; (5) branches

phytomass. In hydromorphic (semi-hydromorphic) environments and in the "near zone", the relative ^{137}Cs content in the tree components may be ranged as follows: wood > bark > branches > assimilative organs. In automorphic areas, the order is different: bark >> wood > branches > assimilative organs. The highest proportion of inner bark to the total ^{137}Cs inventory was observed in aspen

stands, which is in correlation with a high proportion of this component in the total phytomass of this species. The maximum ^{137}Cs content in the assimilative organs is characteristic of oak.

Thus, the contribution of individual structural components to the total inventory of ^{137}Cs in the tree canopy depends primarily on soil properties and regime. Wood contains a larger proportion of radionuclide in (i) hydromorphic and semi-hydromorphic areas, and (ii) in the near zone with a long-term radionuclide leaching from the fallout particles. In the ecosystems on automorphic soils, the wood contribution to the total contamination of the tree canopy is lower that the external bark, in spite of much higher wood biomass. This is particularly pronounced on clayey chernozem soils. Parameterization of ^{137}Cs fluxes among the stand components is, therefore, impossible without differentiation of the environments by the factors of ^{137}Cs transfer to each component and dynamics of these factors.

3.2 RADIONUCLIDE BEHAVIOUR IN THE UNDERSTOREY

The role of understorey as a factor of biogeochemical migration of nutrient and pollutants is often underestimated because of its insignificant phytomass compared to arboreal vegetation. Nevertheless, physical and chemical soil properties, and regimes (especially in upper layers) are known to depend, to a large extent, on the understorey composition and structure [103]. Many forest shrubs and herbaceous species are commonly used by local population as a food source and in the everyday practice (for fuel, construction, etc.). That is why the understorey became an important object of this comprehensive study.

3.2.1. Radionuclide Composition and Main Features of ^{137}Cs Behaviour in the Understorey

The field data suggest that the rate of natural decontamination in the herb layer should be 3 times more intensive compared to arboreal vegetation [8]. This is due to apparent differences in the life cycle of long-lived tree species and short-lived (annual and biennial) forest herbs. On the other hand, herbaceous vegetation is under the tree cover, which makes it exposed to the secondary contamination by the fallout particles removed from the trees and various fragments of the contaminated trees (twig pieces, bud and bark squamae, etc.). This input may prolong surface decontamination of the grass and shrubs, which has a considerable effect on the radionuclide behaviour in general.

This is confirmed by longer presence of ^{144}Ce, ^{95}Zr, ^{95}Nb, and ^{106}Ru in the radionuclide composition of the understorey compared to the overstorey. These radionuclides are known to indicate surface contamination of biological objects [280], and have been observed in the understorey for 4–5 years after the accident (Fig. 31). In the ensuing years, the radionuclide composition of herbaceous and shrub species contained ^{137}Cs and ^{90}Sr only, in spite of a significant proportion of other radionuclides in the soil.

Species-dependent variation of the radionuclide content in the investigated herbaceous species is higher than in the tree species, which may be explained

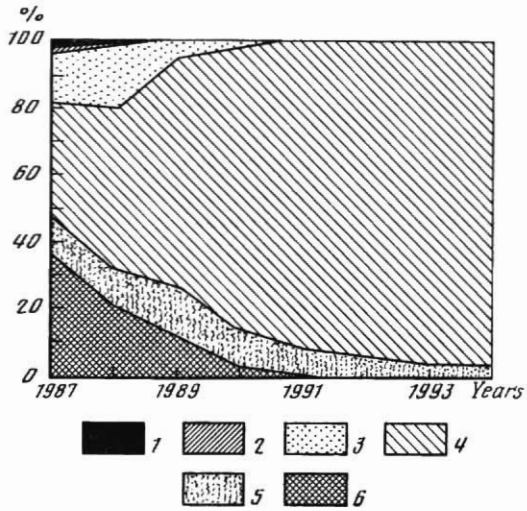

Fig. 31. Averaged dynamics of the radionuclide composition of forest herbaceous plants (30 km exclusion zone): (1) ^{95}Zr; (2) ^{95}Nb; (3) ^{106}Ru; (4) ^{137}Cs; (5) ^{134}Cs; (6) ^{144}Ce

by (i) limited nutrition area of grasses and shrubs compared to trees, which results in less "averaging" of TF depending on spatial variability of the moisture conditions over the key site; and (ii) higher species diversity of herbaceous vegetation, even within a limited area of the key sites.

Variability of ^{137}Cs accumulation by different species (within the same experimental site) is about 45%, it has a pronounced seasonal dynamics: it increases during the growing season from spring to summer and decreases by autumn (Table 38). Seasonal variation makes it difficult to develop a clear and correct classification of plant species by their accumulative capacity [84, 85, 100, 175], in spite of the obvious species-dependent features of ^{137}Cs accumulation. It is principally possible, however, to compare the weighted-average con-

Table 38. Variation of the ^{137}Cs content in herbaceous vegetation (Ci/kg, dry weight, key site D-1, integrated sample for 1989–1993)

Year	Month	Statistical indices						
		n	m	±m	G	V, %	max	min
1989		10	1.39	0.2	0.63	45.8	2.41	0.48
1993	May	7	0.33	0.05	0.15	46.4	0.52	0.09
	June	7	0.28	0.06	0.17	61.5	0.67	0.11
	July	7	0.35	0.1	0.28	79	1	0.08
	August	7	0.43	0.07	0.19	43.4	0.67	0.1
	September	7	0.56	0.12	0.33	58.5	1.25	0.15
	October	7	0.34	0.06	0.16	47.9	0.56	0.11

Footnote: n – sample; m – mean; G – standard deviation; ±m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

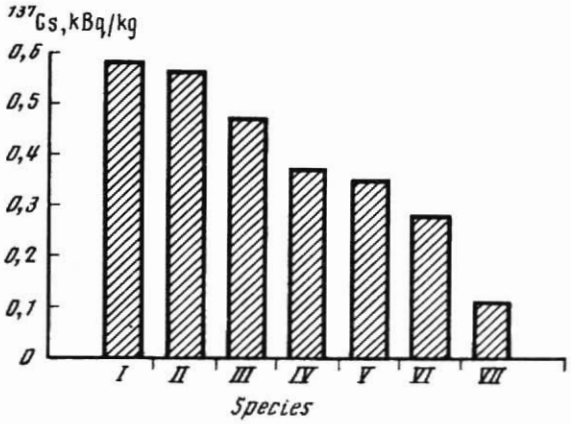

Fig. 32. ^{137}Cs content in various herbaceous species sampled in the same ecotope (weighted average for the vegetative period, key site D-1, 1993).
Species: (I) *Convallaria majalis*, (II) *Pteridium aqnilinum*, (III) *Majanthemum bifolium*, (IV) *Rubus saxatilis*, (V) grasses (bulk sample), (VI) *Polygonatum officinale* (VII) *Peucedanum officinale*

centrations of ^{137}Cs in the phytomass of various herb species for the entire growing period (Fig. 32). Such an index makes it possible to pool the species growing under the same conditions into two basic groups: caesium-concentrators and caesium-discriminators (in accordance with the biogeochemical classification by Perelman [179]).

The field data suggest that May lily (*Convallaria majalis*) and bracken (*Pteridium aquilinum*) should be among the most active concentrators: the weighted-average ^{137}Cs content in these species exceeds that in the other species by a factor of 2-10 (Fig. 32). These data are in agreement with the data by other authors [57, 82, 215]. The minimum accumulative capacity is characteristic of hog fennel (*Peucedanum orioselenium*) and Solomon's seal (*Polygonatum officinale*) which accumulate almost ten times less ^{137}Cs than bracken. In general, species-dependent differences in the radionuclide accumulation by herbs and shrubs do not exceed one order of magnitude.

The growth conditions have a considerable effect on the radionuclide accumulation by herbaceous plants and its dynamics. An increasing trend in the dynamics of radionuclide content is characteristic of those plants inhabiting moist ecotopes. Particularly high $TF^{137}Cs$ is typical of the so-called "hydrophilic" species, such as iris (*Iris pseudacorus*) and sage (*Cyperaceae gen.*). Under the same deposition, these species accumulate by two orders of magnitude more radionuclides than other effective concentrators (*e.g., Convallaria majalis*) in the automorphic area (Fig. 33).

Moss and lichen are known to be the most effective concentrators of ^{137}Cs (Table 39). Unlike higher plants, mosses and lichen possess specific properties enabling them to accumulate a considerable amount of chemical elements. They are very sensitive to any change in the radionuclide content in the soil or other substrate, which makes them the admitted indicators of environmental pollution by radionuclides and heavy metals [27, 121, 162, 301, 309]. Mosses may accumulate 10 times more ^{137}Cs compared to any plant species, and 2-3 times more than lichen. A considerable proportion of ^{137}Cs is, as a rule, concentrated in the

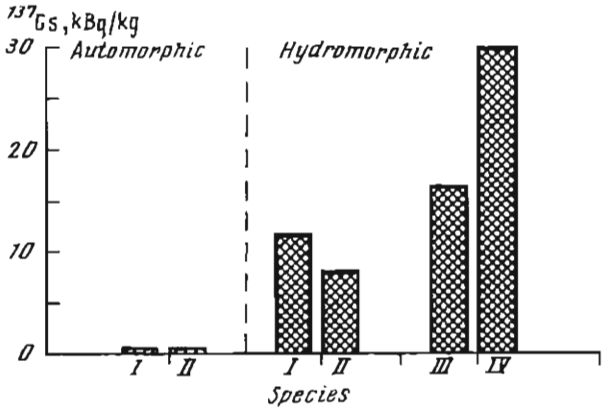

Fig. 33. ^{137}Cs content in herbaceous plants under contrast moisture regimes. Species: (I) grass (bulk sample), (II) *Rubus saxatilis*, (III) *Iris pseudacorus*, (IV) sedge (bulk sample)

dead part of a moss plant. ^{137}Cs content in club moss (*Lycopodium gen.*) is almost the same as in ferns. Flowering plants are the least effective concentrators of all taxonomic groups constituting the understorey. Therefore, the moss (lichen) cover serves as an effective biogenic barrier to retain radionuclides and prevent them from vertical and lateral migration.

Thus, the weighted-average radionuclide content in plants is a useful tool to characterise the accumulative capacity of various plant species for radionuclides. Classification of herbaceous plants by this index is of high significance for the purposes of environmental monitoring. The so-called discriminators may be of interest in terms of agricultural land use in the contaminated territory, whereas the concentrators may be used for the purpose of monitoring, phytoremediation, and as conservative indicators to estimate the maximum possible radionuclide content in the agricultural produce obtained from the contaminated territories.

Table 39. ^{137}Cs content in the moss/lichen cover and herbaceous vegetation in the same ecotope ("near zone", key site Sh-5, 1991)

Species	^{137}Cs, kBq/kg	Species	^{137}Cs, kBq/kg
Mosses		*Lichen*	
Dicranum rugozum:		*Cladina arbuscula*	407
Alive tissues	703	*Grass cover*	
Dead tissues	1073		
Pleurosiun schreberi:		*Lycopodium clavatum*	344.1
Alive tissues	425.5	*Filicales*	332
Dead tissues	740	Integrated grass/cereal sample	146.2
Polytrichum juniperinum (alive tissues only)	703		

3.2.2. The Dynamics of Radionuclide Content in the Understorey

The content of any chemical element in plants varies by time, which is true for radionuclides as well [137, 230, 342].

Seasonal dynamics. ^{137}Cs content in wild herbaceous species varies over a wide range. The species-concentrators are characterised by the widest variation range and the maximum variation coefficient, and for discriminators these values are minimal (Table 40). The radionuclide content, variation, and dynamics in the understorey depend on both species and growth conditions.

In hydromorphic areas, ^{137}Cs content in most species decreases from spring to autumn by a factor of 5–10 (Fig. 34). Such dynamics is most pronounced for stone bramble (*Rubis saxatilis*); other species also exhibit similar seasonal dynamics with a general decreasing trend and more or less manifested variation. The same general dynamics of TF to herbaceous species is characteristic of some stable macroelements (potassium) and trace elements [186].

In automorphic areas, ^{137}Cs content in the herbaceous plants increases during most of a growing season, and starts to decrease as late as by the end of a vegetative season due to radionuclide leaching from the dead plants (Fig. 35). Such a dynamics is typical of many other trace elements as well [20].

In some species, the absolute maximum of ^{137}Cs varies from July to September, which is likely due to their long lasting vegetation and life cycle (Fig. 36). For example, *Convallaria majalis* (blossoming in late May) accumu-

Table 40. Variation of ^{137}Cs content in some herbaceous species (kBq/kg, the exclusion zone, 1993)

Species	Statistical indices					
	m	±m	G	V, %	max	min
Automorphic area (key site D-1)						
Cereals (mixed sample)	0.35	0.05	0.13	36.8	0.52	0.19
Convallaria majalis	0.58	0.09	0.24	40.6	1	0.26
Peucedanum officinale	0.14	0.01	0.02	20.7	0.15	0.08
Polygonatum officinale	0.28	0.05	0.12	42	0.48	0.16
Pteridium aqnilinum	0.14	0.35	62.4	1.25	0.24	0.14
(Majanthemum bifolium)	0.45	0.09	0.22	48.2	0.78	0.25
Hydromorphic area (key site D-3)						
Cereals (mixed sample)	11.53	1.43	3.51	30.4	18.13	6.66
Convallaria majalis	18.62	3.1	7.6	40.8	31.45	6.66
Rubus saxatilis	7.91	3.03	7.43	94	24.05	2.96

Footnote: m – mean; G – standard deviation; ±m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

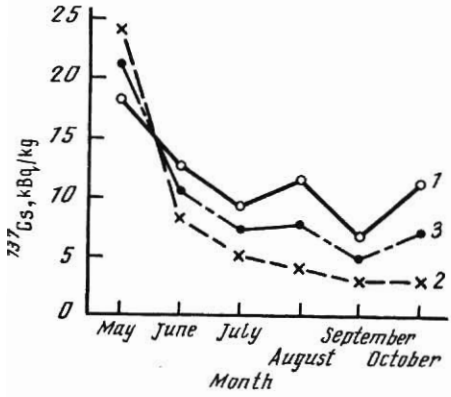

Fig. 34. Seasonal dynamics of ^{137}Cs in herbaceous plants on peat-gley soils in hydromorphic areas (key site D-3, 1993):
(1) grass (bulk sample); (2) *Rubus saxatilis*; (3) total (bulk sample)

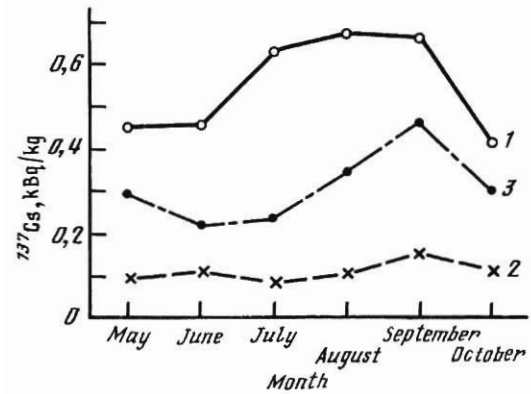

Fig. 35. Seasonal dynamics of ^{137}Cs in various groups of herbaceous plants on podzolic, sandy soils in automorphic areas (average of 1992 and 1993):
(1) "accumulators"; (2) discriminators"; (3) others

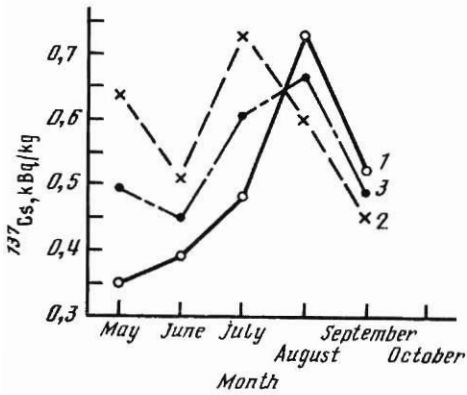

Fig. 36. Seasonal dynamics of ^{137}Cs in various herbaceous plants on podzolic, sandy soils in automorphic areas (Key site D-1, 1994):
(1) *Pteridium aqnilinum*, (2) *Convallaria majalis*, (3) bulk sample

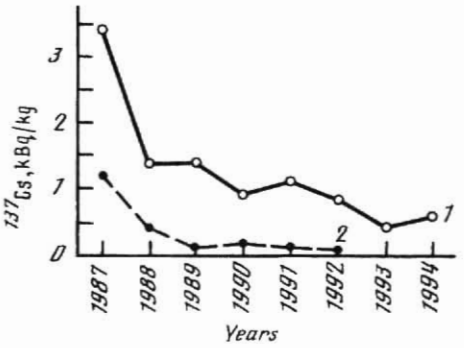

Fig. 37. Multiyear dynamics of ^{137}Cs in herbaceous plants on podzolic, sandy soils in the automorphic areas (weighted average of dominant species):
(1) forest; (2) old-tilled meadow

lates maximum ^{137}Cs by middle July. The plants with later blossoming time have the accumulation maximum in August or September. In wet years, a vegetative period is usually longer and the maximum of the radionuclide content in herbs shifts to a later time. The above-discussed dynamics is most manifested for species-concentrators (Fig. 36).

No clear explanation can be suggested for the observed differences between the radionuclide dynamics in the hydromorphic or automorphic areas. Maybe such a dynamics reflects the corresponding changes in the chemical potential provided by the plant roots. This index depends on both the rate of phytomass increment and the moisture content in the plant [103]. The plant moisture is reported to decrease drastically in the automorphic areas and remains stable in the hydromorphic areas [277], which may affect the radionuclide uptake and accumulation by the plants.

The above-discussed seasonal dynamics of ^{137}Cs in the herbaceous plants should be taken into account when developing a monitoring strategy: the plant species with unpronounced dynamics may be sampled once in the middle of a vegetating season, and other species should be sampled several times in a season. The seasonal radionuclide accumulation in fodder species may cause a higher contamination of the dependent wildlife fauna.

The long-term dynamics of ^{137}Cs content in the understorey was in a general agreement with its dynamics in the overstorey: the radionuclide content in herbs and shrubs decreased monotonously for the first 2–3 years after the accident independently on species or environmental factors (Fig. 37).

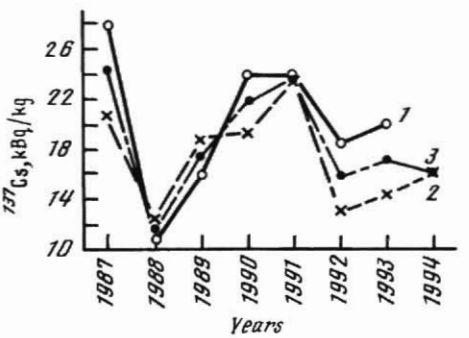

Fig. 38. Multiyear dynamics of ^{137}Cs in herbaceous vegetation in various hydromorphic areas: (1) wet forest; (2) peat bog; (3) average

In the subsequent years, the dynamics became more dependable on the soil-ecological conditions (Fig. 38). Like in arboreal vegetation, ^{137}Cs content in herbs depended on the dynamics of radionuclide availability in different environments and the species-dependent reply to the extreme conditions of 1992.

Comparison of ^{137}Cs dynamics in the herbaceous vegetation in the forest and meadow ecosystems shows the 2–3 times higher accumulation in the forest species compared to the meadow ones under the same deposition (Fig. 37). Thus, herbaceous vegetation is more significant factor of radionuclide migration in the forest than in the meadow environments.

3.3. ACCUMULATION OF RADIONUCLIDES BY HIGHER FUNGI

Higher fungi (mushrooms) are of great significance for rural population of Ukraine and the European part of Russia as a food resource and a commercial forest produce. Particular interest of (radio)ecologists in fungi is due to the exceptional capability of these organisms for concentration of many chemical elements including radionuclides.

Fungi are far much effective accumulators of ^{137}Cs than any other component of a forest ecosystem (Fig. 39). ^{137}Cs concentration in fungi is not only higher than in the plants, but sometimes exceeds the radionuclide content in the soil substrate. On the average, ^{137}Cs concentration in fungi exceeds its concentration in the most contaminated layers of forest litter by a factor of 20. The corresponding differences between fungi and wood may reach 2-3 orders of mag-

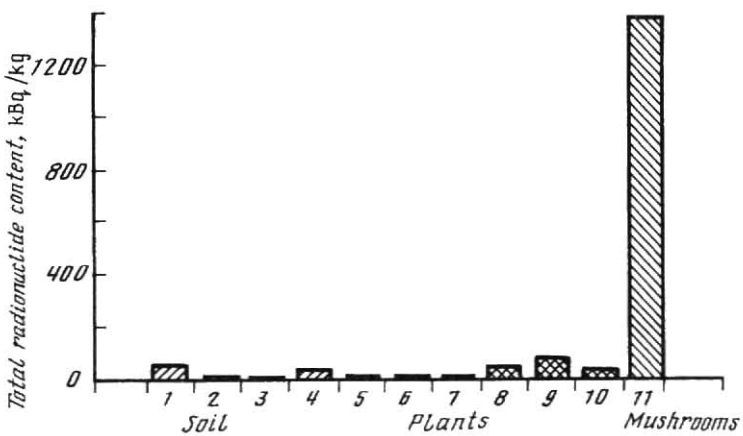

Fig. 39. Total radionuclide content in various components of forest biogeocenoses (average for 1991, "near zone"): (1) A0 (forest litter); (2) mineral soil 0–5 cm; (3) mineral soil 5-10 cm; (4) total soil (A0 + 0–10 cm); (5) pine wood; (6) needles and twigs of the current year; (7) tree canopy (weighted average); (8) herbs and grasses; (9) mosses; (10) lichen; (11) mushrooms (*Xerocomus badius*)

Table 41. Average ^{137}Cs content in forest fungi (the exclusion zone, mean ^{137}Cs deposition is 185 kBq/m^2)

Species	Trophic group	^{137}Cs, kBq/kg (mean)	Variation range
Lycoperdon perlatum	Saprotrophic	6.59	2.11–10.43
Cantharellus cibarius	Saprotrophic	6.88	3.33–11.8
Clitocybe sp.	Saprotrophic	12.06	3.44–18.87
Armillariella mellea	Xylotrophic	14.32	1.15–37
Leccinum aurantiacum	Symbiotrophic	32.08	4.26–81.4
Boletus edulis	Symbiotrophic	34.93	10.73–59.2
Amanita muscaria	Symbiotrophic	35.82	4.07–111
Lactarius necator	Symbiotrophic	52.87	8.51–199.8
Leccinum scabrum	Symbiotrophic	67.9	18.94–177.6
Russula sp.	Symbiotrophic	77.96	1.11–259
Hydnym repandum	Saprotrophic (on forest litter)	94.5	5.99–125.8
Lactarius torminosus	Symbiotrophic	163.54	7.03–256.78
Paxillus involutus	Saprotrophic/ Symbiotrophic	197.1	1.67–555
Xerocomus badius	Symbiotrophic	213.34	192.4–236.8
Lactarius rufus	Symbiotrophic	226.7	3.7–555

nitude [278]. The rate of ^{137}Cs uptake by fungi is even more intensive than potassium [313, 330].

Species is one of the main factors affecting the radionuclide accumulation by fungi *. Species-dependent variation in the accumulation of ^{137}Cs by fungi under the same deposition reaches orders of magnitude (Table 41). The highest variation is likley due to a synergetic superposition of several factors such as weather conditions, and peak of a seasonal dynamics of ^{137}Cs availability. The effect of these factors may be reduced by calculation of the long-term average $TF^{137}Cs$ for each species, which makes it possible to compare the accumulative capacity of the investigated species.

The minimum radionuclide content is characteristic of the so-called saprotrophic ecological group of fungi [311, 320, 331] (see Table 47). This is followed by several representatives of the *xylotrophic* group (e.g., *Armillaria mellea*). The most contaminated are symbiotrophic (mycorrhizal) fungi species that exhibits also the highest variability of TF. This is

* More than 200 species of higher fungi inhabit the contaminated forests in the territories of Russian Federation, Ukraine and Belarus'. The most wide-spread species belong to Rusulla, Boletus, Clitocybe, Lactarius, Amanita, and some other well-known genera [44, 104, 259]

due to a considerable variation of mycelium content down the soil profile (from A0l forest litter layer to the 50-60 cm deep mineral layers) [310, 357], whereas the maximum of ^{137}Cs content in the soil is concentrated in A0f/h layers of the forest litter [280]. The most accumulative capacity is characteristic of *Paxillus incvoluta, Xerocomus badius*, and *Lactarius necator*. These species are suggested as bioindicators of radionuclide contamination in the forest ecosystems [240, 318, 329, 343]. In the future, when ^{137}Cs migrates to deeper soil layers, some other species (for example, *Boletus edulus*) will likely become the most effective concentrator for this radionuclide. This is confirmed by the fact that *Boletus edulus* has a deeper mycelium and was reported to be the most effective concentrator for "weapon" radiocaesium [310].

Statistical analysis of the radionuclide content in various ecosystem components suggests that the coefficient of variation *(V)* of this parameter is similar to that in the vegetation (Table 42).

We believe that the *V* of ^{137}Cs accumulation in both higher plants and fungi is controlled by the same factors. These are (i) spatial distribution of the radionuclides in the soil, and (ii) soil-dependent variation in the radionuclide availability. Spatial variability of ^{137}Cs content in the soil is about 30%, which is 2 times less than the corresponding value for the above-ground vegetation and fungi. Therefore, the significance of the radionuclide availability is the same or even higher than that of the spatial distribution of the deposition. This is directly confirmed by the features of ^{137}Cs accumulation in the same fungi species under the same deposition, but different soil conditions (Table 43). The fungi species growing on peat-gley, hydromorphic soils (accumulative landscapes) accumulate ten times more ^{137}Cs than the same species on soddy-podzolic automorphic soils (eluvial landscapes).

At the same time, the coefficients of pairs correlation between ^{137}Cs concentration in fungi and basic soil properties, such as humus content,

Table 42. Variation of the ^{137}Cs content in basic components of forest biogeocenoses in the "near zone" (1991)

Components of BGC	Statistical Indices					
	m	±m	max	min	G	V, %
Forest litter (A0), Ci/km^2	103.3	1.9	270	11	32.4	31
A0+soil (0–10 cm)	123.2	1.8	282	23.1	31.3	25
Wood, nCi/kg	149.1	14.6	320	20	90	60.4
Phytomass of current year	1723.1	153.8	3900	290	996.9	57.9
Fungi, μCi/kg *(Xerocomus badius)*	372.5	52.2	740	110	209	56.1

Footnote: m – mean; G – standard deviation; ±m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

Table 43. The effect of landscape type (moisture regime) on ^{137}Cs content in the fungus species (kBq/kg, dry weight, 1990, deposition = = 185 kBq/m^2)

Species	Landscape/regime type	
	Eluvial landscape automorphic area	Accumulative landscape hydromorphic area
Lycoperdon perlatum	2.11	5.55
Armillariella mellea	1.52	37
Russula sp.	2.29	170.2
Macrolepiota procera	4.81	8.14
Lactarius necator	8.88	70.3
Clitocybe sp.	14.43	66.6
Paxillus involutus	37	777
Lactarius torminosus	70.3	62.9

exchangeable Ca and potassium, pH, etc., are insignificant or low (Table 44). This is likely due to combined effect of the soil properties or/and inadequacy of the common methods of soil sampling to characterize the whole diversity of the fungus nutrition area. At a first approximation, it is possible to assume a significant correlation between the forest litter thickness and ^{137}Cs accumulation by fungi. Other authors confirm this phenomenon pointing out that $TF^{137}Cs$ to fungi increases in proportion with the forest litter reserves [117].

No correlation was revealed between ^{137}Cs content in fungi and ^{137}Cs deposition in the near zone (Table 45). Low correlation coefficients reflect a high spatial variability of the fallout in the investigated territory [341]. The low correlation between the two indices was reported by other researchers as well [293].

Interestingly, the correlation coefficients presented in Table 45 are negative. In our opinion, such a phenomenon may be due to the effect of fungi mycelium in itself. ^{137}Cs content in the fungal fruit bodies is reported to depend on its previous accumulation in the so-called long-leaved mycelium [304]. Within a limited sampling area, this factor is likely to override the effect of spatial variability of the deposition.

The rate of nutrient and heavy metal accumulation by fungi varies depending on both a growing stage and a structural component [225, 226, 272]. The metal content is reported to be higher in young specimens compared to aged ones, and in the fungi cap compared to the stipe [19], since the metabolic processes run more intensively in the cap than in the stipe. The corresponding differences in ^{137}Cs accumulation, however, are insignificant (Table 46). There is a tendency toward its higher accumulation in the caps, confirmed by other researchers [303]. No significant differences in ^{137}Cs content in young (1–2 days old) or aged (10 days old) fruit bodies were revealed.

Table 44. Coefficient of correlation (r) between ^{137}Cs content in mushrooms and basic soil parameters

Soil parameter	r	P
Thickness of the forest litter, cm	–0.56	0,95
Humus content, %	0.25	0,9
pH of KCl extract	nc*	nc
pH of water extract	0.2	0,5
Exchangeable Ca^{2+}, mg-equiv/100	nc	nc
Mobile K_2O content, mg/kg	0.2	0,5

* No reliable correlation

Table 45. Coefficient of correlation (r) between ^{137}Cs content in mushrooms and various soil layers

Soil layer	r	P
Forest litter A0	–0.22	0,5
0–10 cm	–0.32	0,5
Total A0 + (0–10 cm)	–0.33	0,5

Table 46. ^{137}Cs in the mushroom fruit bodies of various age and in the mushroom caps and stipes (MBq/kg, dry weight, key sites K-3 and Sh-1)

Species	Component/age	Year	Radionuclide	
			^{134}Cs	^{137}Cs
Boletus edulis	Caps	1990	0.11	0.96
	Stipes		0.11	1.0
Lactarius necator	Caps	1991	0.85	8.51
	Stipes		0.74	8.88
Macrolepiota procera	Young	1990	0.01	0.08
	Old		0.01	0.08
Lactarius necator	Young	1991	1.67	17.39
	Old		1.67	17.39

Table 47. Multiyear dynamics of $TF^{137}Cs$ from the soil (forest litter) to some mushroom species, (Bq/kg) : (kBq/m^2)

Species	Year							
	1987	1988	1989	1990	1991	1992	1993	1994
Ukraine								
Exclusion zone, 6 km from ChNPP to W, eluvial landscape, key site Sh-1								
Lactarius necator	28	172	155	240	479	375	nd*	nd
Paxillus involutus	651	3621	1091	2264	1700	594	nd	1321
Russula sp.	nd	nd	200	566	2000	542	nd	92
Exclusion zone, 28 km from ChNPP to S, eluvial landscape, key site D-1								
Lactarius necator	76	95	104	48	nd	5	52	111
Paxillus involutus	506	827	77	200	96	10	182	110
Russula sp.	nd	100	98	12	nd	nd	7	nd
Exclusion zone, 28 km from ChNPP to S, accumulative landscape, key site D-3								
Lactarius necator	nd	nd	375	339	175	nd	1174	nd
Paxillus involutus	nd	nd	2500	nd	1923	1800	2174	1851
Russula sp.	nd	nd	417	821	1346	780	522	697
Russia								
200 km from ChNPP to NE, transit-accumulative landscape , key site Zl-1								
Lactarius sp.	nd	nd	1828	nd	nd	nd	nd	nd
Paxillus involutus	nd	nd	nd	nd	510	nd	nd	nd
Russula sp.	nd	nd	nd	nd	690	nd	nd	nd
260 km from ChNPP to NW, transit-accumulative landscape , key site Kl-1								
Lactarius necator	nd	313	nd	519	904	nd	nd	nd
Paxillus involutus	nd	1581	nd	3463	3132	nd	nd	nd
Russula sp.	nd	nd	1369	1175	663	nd	nd	nd
450 km from ChNPP to NW, transit-accumulative landscape, key site E-1								
Paxillus involutus	nd	nd	nd	nd	nd	43	nd	nd
Suillus luteus	nd	nd	nd	nd	nd	6	nd	nd
550 km from ChNPP to NW, transit-accumulative landscape, key site Pl-1								
Lactarius necator	nd	167	nd	nd	nd	nd	nd	nd
Paxillus involutus	nd	18916	nd	nd	30	2	nd	nd
Agaricus sp.	nd	11534	nd	nd	nd	nd	nd	nd

* No data

Table 48. Multiyear dynamics of the radionuclide composition of *Paxillus involutus* (the exclusion zone, ^{137}Cs deposition is 185–259 kBq/m^2)

Year	Radionuclide composition, %							
	^{141}Ce	^{144}Ce	^{134}Cs	^{137}Cs	^{103}Ru	^{106}Ru	^{95}Zr	^{95}Nb
1987	2.3	11.09	17.6	36.9	2.3	2	8.2	18.8
1988	bd*	4.3	17.5	78.2	bd	bd	bd	bd
1989	bd	bd	14.6	85.4	bd	bd	bd	bd
1990	bd	bd	11.1	88.9	bd	bd	bd	bd
1991	bd	bd	7.6	92.4	bd	bd	bd	bd
1992	bd	bd	6	94	bd	bd	bd	bd
1993	bd	bd	4	96	bd	bd	bd	bd
1994	bd	bd	3.3	96.7	bd	bd	bd	bd

* Below detectable

The long-term dynamics of the radionuclide content in the fungi is complex and depends on (i) location and distance from the accidental unit (i.e. physicochemical properties of the initial fallout), (ii) climatic and soil-ecological conditions (soil type, forest litter pattern), and (iii) species peculiarities, particularly vertical distribution of mycelium in the soil [357]. In general, the long-term dynamics of ^{137}Cs uptake by fungi is presented in Table 47. In the first 2 years after the accident, the radionuclide content in fungi was not very high and determined primarily by surface contamination, which is confirmed by the similar radionuclide content of the fallout and fungi (Table 48) [96]. Fungi exhibit the maximum $TF^{137}Cs$ two-three years after the accident, when a considerable portion of the radionuclides is found in the mycelium-abundant A0f layer of the forest litter. The ensuing years are characterised by a gradual descending trend of ^{137}Cs content in fungi. The $TF^{137}Cs$, however, is very variable by year: the radionuclide content in the fungi was comparatively low during the extremely dry years (1992) and very high in the wet years. The correlation (r) between $TF^{137}Cs$ to fungi and rainfall reaches 0.82.

In some cases, the period of TF increase lasts up to 5–6 years. This may be explained by (i) the species features (for example *Lactarius necator* has deep mycelium, which undoubtedly affects the uptake dynamics [260]) and (ii) soil-ecological factors (for example, deep peat-like forest litter in the Ukrainian Polessie and Bryansk region of the Russian Federation).

The dynamics of TF variation over the entire investigated territory has its own peculiarities. The physicochemical properties of the initial fallout had the maximum effect on the ^{137}Cs accumulation in the first years after the accident. In this period, the maximum TF values were observed in the key sites attributed to the remote zone where the fallout was represented primarily by the gas-aerosol component (Tula and Bryansk regions) [92, 167]. The role of the soil-ecological factor has increased permanently in the subsequent years. Its significance increased in line with the soil capacity for irreversible adsorption of ^{137}Cs, which may be presented in the range as follows: peat-podzolic (gleyic)

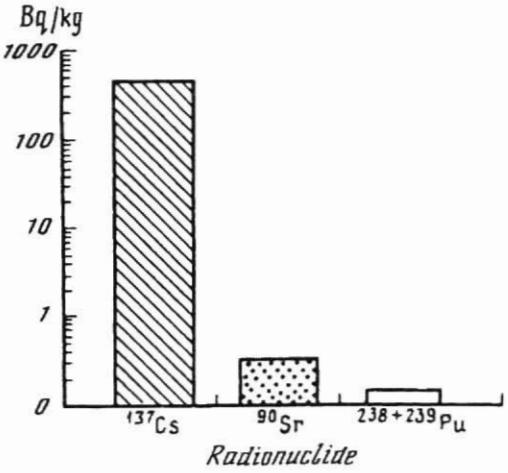

Fig. 40. ^{137}Cs, ^{90}Sr, and $^{238+239}Pu$ content in mushrooms (30 km exclusion zone, 1993)

soil, sand soddy-podzolic soil, loam soddy-podzolic soil, loam light-gray soil, podzolized chernozem. Therefore, the maximum decrease in $TF^{137}Cs$ to fungi (4 orders of magnitude) was observed on chernozems. The minimum decrease was observed in the near zone where the TF dynamics by 1995–1996 still depended on both irreversible adsorption of ^{137}Cs by soil and contunious release of this radionuclides from the fuel particles.

The role of fungi complex in the accumulation of other radionuclides, such as strontium and plutonium, is practically insignificant [328]. Fungi accumulate ^{90}Sr by 3 orders of magnitude less than ^{137}Cs, and the plutonium uptake is even less manifested (Fig. 40). Taking into account a high mycelium reserves in the forest environments (about 200 g/m^2) [335], it is obvious that mycobiota plays a very important role in biogeochemistry of ^{137}Cs.

3.4. RADIONUCLIDE TRANSFER TO FOREST PRODUCE

Estimation of the radionuclide transfer and accumulation in various forest produce makes it vital to re-establish and optimise commercial activities in the moderately contaminated areas. It is also of importance to estimate a potential dose burden (external and internal) on the population from the forest environments, although, the latter task is not in the scope of this study.

As discussed above, the radiocaesium content in the stem wood (including commercial wood) from the same area depends on species, as expressed in the following series: birch > aspen > oak > pine (Fig. 41). Thus, the most commercially valuable species (pine) is least contaminated, and the area of its safe use over the contaminated territory is wider compared to other species. On the other hand, the area appropriate for commercial use of birch (especially for fuel and wood charcoal) is considerably limited, since ^{137}Cs concentration in its ash and charcoal is higher than in its wood by an order of magnitude.

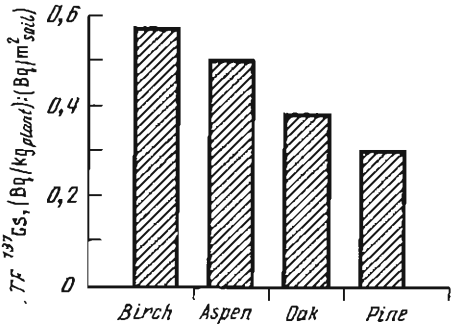

Fig. 41. Multiyear dynamics of $TF^{137}Cs$ from soil to barked wood of various tree species

Ecotope has also a considerable effect on the radionuclide content in commercial wood. The wood from the accumulative landscapes with hydromorphic soils is more contaminated than the wood from the eluvial landscapes with automorphic soils by a factor of 3–38 (see Chapter 3.1.2). The highest dependence of ^{137}Cs content in wood on the soil properties and moisture conditions is characteristic of birch followed by pine and oak.

The wood processing products differ by a radionuclide content: slab (made of marginal, young sap-wood) is more contaminated than cant (made mainly of core wood) by a factor of 1.5–6 (Fig. 42). This is due to a different radionuclide content across the stem (see Chapter 3.1.2, Fig. 15). The manifestation of the differences depends on both species and distance from ChNPP. The maximum differences between the products are characteristic of oak followed by pine, birch, and aspen. In general, specific activity of cant wood does not exceed the maximum permissible level over the entire contaminated territory (except for oak and birch in some areas of the near zone) [53, 196, 204–206]. Normalised concentration of ^{137}Cs in other wood products (timber, box board, edged board, etc.) is much similar to the cant wood.

The radionuclide content in other products of wood processing (resin, turpentine gum, and turpentine) is considerably lower than in the feedstock wood. In general, the products may be ranked by their radioactive contamination as follows: wood charcoal > slab > log > cant > turpentine gum > resin > turpentine. The most contaminated product is wood charcoal whose contamination is higher than wood by an order of magnitude. Contrary to that, contamination of resin and turpentine is very low and practically does not depend on deposition in the area of tree felling. As a rule, these products may be allowed for unlimited use independently on the area of their origin.

Expert's estimations suggest that the cost of non-wood produce from the forestlands exceeds the cost of wood by several times [244]. Most of this produce, however, is much more contaminated than wood and wood products. For example, hey forage in wet habitats is characterised by $TF^{137}Cs$ over 10^{-1} m^2/kg. It means that the hey obtained from the territory with deposition of 37–110 kBq/m^2 may be used as a fodder (under radiological control) only in combination with "clean" fodder amendments. Higher deposition makes the hey unusable at all [53, 196, 204–206]. The so-called conifer flour (a vitamin amendment to the cattle fodder made of pine or spruce needles and twigs) has relatively low concentration of ^{137}Cs, and may be procured in the territories contaminated up to 370 Bq/m^2 (Table 49). Some fun-

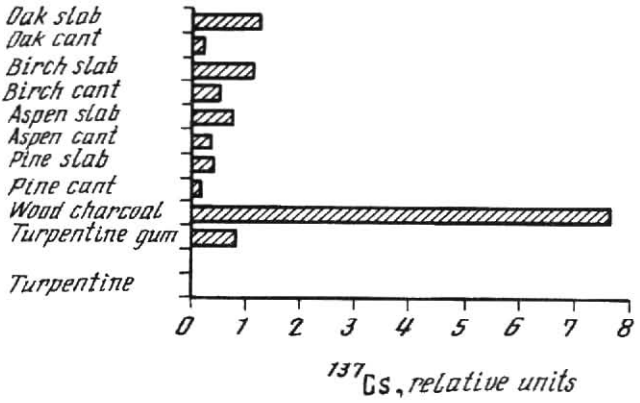

Fig. 42. ^{137}Cs content in various wood produce

gus species used for medical purposes such as *Amanita muscaria* and *Lycoperdon bovista* also do not accumulate a considerable amount of radionuclides [263] compared to other fungi. Medicine herbs were contaminated by ^{137}Cs only, independently on gathering area. The only raw stuff contained a significant portion of ^{144}Ce and ^{106}Ru was external bark (used for some medical purposes). TF^{137}Cs to medicine herbs (dry weight) varied by 30 times depending on species (Table 50). The specimens gathered in hydromorphic areas contained tens times more radionuclide than the specimens of same species collected in automorphic areas.

^{137}Cs content in berries is also ecotope-dependent. In hydromorphic areas, the radionuclide concentration in berries may exceed its concentration in the leaves and is up to three times more contaminated than in the automorphic areas. In wet years, TF^{137}Cs to forest berries tends to increase. Berries of *Rubus idaeus, Rubus saxatilis, Vaccinum myrtillus,* and *Fragaria vesca* are characterised by a positive correlation between the radionuclide content in the stem, fruit, pericarp, and seeds [261]. If compare ^{137}Cs in berries with the maximum permissible levels in food, it is appeared that berries may be picked in the areas with the deposition of less than 140 kBq/m^2. Acorns have about the same ^{137}Cs content as grass vegetation. Since this forest produce is used by local population as a fodder amendment only, acorn may be picked in the areas with the deposition of less than 370 Bq/m^2 [53, 196, 204–206]. Wild apples exhibit the least accumulation capacity of all forest produce.

^{137}Cs concentration in birch sap depended on all above-mentioned factors affecting radionuclide accumulation in tree plants (see Table 51). In the areas with the deposition of 184–32, 560 kBq/m^2 by ^{137}Cs and 74–19, 240 kBq/m^2 by ^{90}Sr, the radionuclide concentration in the birch sap varied from 19 to 326 Bq/l and 36 to 700 Bq/l, respectively (see Table 49). ^{137}Cs and ^{90}Sr were the only radionuclides found in the birch sap. ^{90}Sr content was

Table 49. Statistical indices of ^{137}Cs и ^{90}Sr content in components of fodder pine twigs (the exclusion zone, 1992, average of n = 42)

Parameter		Statistical indeces					
		m	±m	max	min	G	V, %
Needles and twigs of current year							
Phytomass*	g	344.9	20.7	605.9	114	134.3	38.9
	% of total**	35.9	0.62	44.9	27.3	4	11
Moisture content***	%	63.4	0.31	68.9	58.8	2	3.2
^{137}Cs	nCi/kg	1746.7	156.3	3900	290	1012.9	58
	%	66.3	0.76	84.8	53.7	4.9	7.4
^{90}Sr	nCi/kg	328.1	78.6	2100	18	491.2	149.7
	%	28.4	2.5	79.6	4	16.5	57.9
"Old" needles							
Phytomass*	g	373.8	19	589.5	143.4	123.4	33
	% of total**	39.6	0.52	44.4	32.6	3.4	8.6
Moisture content***	%	56.1	3.9	62.1	46.2	2.5	4.4
^{137}Cs	nCi/kg	478.7	47.9	1300	71	310.6	64.9
	%	19.9	0.62	29.9	9.1	4	19.9
^{90}Sr	nCi/kg	409.3	91.9	2391	28	595.4	145.5
	%	31.7	2.1	62.2	5.1	13.7	43.1
"Old" twigs (devoid of needles)							
Phytomass*	g	230.5	12.5	434.1	84.6	81	35.2
	% of total**	24.5	0.54	38.5	20.8	3.5	14.5
Moisture content***	%	59.1	0.42	67.7	54	2.7	4.5
^{137}Cs	nCi/kg	527	47.5	1200	85	307.8	58.4
	%	13.8	0.49	25.4	6.1	3.2	23.1
^{90}Sr	nCi/kg	425.1	85.8	2500	16	556.1	130.8
	%	39.9	2.4	90.2	0.8	15.5	38.9
Fodder twigs in total							
Phytomass*	g	949.2	50.2	1582.8	352.2	325.4	34
	% of total**	100					
Moisture content***	%	59.7	0.32	64.9	54.6	2.1	3.5
^{137}Cs	nCi/kg	942.2	84.4	2168.3	157	547	58
	%	100					
^{90}Sr	nCi/kg	106.2	30.6	985.5	4.8	198.4	186.8
	%	100					

* dry weight, t=105°C
** % of total weight of the fodder twigs
*** weight %

Footnote: m – mean; G – standard deviation; (m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

Table 50. $TF^{137}Cs^{*}$ from the soil to some forest produce in the exclusion zone (means for 1987–1990)

Produce	Type of landscape/moisture regime	
	Eluvial/automorphic	Accumulative/ hydromorphic
Officinal herbs and berries		
Peucedanum orioselinum	2.7	nd**
Cereals (mixed sample)	4.9	83
Plantago lanceolata	1.7	nd
Convallaria majalis	4.9	nd
Majanthemum bifolium	12.7	194.8
Betonica officinale	13	nd
Hipercum perforatum	0.5	nd
Polygonatum officinale	3.9	nd
Pteris aquilina	4.8	nd
Melampyrum nemorosum	4.8	94.3
Rubus idaeus		
(leaves)	6.4	33.9
(berries)	15.9	49.7
Fragaria vesca		
(leaves)	6.6	nd
(berries)	10.2	nd
Vaccinium myrtillus		
(leaves)	nd	50
(berries)	nd	56.6
Rubus idaeus		
(leaves)	nd	51
(berries)	nd	66.3
Other		
Acorn	1	nd
Forest apple (fruits)	0.6	nd

* $TF^{137}Cs = (Bq/kg_{plant}) : (kBq/m^2g_{soil}) = n \cdot 10^{-3}\ m^2/kg$, dry weight

** No data

twice as much as ^{137}Cs, in spite of the fact that ^{137}Cs deposition was higher than ^{90}Sr. Thus, the root uptake of ^{90}Sr by birch is at least twice as high as ^{137}Cs, and ^{90}Sr shall be of particular attention when performing radioecological monitoring of forest produce. $TF^{137}Cs$ to birch sap varies in a wide range: from $1 \cdot 10^{-3}\ m^2/l$ (in the near zone) to $5 \cdot 10^{-4}\ m^2/l$ (in the marginal part of the exclusion zone and the remote zone). $TF^{90}Sr$ is about 2–6 times higher than $TF^{137}Cs$ and varies within a range of $3.6 \cdot 10^{-4}$-$n \cdot 10^{-3}$.

Table 51. TF^{137}Cs and ^{90}Sr to birch sap in the 30 km exclusion zone (Bq/l_{sap} : kBq/m^2_{soil},1989)

Forest type; distance and direction from ChNPP; key site	Deposition, kBq/m^2*	^{134}Cs	^{137}Cs	^{90}Sr	^{137}Cs/^{90}Sr	^{137}Cs	^{90}Sr
		Bq/l				TF, $n \cdot 10^{-4}$ m^2/l	
Marginal part of the exclusion zone							
Mixed, broad-leaved-pine forest, 28 km to S; D-1	159/56	3.3	18.5	35.9	1/2	1.2	6.4
Alder forest, 26 km to S; D-3	148/78	14.4	740	77.7	1/1	5	10
Near zone							
Pine forest (forest margin) 7 km to S; K-2	2405/1036	27.8	133.2	247.9	1/2	0.6	2.4
Pine forest 7 km to S; K-2	1739/740	7.4	40.7	162.8	1/4	0.2	2.2
Single tree in a village, 7 km to SW	2035/888	5.6	18.5	70.3	1/4	0.1	0.78
Mixed, broad-leaved-pine forest, 6 km to W; Sh-1	32560/19129	66.6	325.6	703	1/2	0.1	0.36
Mixed, broad-leaved-pine forest, 10 km to NW	148/81	8.1	36.6	66.7	1/2	2.5	8.2

* Average deposition within the root-available area of a sampling tree: numerator: ^{137}Cs; denominator: ^{90}Sr

It suggests that the spatial variation of ^{90}Sr availability is somewhat less than that of ^{137}Cs. TF^{137}Cs and TF^{90}Sr to the birch sap in the hydromorphic areas are higher than in the automorphic area by an average factor of 4 and 1.5, respectively. It means that the migration ability (and availability) of ^{90}Sr is less dependable on soil properties and regimes compared to ^{137}Cs. There is a weak negative correlation between the intensity of sap production and radionuclide concentration in the sap (Fig. 43). By now (1997), both ^{137}Cs and ^{90}Sr content in the birch sap over the entire contaminated territory is less than the current maximum permissible levels for drinking water [135].

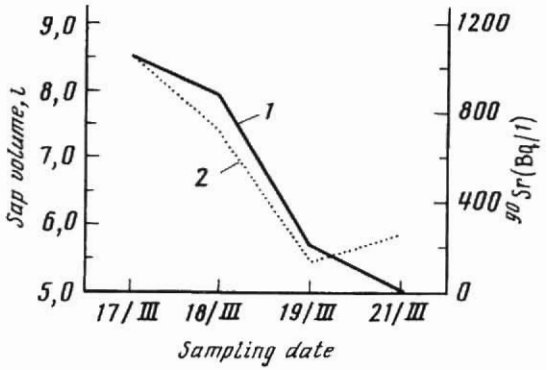

Fig. 43. The dynamics of $TF^{90}Sr$ from the soil to birch sap in the exclusion zone (1989)

When the data on the "Chernobyl" ^{137}Cs in the forest produce are compared with the corresponding data on the global fallout ^{137}Cs , it appears that the Chernobyl $TF^{137}Cs$ exceeds the latter about 3 times. Since the time lag between the fallout events is 22 years, it is possible to estimate the effective half-stay of ^{137}Cs in forest produce. A conservative estimation suggests that the contamination of forest produce will decrease at least twofold during 15 years after the accident.

4. RADIONUCLIDE BEHAVIOUR IN FOREST SOILS

The radionuclide behaviour in the soil determines, to a great extent, the radionuclide root uptake and its further migration by food chains. The radionuclide fate in the soil is determined by a wide spectrum of simultaneously running, often competitive, elementary processes, such as adsorption-desorption, diffusion-mass transport, retention-migration, etc. The intensity of each elementary process depends, in turn, on a combination of several factors such as nature of the radionuclide, physicochemical features of the fallout, soil properties, environmental regimes, etc.

Abundant experimental and field data accumulated over a number of years suggest that the radionuclide interaction with soil is a complex problem, since individual processes may contradictory influence the radionuclide behaviour depending on a wide range of factors and parameters. [113, 133, 154, 155, 231, 273, 345]. In general, we may suggest two basic stages describing the radionuclide behaviour in the soils. At the first stage, the distribution of the radioactive fallout depends primarily on weather conditions, topography, and vegetative cover in the exposed area. At the second stage, radionuclide redistribution depends primarily on a set of factors characteristic of individual climatic zones, soils, and ecosystems (depending on the scope and scale of the study) [152]. Independently on soil and landscape, in all cases, most of radionuclides is retained in the upper soil layer for a long time. However, further distribution of the radionuclide content and mobility (availability) down the soil profile is determined by numerous soil factors, such as pH, clay and organic matter content, mineralogical composition, presence (absence) of local confining beds, etc. [30, 88, 97, 181, 183, 202, 260, 277–280, 324]. A very specific radiological feature of the Chernobyl accident is a combination of the global scale of contamination and high spatial variability of the fallout properties and composition over the contaminated territory. This sets a challenge to develop a long-term forecast of a radioecological situation in various forest ecosystems in the territories of various types of contamination.

4.1. RADIONUCLIDE COMPOSITION OF THE CONTAMINATED SOILS

The investigated territory as a whole is characterised by the extremely various radionuclide composition and fallout properties.

In the Russian Federation, the radionuclide composition is rather uniform. In 1988, ^{137}Cs and ^{134}Cs were the main dose-forming radionuclides, and the proportion of ^{90}Sr did not exceed 1–2% (7.4–29.6 kBq/m^2). Trace amount of

Table 52. The radionuclide composition dynamics in the investigated key sites in 1988 and 1992

Key site	Radionuclide composition, %												
	1988							1992					
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{90}Sr	^{95}Zr	^{95}Nb	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{90}Sr	^{95}Zr
Tula region, Russian Federation													
Pl-1	bd*	18.4	75.2	6.4	bd	bd	bd	bd	6.4	93.6	bd	bd	bd
Pl-2	bd	17.9	76.7	5.4	bd	bd	bd	bd	6.4	93.6	bd	bd	bd
Bryansk region, Russian Federation													
Kl-1	0.04	18.3	75.9	5.8	bd	bd	bd	bd	6.4	93.6	bd	bd	bd
Kl-2	0.05	18.4	75.3	6.2	bd	bd	bd	bd	6.5	93.5	bd	bd	bd
Zl-1	1.5	18.1	74.2	6.2	bd	bd	bd	bd	6.5	93.5	bd	bd	bd
Exclusion zone, Ukraine													
D-1	47.5	5.3	15.6	19.4	7.5	2.6	2.1	0.6	2.7	65.4	0.7	30.6	bd
K-2	48.9	4.7	14.6	20.4	7.9	1.1	2.4	0.6	2.5	66.3	0.9	29.7	bd
Sh-1	52.8	4.6	13.4	17.7	7.6	1.3	2.6	0.7	2.4	59.8	1.1	36.0	bd

* Below detectable

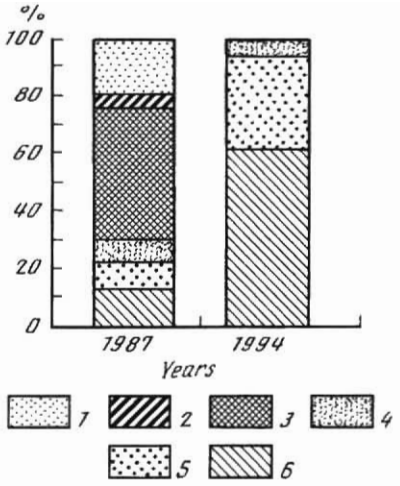

Fig. 44. Average changes in the radionuclide composition in forest soils (30 km exclusion zone):
1 – ^{106}Ru; 2 – ^{95}Zr + ^{95}Nb; 3 – ^{144}Ce; 4 - ^{134}Cs; 5 – ^{90}Sr; 6 – ^{137}Cs

^{106}Ru and ^{144}Ce was registered in Bryansk region, during the first years after the accident only [280]. Later, ^{134}Cs proportion has decreased triple for 5 years, while that of ^{137} Cs has increased.

In the exclusion zone of ChNPP (1987), the radionuclide composition was represented by ^{144}Ce (47–52%), ^{106}Ru (17–20%), ^{137}Cs (15%), ^{90}Sr (8%), ^{134}Cs (5%), and a negligible proportion of ^{95}Nb and ^{95}Zr (Table 52). In spite of the distance and position of the key sites relative to the accidental unit, the fallout properties and the radionuclide composition did not vary much over the exclusion zone. Currently, the radionuclide composition in the territory is, in fact, constituted by ^{137}Cs, ^{134}Cs, and ^{90}Sr only (a proportion of plutonium isotopes is negligible) (Fig. 44). The average ^{90}Sr/^{137}Cs ratio over the exclusion zone is about 0.6 and tends to decrease as the distance from the accidental unit decreases. The relative proportion of ^{241}Am in the soil tends to increase, though it currently constitutes less than 0.5% of the total deposition [131]. Therefore, the observed increase in the ^{241}Am fluxes in the chain "soil-plants-animals-human being" is practically insignificant. The same proportion (about 0.5%) is characteristic now of the short-lived ^{144}Ce and ^{106}Ru. In 1995, these radionuclides were registered only in the upper two centimetres of the soil profile. The radionuclide composition of deeper soil layers is represented by ^{137}Cs and ^{90}Sr only.

4.2. SPATIAL DISTRIBUTION OF THE RADIONUCLIDES IN THE SOILS

Specific features of the Chernobyl accident (a violent explosion, followed by a severe fire) caused a high spatial variability of the initial distribution of radionuclides over the contaminated territory. The main feature of the large-scale contamination pattern is believed to be a sharp difference between the so-called "plum-affected zone" (up to 60 km around the accidental unit) and the "discharge zone" (over 60–250 km from ChNPP) [96, 203]. The radionu-

Table 53. Spatial variability of ^{137}Cs in the investigated soils (Ci/km^2, 1992)

Key site	Statistical indices						
	n	m	σ	± m	V,%	max	min
Tula region, Russian Federation							
Pl-2	16	9.1	2.12	0.53	23.3	13.7	6.1
Kaluga region, Russian Federation							
Kh-2	24	9.6	0.6	2.17	22.6	13.3	6.8
Bryansk region, Russian Federation							
Nz-1	26	39.0	11.3	2.75	28.2	63.0	15.4
30 km exclusion zone, Ukraine							
Sh-5	289	103.4	32.4	1.91	32.4	270.0	11.2

Footnote: m – mean; G – standard deviation; ±m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

clide pattern within the plum-affected zone is in general agreement with the lognormal distribution. At a greater distance from ChNPP, the occurrence of an extreme radionuclide content in the limited (micro) areas is shifted to the right relative to the Gaussian distribution, but is not lognormal [7, 92, 289]. In other words, the distribution of radionuclides approximates the Gaussian distribution (characteristic of the global fallout) as the distance from ChNPP increases [216].

Spatial variability of the radionuclide content in the forest soils of the Russian Federation is within the range of 22 to 30% (Table 53). There is some tendency toward increase in the spatial variability on drawing on to the accidental unit. A variation range of the radionuclide content (min-max) within the boundaries of 30 km exclusion zone is by an order of magnitude higher than that in the remote zone.

Spatial variability of radionuclides in the soils depends on the ecosystem type as well. Maximum and minimum of this index is, as a rule, attributed to young pine forests and mature mixed and pine forests, respectively. This is due to (i) direct effect of the tree crones of different age on the initial distribution of the fallout, and (ii) specific microtopography of the soil surface in mature forests that causes a further spatial redistribution of radionuclides within specific micro-areas [67].

Spatial variability of the deposition depends on the chemical nature of the radionuclide, the depth of a soil layer, the fallout properties, and some other factors.

For gamma emitting radionuclides, the least variation is characteristic of ^{137}Cs, whereas in case of ^{144}Ce and ^{106}Ru it is much higher. This is due to some

Table 54. Statistical indices of the radionuclide content in the automorphic areas (Ci/km^2, the exclusion zone, 1991)

Layer	m	± m	σ	V, %	max	min
			^{137}Cs			
Forest litter (A0)	103.4	1.91	32.43	31.0	270.0	11.0
0-5	18.23	0.82	13.9	76.2	130.0	1.7
5-10	1.68	0.11	1.84	109.3	19.0	0.13
0-10	19.68	0.83	14.09	72.0	132.7	1.98
0+(0-10)	122.9	1.84	31.31	25.0	282.5	23.1
			^{134}Cs			
Forest litter (A0)	10.28	0.20	34.03	33.0	28.0	1.0
0-5	1.83	0.08	1.41	76.9	13.0	0.15
5-10	0.17	0.011	0.195	114.9	2.1	0.0
0-10	1.97	0.08	1.40	71.0	13.22	0.17
A0+(0-10)	12.52	0.19	3.29	27.0	29.27	23.0
			^{144}Ce			
Forest litter (A0)	17.53	0.27	4.66	27.0	30.0	1.9
0-5	1.38	0.12	2.11	152.1	22.0	0.0
5-10	0.015	0.004	0.064	428.4	13.0	0.0
0-10	1.12	0.12	2.06	184.0	22.0	0.0
A0+(0-10)	18.94	0.25	4.19	22.0	32.2	3.2
			^{106}Ru			
Forest litter (A0)	12.25	0.34	5.81	47.0	77.0	0.0
0-5	0.84	0.10	1.72	206.7	13.0	0.0
5-10	0.0045	0.0023	0.04	867.7	0.54	0.0
0-10	0.84	0.10	1.72	205.0	13.0	0.0
A0+(0-10)	13.09	0.33	5.63	43.0	77.0	0.0

Table 54. (continuation)

Layer	m	± m	σ	V, %	max	min
			Total Σγ			
Forest litter (A0)	143.4	2.51	42.2	29.0	326.0	15.6
0-5	22.28	1.08	18.32	82.2	178.9	1.85
5-10	1.87	0.12	2.03	108.7	21.0	0.13
0-10	23.89	1.09	18.54	78.0	181.82	2.15
A0+(0-10)	167.3	2.35	39.95	24.0	341.0	30.3

Footnote: m – mean; G – standard deviation; ±m – mean error; V, % – variation coefficient; max, min – maximal and minimal values, respectively

specific features of the initial fallout (Table 54). ^{144}Ce and ^{106}Ru are known to be attributed chiefly to the so-called "fuel", large-size component of the fallout, and most ^{137}Cs is concentrated in the "gas-condensed", highly dispersed component [72, 91, 140, 142]. The latter is an apparent reason for lower spatial variability of ^{137}Cs.

Spatial variability of the radionuclide content over the territory depends also on the depth, which is differently manifested for automorphic and hydromorphic areas. In the automorphic areas, the coefficient of variability (V) for both the total deposition and individual radionuclides increases drastically with depth (Table 52, Fig. 45). This is caused primarily by individual migration activity of different radionuclides [236, 242], variation of the forest litter parameters, and different density (i.e., retention capacity) of the moss cover. Considerable increase in V with depth in the automorphic soils indicates that the initial variability of the radionuclide content increases with depth, as radionuclides leach down the soil profile. The resulting tongue-like character of the lower boundary of the radionuclide vertical distribution in the soil is apparently the main cause of a higher spatial variability of the radionuclide content in plants than in the soil. The total deposition is the least variable and seems to reflect the spatial variability of the initial fallout over the territory [236, 242, 277]. Interestingly, the variation of the radionuclide content in the investigated soils is much similar to that of heavy metals, trace elements, and some macro-elements (Ca and K) [50, 102, 163]. The only significant difference is that the variation of radionuclides increases with depth more actively compared to stable elements. These differences between radionuclides and stable elements will likely smooth with a passage of time.

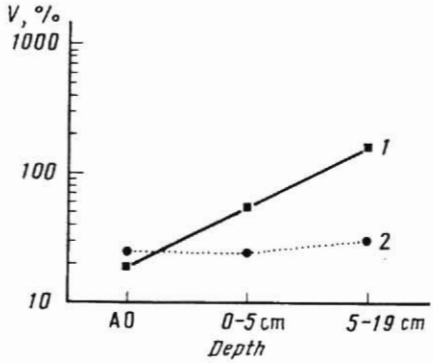

Fig. 45. The coefficient of variation of ^{137}Cs content in the soil profile:
(1) automorphic and (2) hydromorphic areas (1994)

In the hydromorphic soils, the variation coefficient of the radionuclide content does not depend on depth as clearly as in the automorphic soils (Fig. 44), which makes it possible to say about the "frontal" migration of the radionuclides down the soil profile. This phenomenon may be explained by several reasons, such as (i) higher moisture that promotes both the convective and diffusive transfer in the soil [18, 139, 254], and (ii) higher content of soluble organic compounds which are known to promote radionuclide migration by forming mobile radionuclide-organic compounds [5].

Spatial variability of the deposition decreased almost twofold by the fifth year after the accident (Table 55). This is likely due to destruction of the fallout particles and a more uniform redistribution of mobile radionuclides in the soil with a passage of time.

A better understanding of the radionuclide distribution processes in the "near zone" and in the remote zone may be obtained from the corresponding small-scale schemes (Fig. 46). The distribution of ^{137}Cs in the exclusion zone has a mosaic-like, concentric-confined character and does not correspond to the local topography. Such a distribution is characteristic of the initial fallout in the vicinity of ChNPP [94, 96]. This is confirmed by the same character of ^{144}Ce distribution that is a marker of the "fuel" component (Fig. 47). Thus, the observed spatial distribution of both ^{137}Cs and ^{144}Ce (and the deposition as a whole) in the "near zone" is still determined by spatial variability of the initial fallout, i.e., no significant redistribution of the radionuclides by the topographical elements has occurred for the recent 8–10 years.

Table 55. The dynamics of the coefficient of variation (V, %) of the deposition in the key sites

Year	Key sites			
	D-1	D-3	K-2	Sh-1
1986	36	39	bd	bd
1987	41	38	31	43
1991	bd*	bd	bd	24
1994	17	17	bd	bd

* Below detectable

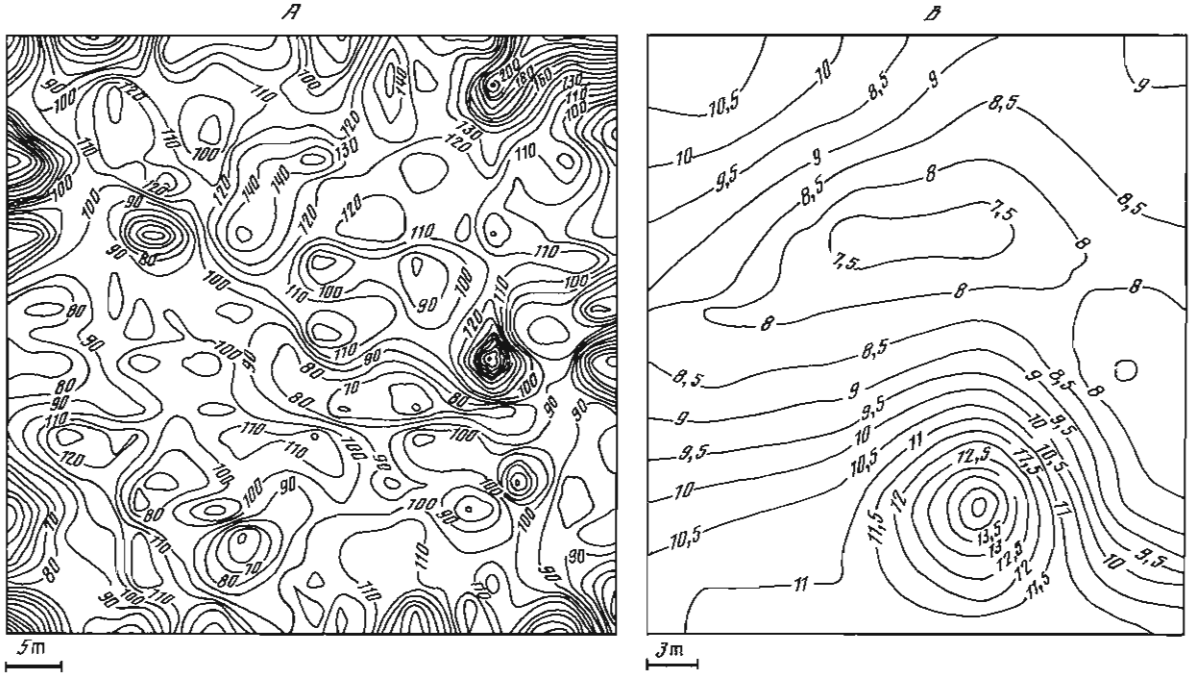

Fig. 46. Spatial distribution of ^{137}Cs in the forest litters (Ci/km^2):
(A) 5 km from ChNPP; (B) 500 km from ChNPP

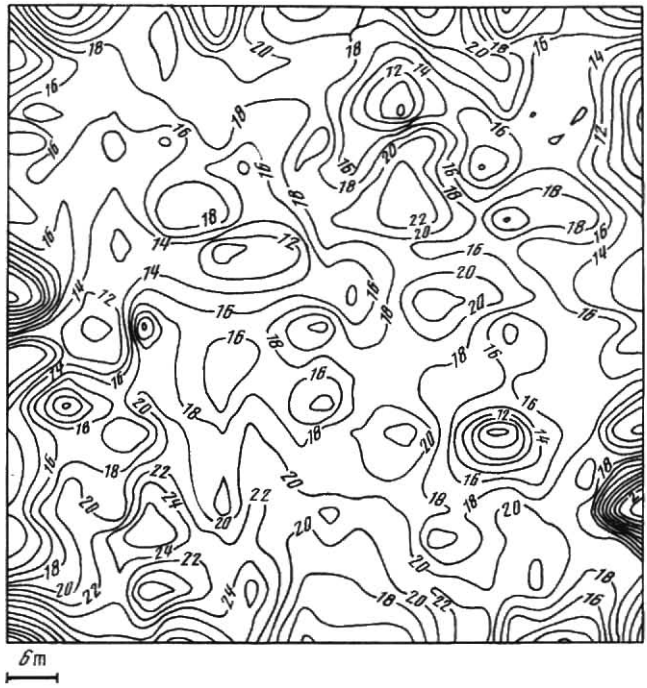

Fig. 47. Spatial distribution of ^{144}Ce in the forest litter (Ci/km^2, 5 km from ChNPP)

In the remote zone, spatial distribution of ^{137}Cs deposition is much different from that in the exclusion zone (Fig. 46, 48). The corresponding isolines are well shaped and follow local topographical contours. There are only few localities displaying the mosaic-like "hot spots", in which the radionuclide pattern resembles that in the exclusion zone. It is of interest that such hot spots are of a more frequent occurrence in the automorphic areas compared to the hydromorphic areas and environments. This is likely due to higher radionuclide mobility and faster smoothing of the initial heterogeneity in the fallout distribution. The absence of a pronounced mosaic-like pattern and well shaped isolines suggest that the processes of radionuclide migration are more manifested and more influential on the distribution of radionuclides in the remote territories compared to the exclusion zone.

In addition, the schemes demonstrate some specific features of the radionuclide migration down the soil profile (Fig. 49). The mosaic-like distribution of ^{144}Ce in the soil layer of 5–10 cm is the same as in the forest litter (0–5 cm). This indicates that the vertical migration of this radionuclide is attributed to some microlocalities, such as soil fractures, worm-holes, etc., where the fallout particles may be transported down without destruction (for example, by soil fauna, *lessivage, etc.*) [118, 40. 55]. This is also confirmed by the data of soil autoradiography: although, the absolute content of the "hot" particles decreases down the soil profile, their proportion in each soil layer remains stable [72].

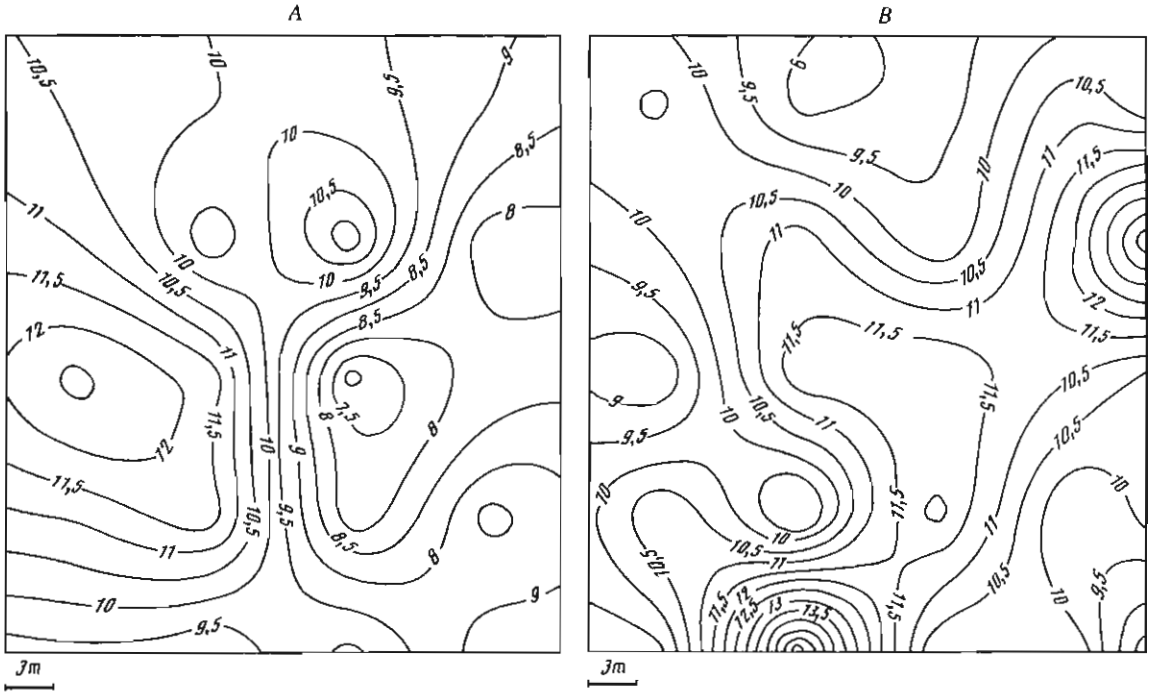

Fig. 48. Spatial distribution of ^{137}Cs in the soils (Ci/km^2, 500 km from ChNPP):
(A) eluvial landscape, automorphic area, mixed forest; (B) accumulative landscape, hydromorphic area, sedge bog

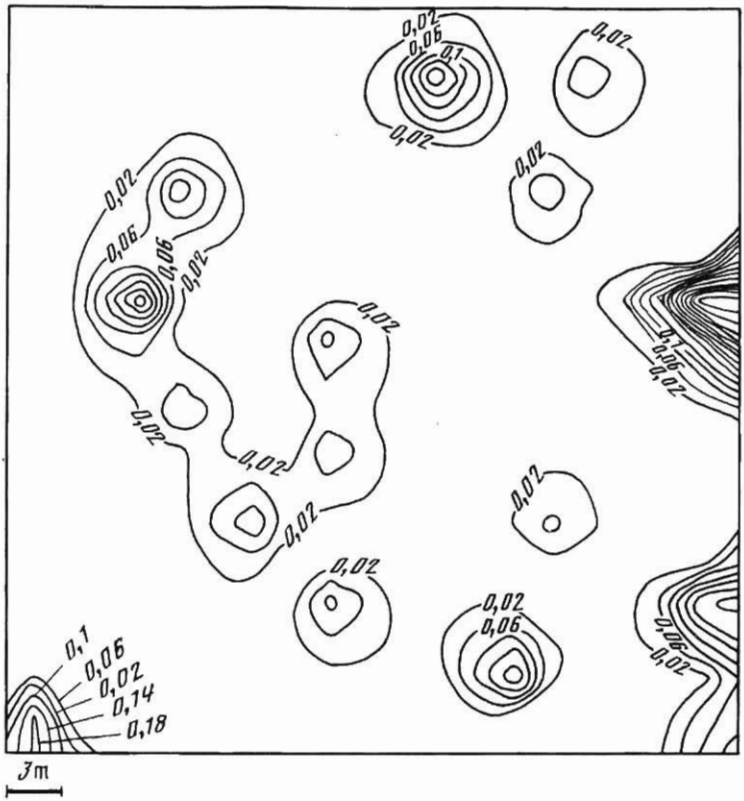

Fig. 49. Spatial distribution of ^{144}Ce in the upper 10 cm mineral soil layer (Ci/km^2, 5 km from ChNPP)

Thus, the manifestation of mosaic-like distribution of the Chernobyl-derived radionuclides decreases as the distance from the accidental unit increases. In the automorphic soils, spatial variability of the deposition increases with depth, and the active radionuclide migration is attributed to specific microzones (soil fractures, worm-holes, etc.). In the hydromorphic areas, spatial variability of the deposition does not depend much on depth, which suggests a "frontal" radionuclide migration. Lateral redistribution of radionuclides at the microtopographical scale is more intensive in the hydromorphic rather than automorphic areas, which is confirmed by a more pronounced mosaic-like character of the radionuclide distribution in the latter. In the hydromorphic areas, the spatial radionuclide pattern loses its initial structure 5–8 years after the accident, and becomes topography-dependent. In the automorphic areas, the initial pattern has still remained 10 years after the accident.

4.3. DEPOSITION DYNAMICS IN THE SOIL

Radionuclide deposition in the soils is known to be a basic criterion of the radioecological situation in a contaminated territory [96]. The long-term monitoring suggests that the changes to the total deposition in the territory (σ) since 1986 has primarily occurred as a result of radioactive decay. Vertical and later radionuclide transport beyond the ecosystem boundaries did not exceed the parts of a per cent ($\sigma \cdot n \cdot 0.1\%$) per year [236], which is incomparable with the rate of radioactive decay. Annual incorporation of the radionuclides (even the most "biologically significant" caesium and strontium) to the biological cycle in most cases also does not exceed this value [276–280]. Variation in the radionuclide composition over the contaminated territory causes the corresponding differences in the long-term deposition dynamics depending on the proportion of short-lived radionuclides (Table 56). The most manifested decrease in σ is characteristic of the immediate surroundings of the accidental unit (the "near zone") where over 50% and 17% of the radionuclide composition was represented by short-lived radionuclides (including ^{144}Ce and ^{106}Ru) and long-lived ^{137}Cs, respectively (data of 1986–1987). The remote zone (Bryansk and Tula regions), where over 70% of deposition was represented by ^{137}Cs, and the proportion of ^{144}Ce and ^{106}Ru was negligible, is characterised by a considerably slower decrease in the deposition.

Table 56. The dynamics of the total deposition of gamma-irradiating radionuclides in the investigated soils (MBq/m^2)

Key site	Year								
	1986	1987	1988	1989	1990	1991	1992	1994	1995
30 km exclusion zone, Ukraine									
D-1	7.43	1.16	0.61	0.42	0.28	0.22	0.18	0.16	0.15
D-3	7.19	1.16	0.62	0.46	0.28	0.23	0.2	0.17	0.16
K-2	87.87	14.56	7.93	5.73	3.73	2.97	2.52	2.22	2.12
Sh-1	1355	260.8	123.7	82.2	51.3	39.7	33.2	28.5	23.9
Tula region, Russian Federation									
Pl-1	bd*	bd	0.46	0.42	0.38	0.36	0.34	bd	bd
Pl-2	bd	bd	0.48	0.44	0.4	0.38	0.36	bd	bd
Bryansk region, Russian Federation									
Kl-1	2.58	bd	0.81	0.72	0.67	0.63	0.6	bd	bd
Kl-2	2.31	bd	0.73	0.66	0.6	0.56	0.54	bd	bd
Zl-1	5.65	bd	1.82	1.65	1.43	1.39	1.32	bd	bd

* Below detectable

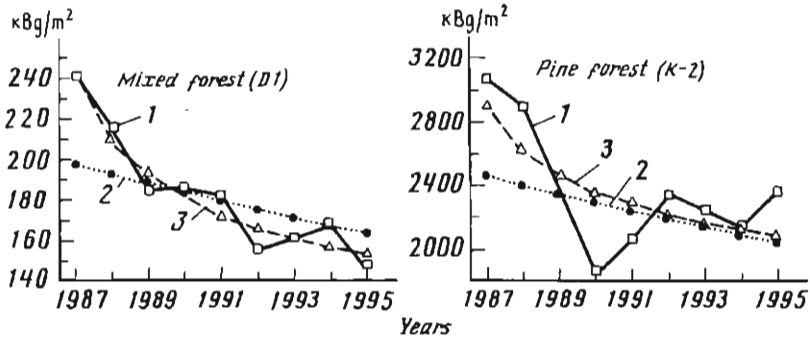

Fig. 50. The observed and calculated dynamics of ^{137}Cs in forest soils:
(1) field data; (2) data calculated by the "share" method; (3) data calculated by the "regression" method

With this general features, the long-term dynamics of the deposition exhibits a sharp variation by years. This is likely due to an insufficient sample to reflect the true average deposition under conditions of its high spatial heterogeneity. This circumstance impeded analysis of the field data. Therefore, the necessary data pool was obtained using the decay-corrected data on the deposition at each sampling site for several years. The total sample was averaged, and the deposition dynamics was recalculated versus the average. This method made it possible to smooth the effect of spatial heterogeneity and obtain statistically reliable data. The deposition dynamics calculated using this method is in a good agreement with the time series obtained in the field (Fig. 50).

The long-term (over 10 years) monitoring made is possible to approximate the observed deposition dynamics using the regression analysis. The calculations suggest that the deposition dynamics (σ) is well approximated by the following equation:

$$Y = e^{a0} \cdot x^{a1},$$

where a_0 and a_1 are the coefficients shown in Table 57.

Table 57. The coefficients of regression for the deposition dynamics in various key sites

Coefficient	D-1	D-3	K-2	Sh-1
a0	1.88	1.858	4.361	7.098
a1	–0.2118	–0.1798	–0.1525	–0.2426

4.4 VERTICAL DISTRIBUTION OF RADIONUCLIDES IN THE SOIL

Radionuclide redistribution down the soil profile determines, to a large extent, a radiological situation in forest ecosystems, and is one of the most influential factors of the radionuclide availability and accumulation. Vertical radionuclide migration in forest ecosystems depends on several transport processes [97, 183, 260, 277–280). A specific structure of forest soils makes it reasonable to analyse the processes of radionuclide migration separately in forest litter (A0) and mineral horizons.

4.4.1. Radionuclide Content and Dynamics in Forest Litter

Forest litter is known to be a very important factor of influencing the behaviour of all chemical elements in forest ecosystems [8, 90, 102, 103, 238, 239, 242).

A most informative index of the vertical migration of radionuclides is their distribution among mineral soil layers and forest litter. In 1992, forest litter in the contaminated territory contained from 27 to 91% of ^{137}Cs deposition in the soil (Table 58). The radionuclide proportion in mineral layers increases as the distance from ChNPP increases. However, the range of variation of this index is rather uniform in all investigated areas independently on the distance. This suggests that the radionuclide migration be affected by soil factors (such as the thickness and structure of forest litter) rather than the initial fallout properties (Fig. 51).

The effect of forest litter on the rate of radionuclide redistribution between the soil and forest litter depends on the thickness of the latter. The closest correlation is observed for the forest litter thickness of 3.5–4.0 cm. It is less mani-

Table 58. The range of variation of the ^{137}Cs content in forest litters (data of 1992)

Area	Range of variation of ^{137}Cs content (%)	
	By mean values	By individual measurements
	Russian Federation	
Tula region	0.1–0.5	0.1–1.0
Kaluga region	26.5–76.0	8.9–91.8
Bryansk region	33.3–64.8	7.1–87.4
	Ukraine	
30 km zone of ChNPP	58.3–90.5	9.5–95.5

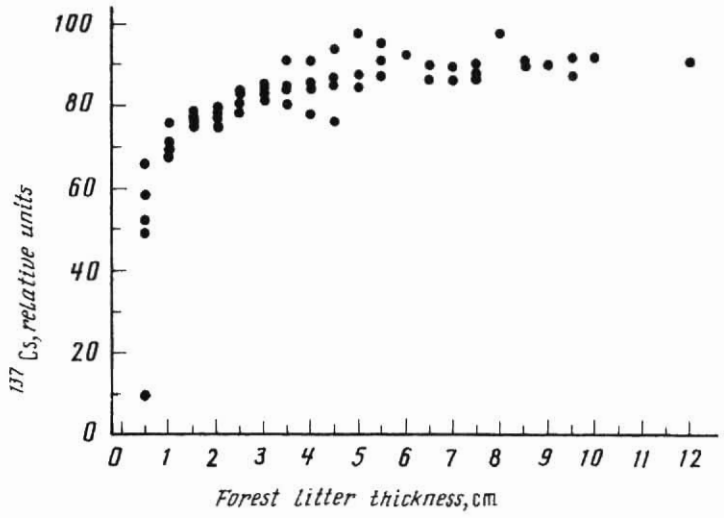

Fig. 51. Retention capacity of the forest litters of various thickness for ^{137}Cs

fested when the thickness is 3.5–5.5 cm, and almost negligible when the thickness is 6.0–12.0 cm.

In the interval of 6–12 cm, retention capacity of a forest litter is practically independent on its thickness. This picture is characteristic usually of dead-covered soils under spruce forests and moss-covered soils under pine forests with "mor" (or "mor-moder") types of organic matter, or in the stem-adjacent area (Table 59). The maximum retention capacity of the soil litter in these forest types is determined by two factors:

1. A low transformed organic matter with a low content of a mineral component [197]. These features combined with a thick (>6 cm) organic layer lead to a capillary rapture, which slows down water and element exchange within the forest litter and promotes radionuclide retention in the layer.

2. Accumulative capacity of soil biota (particularly microorganisms) is most pronounced in coniferous cenoses. Fungus mycelium is known to be a very effective concentrator of radionuclides, therefore, mycobiota may contain 10 to 60% of the total ^{137}Cs inventory in the soil [304, 312, 335, 357].

3. A manifested moss cover over the forest litter promotes radionuclide retention in the upper organic layer (Table 60). This is due to the above-discussed capability of mosses (lichen) for effective accumulation of radionuclides.

In hydromorphic areas, the least radionuclide content in the forest litter is characteristic of back (eutrophic) bogs under alder forest. In sphagnum (oligotrophic) bogs the maximum radionuclide content is attributed to the upper sphagnum layer (see Table 57). In the latter case, the maximum of ^{137}Cs inventory is located in the sphagnum residues dated 1986–1988 (0–5cm layer) which is quite explainable. Below this upper layer, the radionuclide content decreases drastically, with a rather uniform distribution down the deeper sphagnum layers (Table 61).

Table 59. The effect of thickness and composition of the forest litter (A0) on ^{137}Cs uptake by vegetation (n = 5–30)

Key site	Type of biogeocenosis (A0l composition)	Thickness (cm)			^{137}Cs content, % of total in the soil profile		
		m	max	min	m	max	min
L-1	Broad-leaved-pine forest (leaves and needles)	3.9	7	1.5	26.5	50.1	10.3
E-1	Broad-leaved/spruce forest (leaves and needles)	4.9	7.5	2.5	52.8	69.1	21
Kh-1	Pine forest (needles)	4.3	5	3.5	59.3	78	16
Sh-5	Pine forest (needles)	4.5	12	0.5	83.4	98.4	9.5
E-3	Spruce forest (moss and needles)	7.3	7.5	6.5	91.8	93	68.4
L-2	Pine forest on a sphagnum bog (dead sphagnum)	11.3	18.5	5.5	76	89	60
Kh-4	Alder forest (alder leaves)	3.1	4	2	23.1	38	12

Footnote: m- mean; max, min - minimum and maximum, respectively

In the absence of a manifested moss cover, or in the case of a thin or peatlike forest litter, the rate of radionuclide migration from the forest litter to lower layers of hydromorphic soils increases, which is apparently due to a high moisture in the soil profile and a high content of the decomposed organic matter (Fig. 52, 53). For example, about 80% of total radionuclide inventory was replaced from the soil litter to the mineral soil in the hydromorphic alder forest by 1995.

Deciduous forests form, as a rule, a thin forest litter with an incomplete profile. Its accumulative capacity is comparatively low, and radionuclides are actively replaced from the forest litter to mineral horizons (Table 59).

Even lower accumulative capacity is characteristic of a thin and fragmentary forest litter under forest-steppe vegetation on chernozems and gray forest soils [280]. The structure of such forest litters is usually incomplete and composed of fresh litterfall that is being intensively decomposed by soil fauna.

Thus, radionuclide migration in the forest landscapes is determined primarily by the forest litter presence and its features. This specific soil horizon serves usually as an effective biogeochemical barrier preventing radionuclides from fast migration down the soil profile. Retention capacity of a forest litter

Table 60. The effect of moss cover on the retention capacity of the forest litter (data of 1992, average of n = 16)

Key site	Type of biogeo-cenosis	Thickness of forest litter (cm), presence of moss cover*	Layer	^{137}Cs, % **
Kl-1	Mixed, broad-leaved-pine forest	4.5 (+)	Forest litter	50.9
			Mineral layers	49.1
Zl-1	Pine forest	3.5 (–)	Forest litter	35.3
			Mineral layers	64.7
Nz-2	Pine forest	4.4 (–)	Forest litter	35.7
			Mineral layers	64.3
Km-1	Pine forest	4.3 (+)	Forest litter	59.9
			Mineral layers	40.1

*(+) moss cover is well manifested ; (-) moss cover is absent
**% of the total deposition (forest litter + mineral layers))

Table 61. Vertical distribution of ^{137}Cs in the upper peat layers of a sphagnum bog *(Sphagnum girgensohnii Russ)* (% of total content in the 0–14 cm layer)

Depth, cm	Year of formation	^{137}Cs (% of total)	Depth, cm	Year of formation	^{137}Cs (% of total)
0–2.0	1992	14.2	8.0–9.5	1977–1979	3.3
2.0–3.5	1989–1991	19.3	9.5–11.0	1974–1976	4.1
3.5–5.0	1986–1988	24.0	11.0–12.5	1971–1973	4.4
5.0–6.5	1983–1985	18.5	12.5–14.0	1968–1970	5.6
6.5–8.0	1980–1982	6.8	Total 0–14	1968–1992	100.0

depends on its type, sub-layer structure, thickness, and manifestation of moss (lichen) cover. The least retention capacity is characteristic of incomplete, thin forest litters under deciduous forests where the rate of radionuclide migration (in particular, ^{137}Cs) to the mineral soil layers is maximum. In general, forest litters may be ranked by their retention capacity as follows: coniferous forests with dense moss cover > coniferous forests with unpronounced rare moss > mixed broadleaved coniferous forests > deciduous forests.

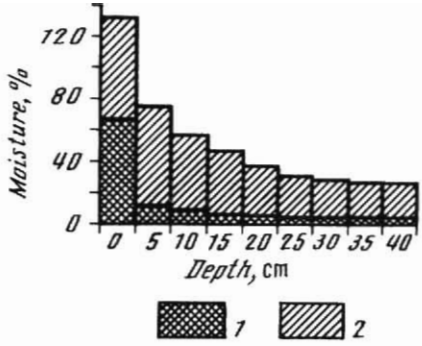

Fig. 52. Vertical distribution of the soil moisture in the automorphic (1) and hydromorphic (2) forest soils (July)

Individual subhorizons of forest litter (A0l, A0f, A0h) possess as a rule different retention capacity, and radionuclides are uniformly distributed within the forest litter layer (Fig. 53). During the first months after the accident, more than 90% of radionuclides were concentrated in the upper organic subhorizon (A0l or A0f). In the ensuing years, the radionuclide behaviour in forest litters depended on many factors.

The long-term dynamics of ^{137}Cs content in the leaf subhorizon A0l is characterised by a pronounced, monotonous, descending trend, particularly in the first years after the accident (Fig. 54). Later on, the rate of decrease slows down drastically, and by the 4–5th year after the accident, radiocaesium proportion in the A0l layer drops to about 1% of the total deposition. This is mainly due to fast decomposition of the A0l layer and its mechanical overlaying with a fresh, less contaminated litterfall. This natural decontamination of the upper subhorizon is most typical of coniferous biogeocenoses with a complete, well manifested forest litter (3 subhorizons). In the investigated deciduous forests, natural decontamination of A0l layer takes longer time, in spite of faster decomposition and replacement (average renewal cycle for deciduous and coniferous forests is about 1 and 2–3 years, respectively). This slower radionuclide dynamics in the deciduous forests may be explained by two main reasons: (i) inadequacy of coniferous A0l layer to that in deciduous cenoses, and (ii) higher contamination of leaves compared to needles [98].

The dynamics of ^{137}Cs in A0f (fermentative) and A0h (humus) subhorizons is different from that in A0l horizon. The radionuclide proportion in these layers increases up to some maximum level, then decreases and remains stable at some average level depending on ecosystem, landscape, thickness, and structure of the forest litter as a whole. Reaching the equilibrium radionuclide content in A0f subhorizon takes about 10 times longer time compared to A0l. The equilibrium content in A0f is somewhat lower than in the A0h layer. In the marginal part of the exclusion zone and the remote contaminated areas, reaching the equilibrium level takes about 8–9 years. This process is longer in the "near zone" (for each sublayer), which is likely due to specific physicochemical properties of the initial fallout in the vicinity of ChNPP [2–6]. The equilibrium level is usually higher in

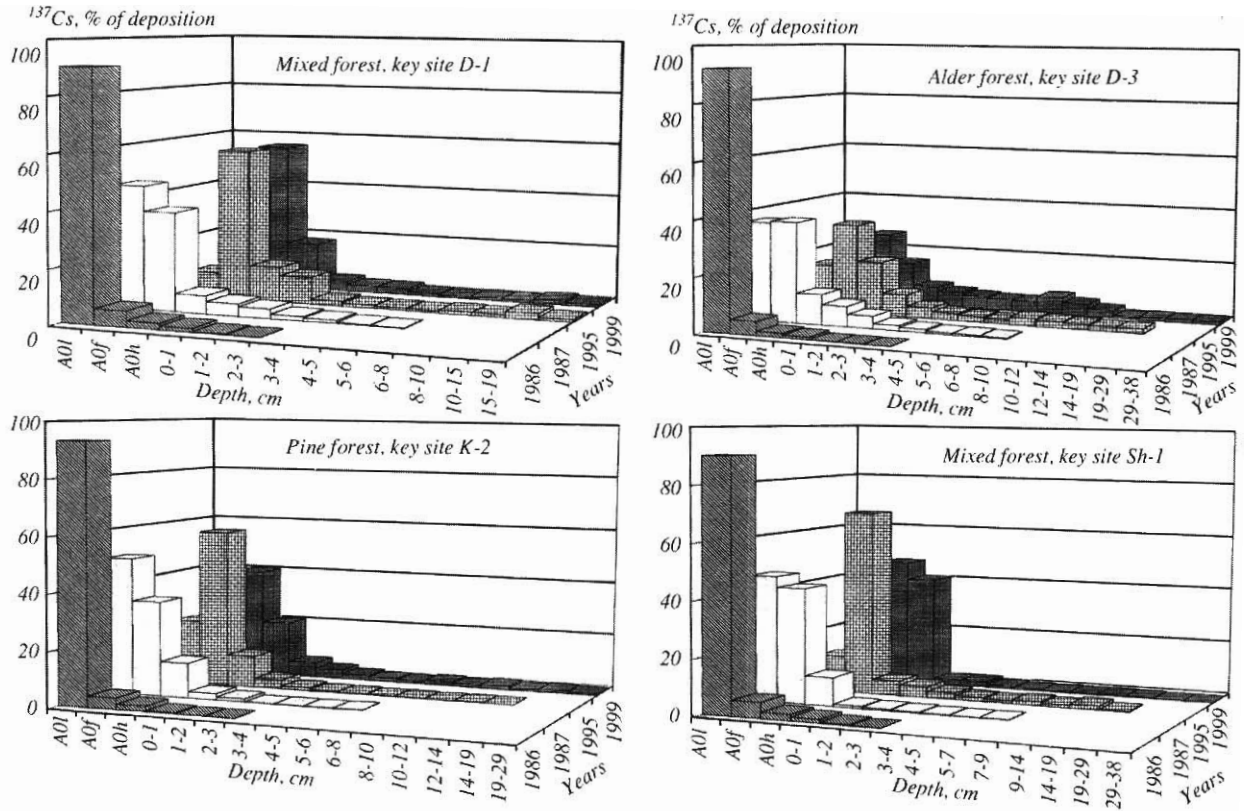

Fig. 53. Multiyear dynamics of ^{137}Cs vertical redistribution

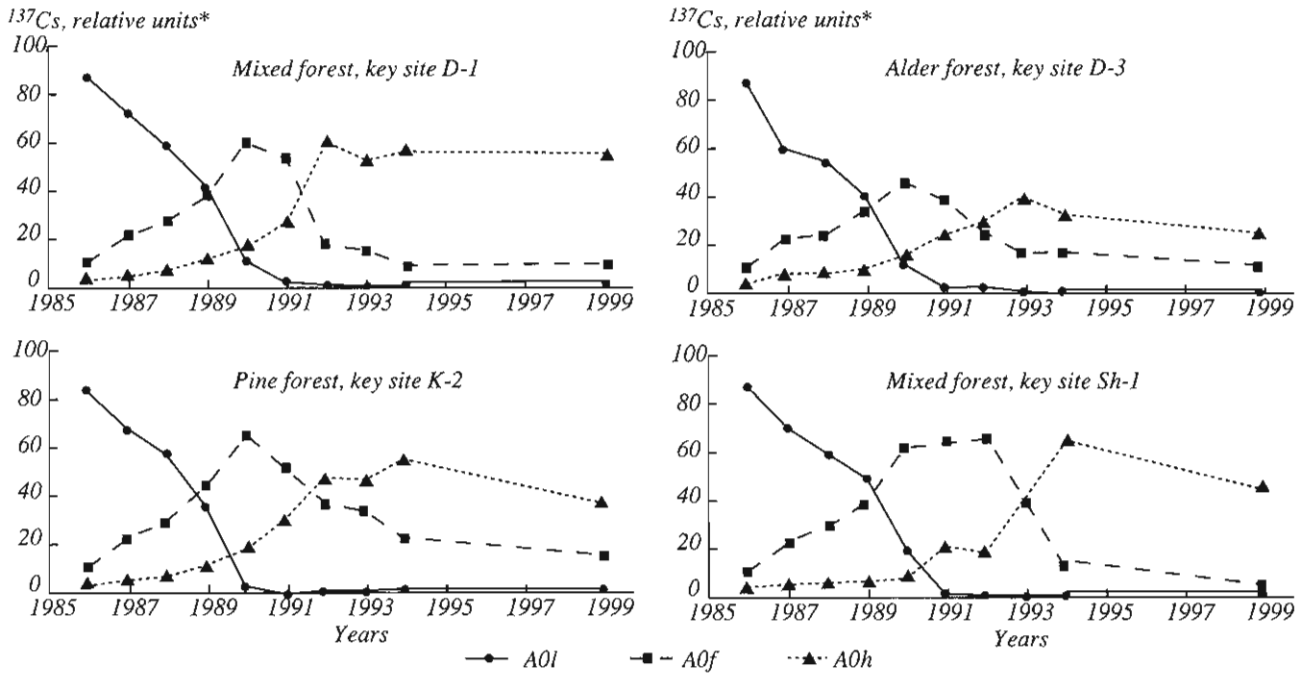

Fig. 54. The long-term dynamics of ^{137}Cs redistribution in the forest litter at various key sites:
(1) A0l; (2) A0f; (3) A0h
* % of the inventory in the forest litter

Table 62. Multiyear dynamics of ^{137}Cs redistribution among the forest litter and mineral layers of forest soils (% of the total content in the soil profile)

Key site	Layer	Years								Mean annual loss from A0
		1987	1988	1989	1990	1991	1992	1994	1995	
		30 km exclusion zone (Ukraine)								
D-1	A0*	93.9	92.9	92.4	88	83	80.4	68.9	66.3	3.74
	ML	6.1	7.1	7.6	12	17	19.6	31.1	33.7	nd**
D-3	A0	93.4	88.2	85.6	74.6	66.4	58.3	60.1	51.3	5.41
	ML	6.6	11.8	14.4	25.4	33.6	36.7	39.9	48.7	nd
K-2	A0	97.4	95.9	91.8	90.4	89.9	90.5	86	82.6	1.93
	ML	2.6	4.1	8.2	9.6	10.1	9.5	14	17.4	nd
Sh-1	A0	94.1	91	93.3	91.6	88.2	86	82.8	81.5	2.05
	ML	5.9	9.0	6.7	8.4	11.8	14	17.2	18.5	nd
		Tula region (Russian Federation)								
Pl-1	A0	nd	58.3	42.1	15.2	5	0.5	nd	nd	14.2
	ML	nd	41.7	57.9	84.8	95	99.5	nd	nd	nd
Pl-2	A0	nd	44.9	35.4	11.5	2.4	0.1	nd	nd	14.3
	ML	nd	55.1	64.6	88.5	97.6	99.9	nd	nd	nd
		Bryansk region (Russian Federation)								
Kl-1	A0	nd	79.2	75.3	70.5	69.1	62.5	nd	nd	5.4
	ML	nd	20.8	24.7	29.5	30.9	37.5	nd	nd	nd
Kl-2	A0	nd	80.4	70.1	57	57.2	50.9	nd	nd	7.01
	ML	nd	19.6	29.9	43	42.8	49.1	nd	nd	nd
Zl-1	A0	nd	91.5	90	80.9	70.8	58.1	nd	nd	6
	ML	nd	8.5	10	19.1	29.8	41.9	nd	nd	nd

* A0 – forest litter; ML– mineral soil layers

** No data

coniferous forests on automorphic soils, compared to the cenoses growing in hydromorphic areas.

Reaching the equilibrium value in A0h layer of the forest litter requires even longer time than in A0f. The maximum content of ^{137}Cs in the latter is reached by the 5–6th year after the fallout, and the steady state is reached by the 8th year, whereas the radionuclide content in A0h subhorizon still increases in the most localities, though the rate of increase has been slowing down since 1992. The only exclusion is the forest environments in the "near zone" where an increase in the radionuclide content still continued in 1996 (10 years after the accident). It may be supposed that, unlike in the A0f layer, the radionuclide content in the A0h layer will never exhibit a pronounced peak.

Thus, the key factors of the radionuclide redistribution within the soil litter are (i) permanent addition of the low-contaminated organic matter ("clean" litterfall), and (ii) high rate of transformation. The dynamics and intensity of decontamination processes depends on a subhorizon of the forest litter. Leaf (A0l) layer exhibits the highest rate of decontamination: ^{137}Cs content in this layer decreased twofold by the second year after the accident and reached its equilibrium value (about 1% of the total deposition) by the 4–5th year after the fallout. The corresponding quasi-equilibrium radionuclide content in A0f layer (10–20%) is reached by the 8–9th year after the accident. The corresponding equilibrium in A0h layer is not reached yet. Thus, the half-stay period (effective half-life) of radionuclides should be calculated for each subhorizon separately, taking into account the above-discussed features of the radionuclide dynamics.

Proceeding from the radionuclide pattern of 1995–96 (on the average, over 70% of the deposition in the contaminated territory is still concentrated in the forest litter), the rates of annual radionuclide migration and transport to the mineral soil layers are presented in Table 62.

In the automorphic areas, the average rate of annual replacement of the radionuclides from forest litter to deeper mineral soil layers is about 1.9–3.7% in Ukrainian Polessie and 5–6% in the Russian Federation. In hydromorphic forest environments, the corresponding value is about 7% in the entire contaminated territory.

The rate of annual radionuclide replacement from forest litter to the mineral layers depends on the following factors:

1. **Soil properties and regimes**. In hydromorphic areas, the rate is much higher than in the automorphic areas. This is due to the above-discussed specific features of hydromorphic and automorphic soils, on the one hand, and different pedogenetic processes running under the coniferous and mixed stand, on the other hand [59, 157].

2. **Distance from the accidental unit**. The rate of ^{137}Cs migration from forest litter to the mineral soil layers in the territory of the Russian Federation is higher than in the exclusion zone by a factor of 1.5.

3. **Weather conditions**. Annual rate of ^{137}Cs migration from forest litter to the mineral horizons varies by 0.5–11%. The variation is well agreed with the dynamics of rainfall during the vegetative season: the migration rate increases drastically in the wet years.

4.4.2. Radionuclide Content and Dynamics in Mineral Soil Layers

A proportion of radionuclides replaced to mineral (organic-mineral) soil horizons laying under the forest litter is a reciprocal of the retention capacity of the latter. Unlike A0, mineral layers of forest soils receive usually mobile forms of radionuclides (except for the cases of *lessivage*, and the areas devoid of forest litter). Consequently, radionuclide distribution down the mineral soil profile depends primarily on the soil properties rather than specific features of the initial fallout.

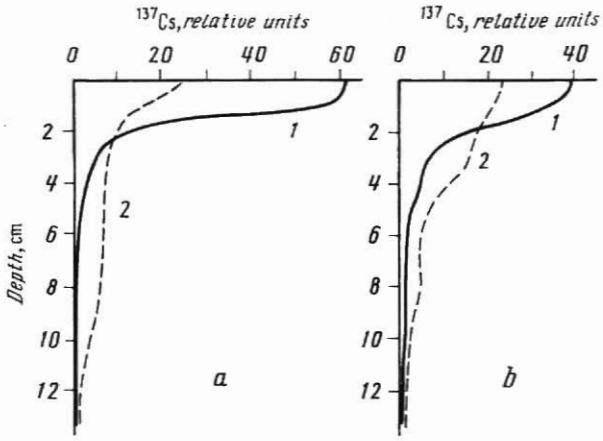

Fig. 55. Vertical distribution of ^{137}Cs in the mineral layers of forest soils (% of the total inventory in the mineral layers):

(a) exclusion zone (key sites D-1 and D-3); (b) "remote" zone" (Bryansk Region, key sites Kl-1 and Kl-2)

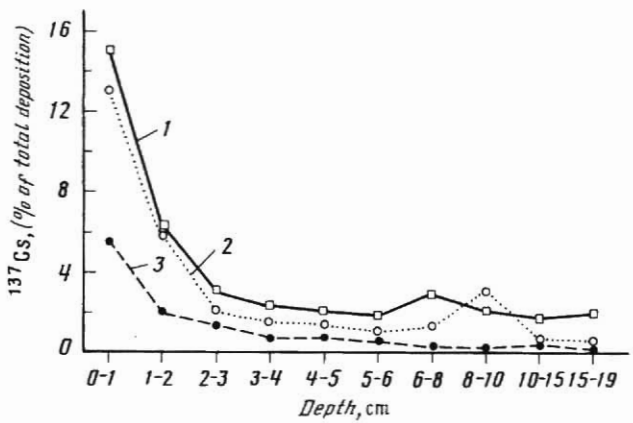

Fig. 56. Vertical distribution of ^{137}Cs in the mineral soil layers in various forest biogeocenoses (% of the total deposition):

(1) broad-leaved/pine forest, hydromorphic area; (2) broad-leaved/pine forest, automorphic area; (3) pine forest, automorphic area

In automorphic soils, ^{137}Cs is retained in the upper 1–2 cm of the mineral profile laying immediately under the forest litter (Fig. 55). Below this thin layer, the radionuclide content decreases drastically and reaches the background level at the depth of 30–70 cm depending on the deposition. In other words, 10 years after the deposition, the maximum penetration depth of radiocaesium in the automorphic areas does not exceed this range (30–70 cm).

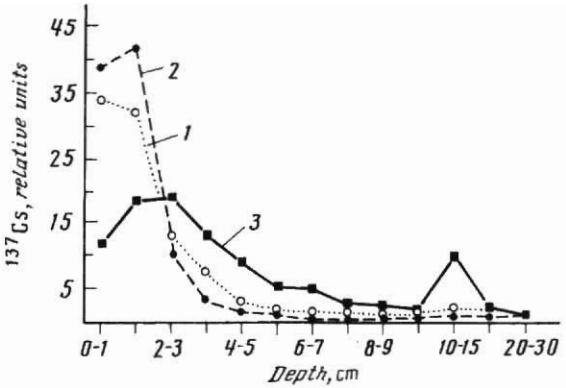

Fig. 57. Vertical distribution of ^{137}Cs in meadow soils in various biogeocenoses, % of deposition:

(1) boggy meadow (a plate-like depression); (2) old-tilled automorphic area; (3) floodplain meadow (the Pripyat' river valley)

In hydromorphic soils, the intensity of radionuclide migration is about 2–3 times higher than in the automorphic soils (Figs. 53 and 55). The radionuclide concentration decreases down the soil profile much slower, and the radionuclide accumulation beneath the forest litter is not as pronounced as in the automorphic soil. This is likely due to weak irreversible adsorption of ^{137}Cs in the upper horizons of peat or peat-like hydromorphic soils. The maximum migration rate of ^{137}Cs is observed in hydromorphic soils under alder forest. The migration rate decreases in the alluvial soils on the Pripyat river floodplain, and reaches the absolute minimum in the oligotrophic sphagnum bogs. In the latter, ^{137}Cs proportion in the lower horizons is much less compared to other hydromorphic soils. This is apparently due to high sorption capacity of sphagnum and its specific stratified structure impeding water flow through the sphagnum layers.

Hydromorphic soils are often attributed to the topographical depressions with a high water table (floodplains, bogs, plate-like depressions, etc.). High rates of vertical radionuclide migration in the hydromorphic areas makes the latter a preferable object of radioecological monitoring, since a significant portion of radionuclides may enter ground water in such "critical areas".

Type of ecosystem has a considerable effect on the radionuclide distribution down a mineral soil profile (Fig. 56). A high rate of ^{137}Cs migration from forest litter to beneath mineral layers is characteristic of deciduous forests, which is likely due to a specific composition of the litterfall, a high rate of its transformation, a high content of mobile organic compounds, and some other pedogenetic features. Both the decomposition rate and the flux of mobile compounds are known to be higher in the soils under deciduous and mixed forests compared to pure coniferous cenoses [208, 209]. The rate of radionuclide migration in the latter is higher under aged stand compared to young trees.

Radionuclide migration under meadow vegetation is considerably less intensive than that under the forest canopy (Fig. 57). The maximum radionuclide content in the first is still detected in the upper 2–3 cm [184, 195]. Below this layer, the radionuclide content decreases sharply and reaches the

Table 63. Variation of the radionuclide composition down the soil profile (1990, %)

Depth (cm)	30 km exclusion zone (D-1)				Tula region, Russian Federation (Pl-1)			
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru
0–1	18.5	8.6	57.7	15.2	bd*	10.7	84.4	4.9
1–2	20.0	7.9	59.8	12.3	bd	12.4	87.6	bd
2–3	18.8	8.3	60.1	12.8	bd	11.8	88.2	bd
3–4	17.2	9.2	73.6	bd	bd	11.8	88.2	bd
4–5	13.2	9.3	77.5	bd	bd	11.6	88.4	bd
5–6	bd	11.4	88.6	bd	bd	12.6	87.4	bd
6–7	bd	11.5	88.5	bd	bd	12.6	87.4	bd
7–8	bd	11.5	88.5	bd	bd	12	88	bd
8–9	bd	12.0	88	bd	bd	11	89	bd
9–10	bd	12.0	88	bd	bd	11	89	bd
10–11	bd	12.0	88	bd	bd	10.5	89.5	bd
11–12	bd	bd	100	bd	bd	11	89	bd
12–13	bd	bd	100	bd	bd	11.2	88.8	bd
13–14	bd	bd	100	bd	bd	11.8	88.2	bd
14–15	bd	bd	100	bd	bd	10.1	89.9	bd
15–20	bd	bd	100	bd	bd	bd	bd	bd

* Below detectable

background at the depth of 6–7 cm. The only exclusion is soils on wet floodplain where the migration rate is abnormally high compared to the neighbouring forest soils.

The process of radionuclide migration to deeper soil layers is accompanied by farther differentiation of the radionuclide composition: the proportion of ^{137}Cs increases and proportion of ^{144}Ce and ^{106}Ru decreases down the soil profile (Table 63). This is apparently due to different chemical properties of radionuclides [135, 154]. $^{137,134}Cs$ and ^{144}Ce exhibit the highest and lowest migration rates, respectively (the behaviour of ^{90}Sr is considered below in Chapter 4.5).

The migration rate of ^{106}Ru is higher in pure pine forests compared to deciduous and mixed cenoses. This is likely due to the fact that mobile ruthenium may exist in soils in both cationic and anionic radionuclide-organic forms [211]. The mobility of anionic ^{106}Ru increases by 2–3 orders of magnitude in the presence of fulvic acids [42]. Ruthenium mobility is reported to be particularly sensitive to pH variation [211, 317]. All these factors may have a synergetic effect on the ^{106}Ru migration rate in the soils under pine stand [60, 166].

Unlike in forest litters, the radionuclide content in the mineral soil horizons is characterised by a monotonous increase with time (Fig. 58). The most intensive transport of ^{137}Cs to the upper 5-cm soil layer takes place in bog soils

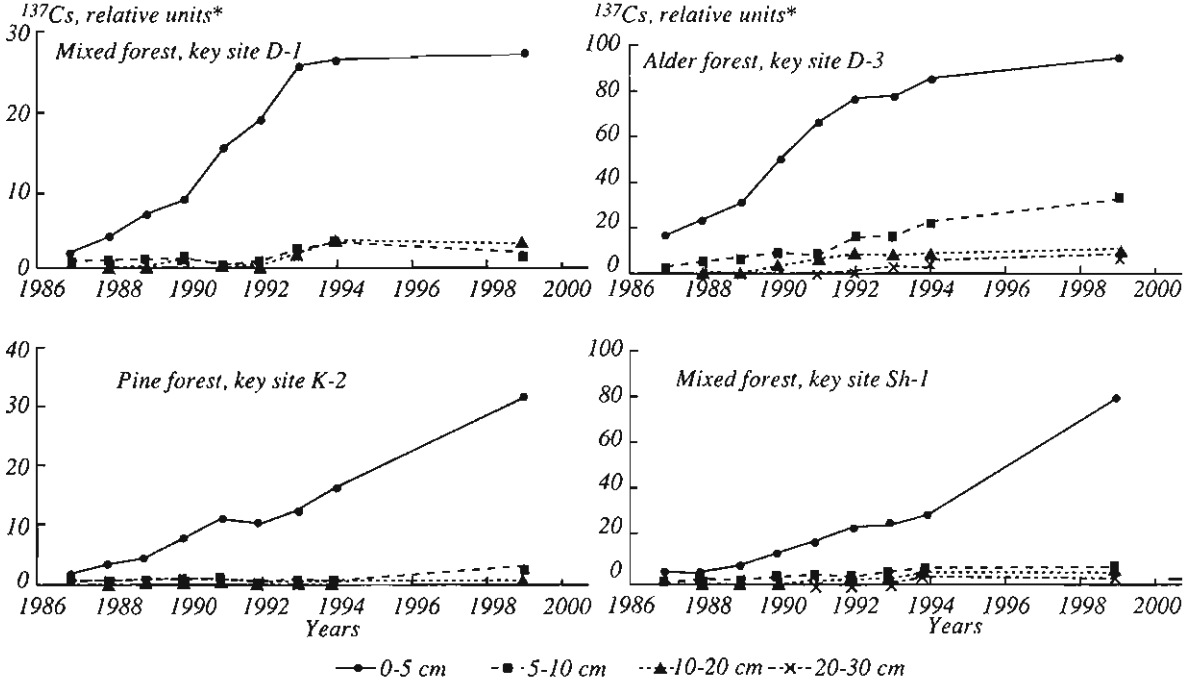

Fig. 58. The dynamics ^{137}Cs vertical distribution in the mineral layers of various forest soils (30 km exclusion zone, Ukraine)
* % of deposition

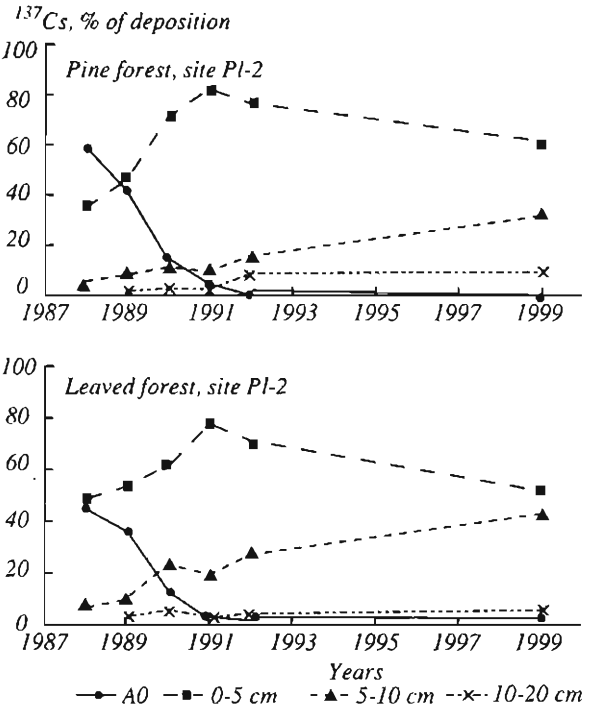

Fig. 59. The dynamics of ^{137}Cs vertical distribution in the forest litter and mineral layers in various forest soils ("remote zone", Bryansk region, Russian Federation)

(except for sphagnum bogs). In the automorphic areas of the remote zone, deciduous and mixed forests provide the most favourable conditions for vertical caesium migration. The least migration rates of this radionuclide are observed in the automorphic soils under pine cenoses in the vicinity of ChNPP. An annual increment of ^{137}Cs in the mineral soil layer of 0–5 cm is about 4% in hydromorphic areas, 3% in automorphic areas of the remote area, and 1.5% in the close vicinity of ChNPP (the "near zone"). The corresponding values for the layer of 5–10 cm are 1.0, 0.5, and 0.1–0.4%.

In deciduous forests of the forest-steppe zone, the dynamics of ^{137}Cs migration in chernozems is similar to that in soddy-podzolic sand soils under broad-leaved coniferous and pure coniferous cenoses. Surprisingly, the rate of ^{137}Cs migration to the mineral horizons of chernozems is higher than in soddy-podzolic soils (Fig. 59, Table 62). Six to seven years after the fallout, the upper 5 cm layer of chernozem soils contained about 70% of total deposition, i.e., the average rate of annual migration in the automorphic chernozem exceeded 10%, which was higher than the corresponding rate in any hydromorphic sandy soil. A similar high rate is characteristic of the layer of 5–10 cm (4%). Thus, there is some specific process that determines an abnormally high rate of radionuclide redistribution down the chernozem profile. Let us analyse this interesting phenomenon in a more detail.

Table 64. Earthworm population in various forest ecosystems (individuals per sq.m)

Region	Forest type	Population	Reference
Tula region	Broad-leaved	200–300	[266]
Bryansk region	Mixed, broad-leaved/coniferous	96	[271]
	Pine	18	[271]

In principle, there is a rather limited number of processes which can have effect on the mass redistribution in the soil profile: diffusion, mass transport by a filtering water, and biogenic transport (activities of soil fauna). The first two pathways cannot play a significant role in the case: the diffusion rate in chernozems is extremely low, and the radionuclide transport by the intrasoil flow is less than a tenth of a percent per year even in sand soils [242]. In chernozems, the mass-transport shall be even less, since the rainfall in the forest-steppe natural zone does not provide sufficient water percolation [52]. Solubility of the initial fallout also cannot serve as a key factor, since the fallout properties in Tula region (chernozems) are the same as those in Bryansk region (soddy-podzolic soils). Thus, the biogenic transport is likely to be the only process determining an abnormally fast radionuclide migration in chernozems. This is confirmed by the fact that such a high migration rate is observed only in the layer of 0–10 cm where the soil fauna is most active.

It is reported that soil invertebrates, in particular earthworms, can replace up to 600 t/ha of soil per year by blending soil layers [70, 178, 266, 271]. The intensity of soil blending is directly proportional to the earthworm population in the soil (Table 64). Earthworms can replace more than 50% of radionuclides from the soil surface to deeper horizons in a rather short time (weeks and months). In this connection, it is of interest that the radionuclide content in the worm coprolites is twice as much as in the surrounding soil [249]. Differences in the earthworm population may be sufficient to cause observable variation of the radionuclide distribution even in the same soil type (chernozem) under coniferous (pine) and mixed (broad-leaved coniferous) forest. The earthworm population is much higher in the latter because of a favourable food resource [178]. The effect of soil invertebrates is believed to be a possible reason of a considerable and fast increase in ^{137}Cs content in the ground water sources in the chernozem zone [222].

In other chernozem soils, the contribution of soil fauna to vertical radionuclide migration is not as high, since the earthworm abundance in the deep-frosted soils decreases. That is why, the vertical distribution of radionuclides in the profile of leached chernozem in the East Ural is very different from the corresponding distribution in the leached chernozems in the Russian Plain.

Thus, the rate of the vertical radionuclide redistribution depends on a number of pedogenetic processes, and various factors and indices may set the pace depending on soil properties, weather conditions and time after the accident. A

specific feature of all forest soils is forest litter (A0), that is another factor complicating the radionuclide behaviour in these soils. To describe and predict the vertical radionuclide migration in forest soils, forest litter shall be considered separately from the rest of the soil profile, since radionuclide migration and retention in these soil components are controlled by different factors and processes. Describing the worm-abundant forest-steppe and steppe soils (and also floodplain and sod soils), soil fauna shall be taken into account as a significant factor of radionuclide redistribution.

4.4.3. Vertical Distribution of ^{40}K

Distribution of natural radionuclide ^{40}K in the soils is practically the same as its stable isotopic carrier ^{39}K. The latter is known to be chemically similar to caesium (the so-called non-isotopic carrier). Therefore, the present distribution of ^{40}K down the soil profile may serve as an approximation of the future distribution of ^{137}Cs over a long period of time (several decades). In general, the investigated soddy-podzolic sand soils of Ukrainian Polessie have a rather uniform profile of ^{40}K, without its significant accumulation in or loss from the soil horizons (Fig. 60). Such a distribution of potassium is characteristic of local sandy soils poor of clay minerals [33, 46]. Although ^{40}K (and the total stable potassium) content decreases with depth, its corresponding reserves increase because the soil bulk density increases down the soil profile.

Taking into account the data on ^{40}K, it may be supposed that the highest ^{137}Cs concentration in the automorphic soddy-podzolic soil profile will remain in the forest litter. ^{137}Cs share in this horizon (relative to the total deposition) will decrease gradually with the annual rate of 2-4% (Table 65, see also the next chapter for details). The vertical gradient of ^{137}Cs in the soil profile will decrease, and its distribution will smooth to approximate finally to the present distribution of stable potassium (and ^{40}K). Such a distribution has already been reached in some hydromorphic soils where forest litter exhibits the maximum concentration of ^{137}Cs, but the maximum inventory of this radionuclide is attributed to the mineral layers. At the same time, the total loss of ^{137}Cs from the root-abundant soil layer is unlikely to be high because, on the one hand, caesium will be irreversibly adsorbed by the soil (in spite of low adsorption capacity of sands) and, on the other hand, it will be "intercepted" by the plant roots and included to the biological cycle [242, 277].

4.4.4. Forecasting Radionuclide Redistribution Between Forest Litter and Mineral Soil Layers

The radionuclide content dynamics in forest litter for 1986-1997 is well approximated by the equation:

$y = a_0 + a_1/x$; (Figs. 61, 62),

where a_0 and a_1 are the empirical coefficients depending on ecosystem as follows:

1. Coniferous cenoses: $a_0 = -3090$; $a_1 = 6.33 \cdot 10^6$

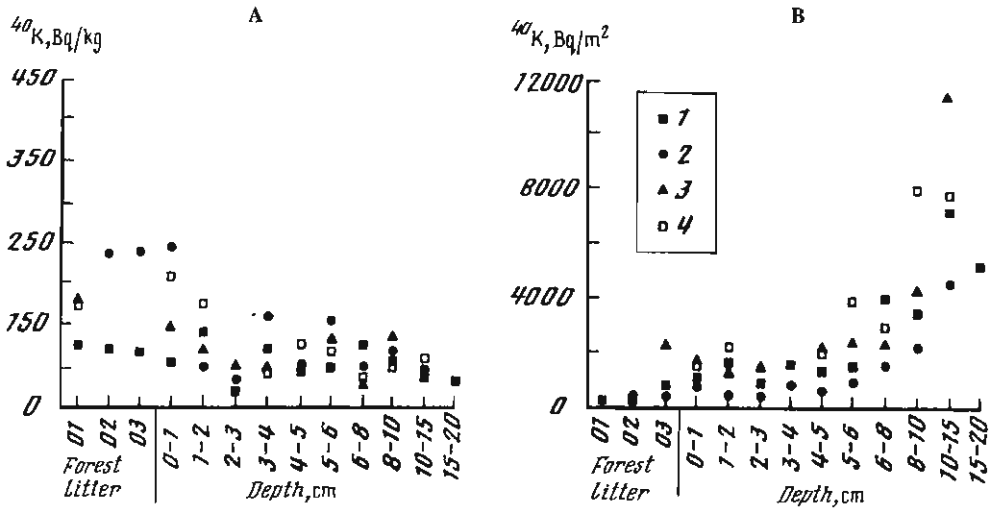

Fig. 60. The content (A) and reserves (B) of ^{40}K in forest litters and mineral soil layers; key sites: (1) D-1; (2) D-3; (3) K-2; (4) Sh-1

Table 65. A forecasted dynamics of ^{137}Cs redistribution between the forest litter and mineral soil layers (% of the total deposition in the soil profile)

Years	Key sites			
	D-1	D-3	K-2	Sh-1
	Forest litter (A0)			
1996	65.49	45.49	82.34	79.91
1997	62.07	40.27	80.75	78.28
1998	58.66	35.06	79.16	76.65
1999	55.25	29.85	77.58	75.02
2000	51.84	24.65	75.99	73.4
2001	48.44	19.45	74.41	71.77
2002	45.04	14.26	72.83	70.15
2003	41.65	9.074	71.25	68.53
2004	38.25	3.893	69.67	66.91
2005	34.86	–	68.1	65.29
	Mineral layers			
1996	34.51	54.51	17.66	20.09
1997	37.93	59.73	19.25	21.72
1998	41.34	69.94	20.84	23.35
1999	44.75	70.15	22.42	24.98
2000	48.16	75.35	24.01	26.6
2001	51.56	80.55	25.59	28.23
2002	54.96	85.74	27.17	29.85
2003	58.35	90.93	28.75	31.47
2004	61.75	96.11	30.33	33.1
2005	65.14	101.3	31.9	34.71

2. Mixed broad-leaved coniferous forests: $a_0 = 6856$; $a_1 = -1.36 \cdot 10^7$
3. Alder forest: $a_0 = -1.04 \cdot 10^4$; $a_1 = 2.08 \cdot 10^7$
4. Mixed broad-leaved coniferous forests in the 30 km exclusion zone: $a_0 = -3178$; $a_1 = -6.5 \cdot 10^6$

The radionuclide content dynamics in the mineral soil layers is approximated by a similar equation (Figs. 61, 62) with the following coefficients:

1. Coniferous cenoses: $a_0 = -3190$; $a_1 = -6.33 \cdot 10^6$
2. Mixed broad-leaved coniferous forests: $a_0 = 6856$; $a_1 = -1.36 \cdot 10^7$
3. Alder forest: $a_0 = 1.05 \cdot 10^4$; $a_1 = -2.09 \cdot 10^7$
4. Mixed broad-leaved coniferous forests in the 30 km exclusion zone: $a_0 = 3278$; $a_1 = -6.5 \cdot 10^6$

Using these equations, we calculated probable radionuclide partitioning in forest litters and mineral soil layers up to 2005 (Table 65). Over 50% of the deposition will be likely replaced from forest litter to the mineral soil

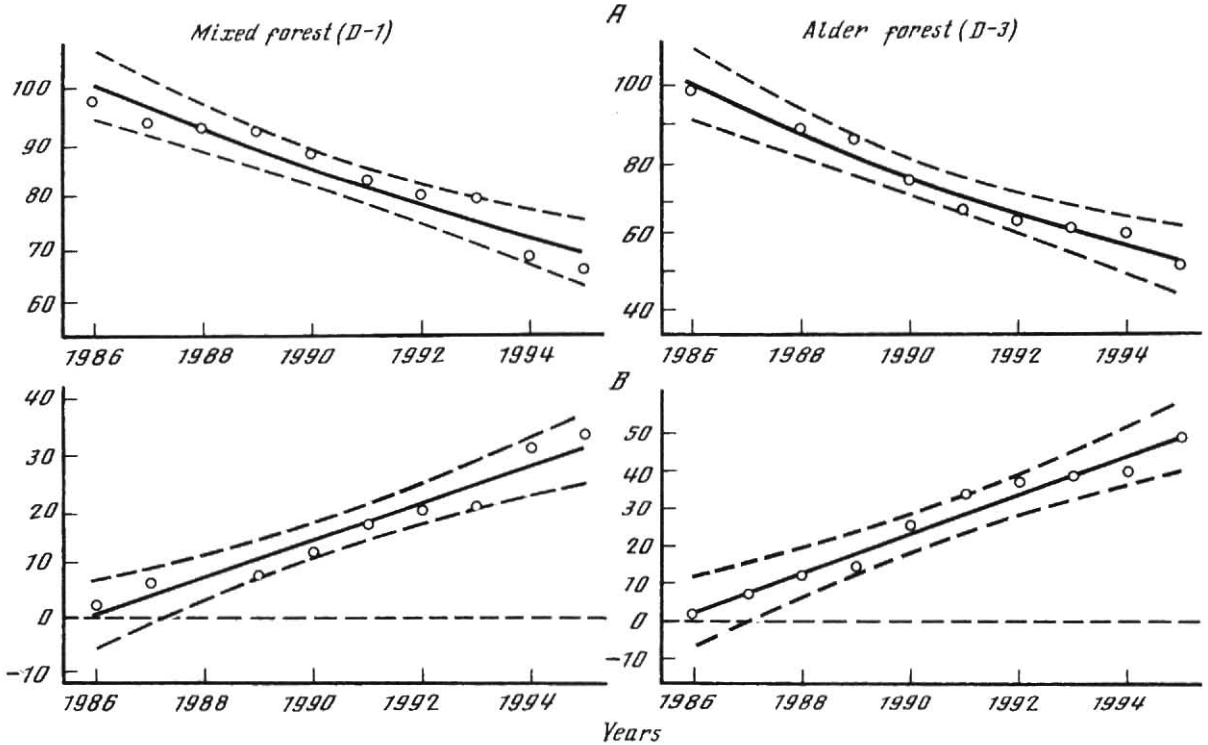

Fig. 61. The dynamics of ^{137}Cs content in (A) forest litter and (B) mineral soil layers (marginal areas of the 30 km exclusion zone, %)

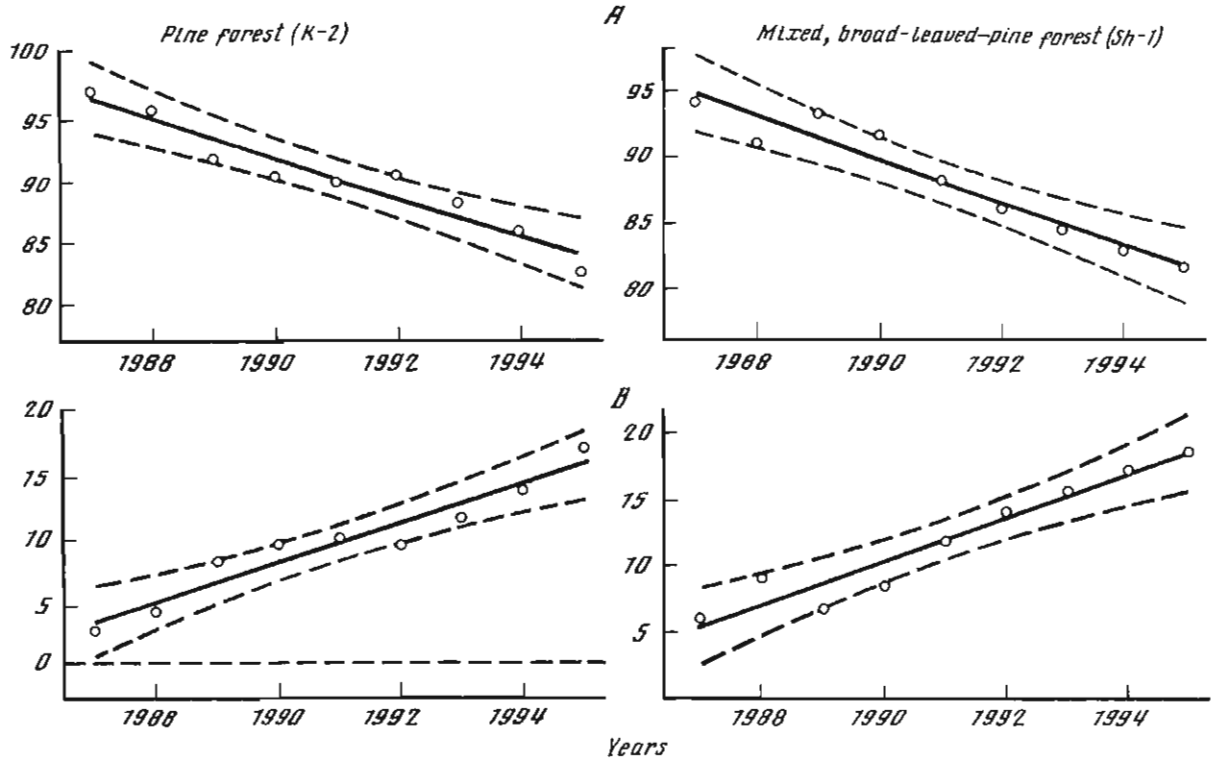

Fig. 62. The dynamics of ^{137}Cs content in forest litter (A) and mineral layers (B) (the near zone, 5–7 km from ChNPP, %)

horizons in automorphic areas. In hydromorphic areas, quasi-equilibrium in the system "stand–forest litter–mineral layers" is expected by 2005. In the "near zone" (closer than 5 km), up to 60–70% of the deposition will remain in the forest litter.

4.5. VERTICAL DISTRIBUTION OF ^{90}Sr

Radiological hazard of ^{90}Sr is the same or even higher than ^{137}Cs, since radiostrontium is a long-lived radionuclide ($T_{1/2} \approx 28$ years) similar to calcium by its chemical properties.

$^{137}Cs/^{90}Sr$ ratio varies over the investigated territory from 1.7–2.2 (in the exclusion zone) up to 42 in the remote zones of the Russian Federation, which is due to the composition of the initial fallout [96]. Vertical distribution of ^{90}Sr in the soil is generally similar to ^{137}Cs. Like ^{137}Cs, ^{90}Sr content is maximal in the upper organic layer, and decreases drastically down the mineral soil profile. It means that vertical distribution of both radionuclides is determined by the same basic processes, i.e., diffusion, mass transport by water flow ("convection"), and biogenic transport. The distribution of ^{90}Sr has, however, some specific features. Normally, a higher portion of ^{90}Sr than ^{137}Cs is transferred from forest litter to mineral layers. The accumulation of this radionuclide in the first mineral layer is less manifested than ^{137}Cs, and the vertical distribution of ^{90}Sr down the mineral profile is smoother. These facts suggest that the distribution of ^{90}Sr is more dependable on the mass transport than ^{137}Cs, which is due to different chemical properties of these radionuclides [125, 212, 223]. Unlike ^{137}Cs, the highest proportion of ^{90}Sr in the "solid-liquid" soil system is represented by its exchangeable [129, 156], i.e., potentially mobile forms. Therefore, ^{90}Sr migration down the soil profile with an intrasoil water flow is more intensive, ^{90}Sr is distributed more uniformly, and penetrates deeper than ^{137}Cs. The data of in situ lysimetric studies confirm that the proportion of ^{90}Sr in the lysimetric waters is about one order of magnitude higher than ^{137}Cs. The proportion increases with depth, since a considerable proportion of ^{137}Cs is fixed irreversibly from the liquid soil phase by the mineral components [114].

Variation of the physicochemical properties of the initial fallout over the contaminated territory [130] has affected not only the $^{137}Cs/^{90}Sr$ ratio, but ^{90}Sr vertical distribution as well (Fig. 63). The proportion of ^{90}Sr accumulated in forest litter decreases as the distance from ChNPP increases, i.e., the migration ability of radiostrontium in the "near zone" is minimum. Laboratory experiments show that the ^{90}Sr extractability from the soil samples increases as the distance from the accidental unit increases [75].

The smoothest distribution of ^{90}Sr down the soil profile takes place in the forest-steppe chernozems (Tula region, the Russian Federation) (Fig. 63). Since 1995, vertical distribution of ^{90}Sr in these soils is similar to the distribution of organic matter and nutrient, and (like ^{137}Cs) depends on the biogenic transport rather than either diffusion or infiltration mechanisms of migration.

The fallout properties in the investigated area being equal, ^{90}Sr migration is more manifested in purely coniferous forests than in mixed forests. This is likely due to different humus composition in the soils under coniferous and decid-

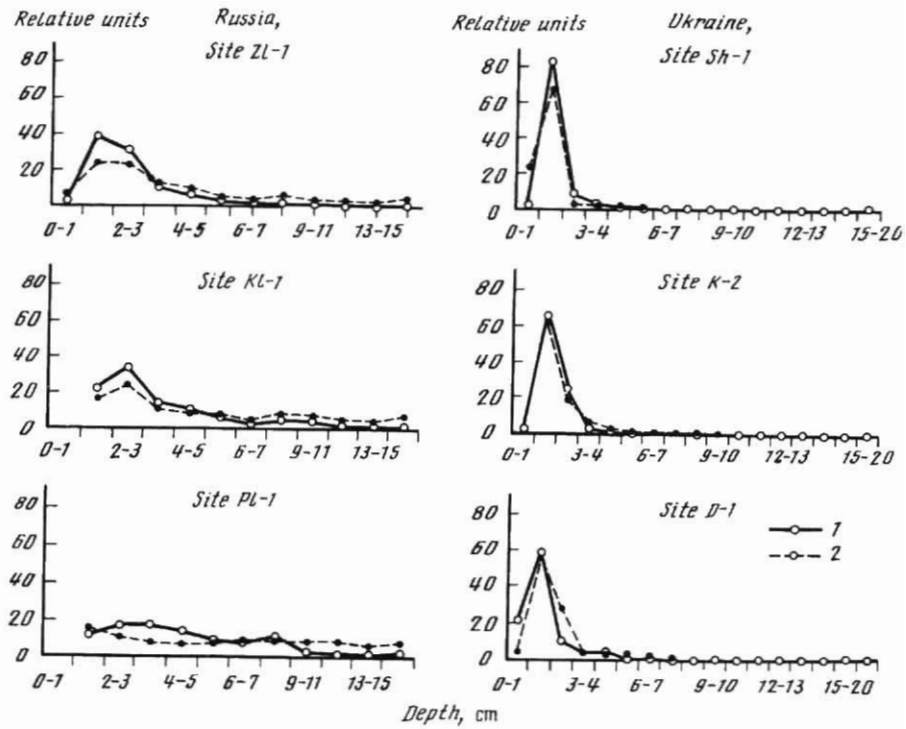

Fig. 63. Vertical distribution of (1) ^{137}Cs and (2) ^{90}Sr in the investigated forest soils (% of deposition)

uous forests. The first is normally poor of high-molecular humus acids and has the fulvate humus composition [166]. ^{90}Sr is believed to form intimate radionuclide-organic mobile compounds with fulvic acids, which promotes its migration ability. In addition, a higher hydrolytic acidity of these soils decreases absorption of ^{90}Sr by the soil solids [171]. The effect of low-molecular soluble organic products of decomposition of fresh litterfall and forest litter also affect the migration activity of radiostrontium [264, 265].

The soil moisture regime has a considerable effect on ^{90}Sr vertical distribution (Fig. 64). In hydromorphic soils, the proportion of ^{90}Sr replaced from forest litter to the mineral layers is by a factor of 1.5 higher than in the automorphic soils. On the other hand, fast migration of ^{90}Sr is also characteristic of automorphic soils in old-tilled areas (Fig. 64). In this case, fast migration of ^{90}Sr is likely to be due to a relatively high content of Ca in the agriculturally developed soils. Calcium is known to compete ^{90}Sr from absorption by the soil, which increases the mobility of the latter [34, 191].

In general, the soils in the region are extremely poor of clay minerals, which promotes a relatively high mobility of ^{137}Cs. Organic horizons of the soils are capable of ^{90}Sr retention rather than ^{137}Cs [315, 316]. Both the abovementioned factors promote the ^{137}Cs mobility and reduce the ^{90}Sr mobility. It

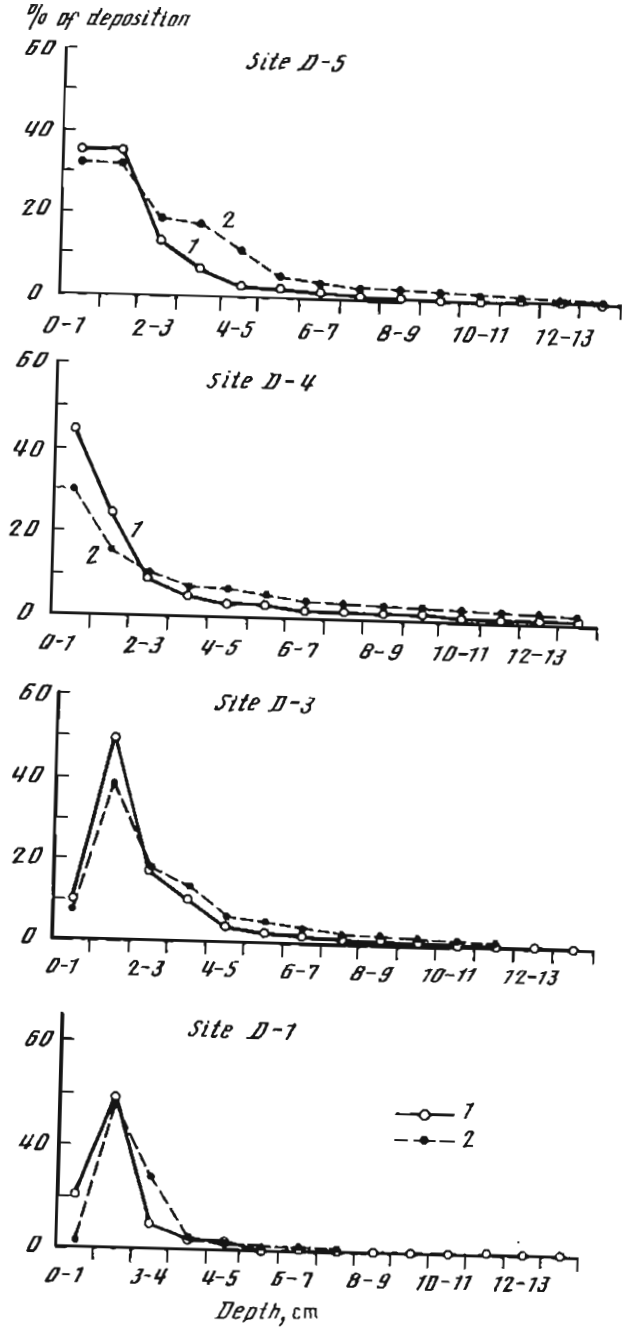

Fig. 64. Vertical distribution of (1) ^{137}Cs and (2) ^{90}Sr in the forest soils depending on biogeocenosis (% of deposition, 30 km exclusion zone, geochemical profile D-1–5)

may be supposed, however, that as soon as a considerable portion of ^{90}Sr be replaced to deeper soil horizons, the rate of its vertical migration (up to the water table) may increase dramatically, since deeper soil layers are practically devoid of any geochemical barrier for ^{90}Sr.

Thus, both ^{90}Sr and ^{137}Cs are rather similarly distributed down the profile of the investigated forest soils. Their migration depends primarily on the same processes, namely, diffusion and the biogenic transport. At the same time, migration of ^{90}Sr is more dependable on the mass transport with intrasoil water compared to ^{137}Cs. The rate of ^{90}Sr vertical migration increases away from ChNPP. The highest intensity of its migration is characteristic of leached chernozems where the vertical distribution of ^{90}Sr is similar to that of mineral nutrients and humus. Other conditions being equal, a higher intensity of ^{90}Sr migration is characteristic of pine forests, hydromorphic areas, and old-tilled areas.

5. CHEMICAL FORMS OF RADIONUCLIDES AND THEIR MOBILITY IN FOREST SOILS

As soon as the fallout particles deposit on the soil surface, they become a subject of physical destruction and chemical leaching. The radionuclides incorporated in the particles are released into the soil solution, and interact with soil liquid and solid components, organic matter, and biota. This results in the formation of soluble and insoluble radionuclide-organic compounds, exchangeable absorption of radionuclides by the soil, irreversible fixation of radionuclides within the crystal lattice of soil minerals, etc. It is obvious that both the vertical distribution of radionuclides in the soil profile and the uptake by plants depend, to a great extent, on the chemical forms of radionuclides in the soil.

5.1. PHYSICOCHEMICAL FORMS OF RADIONUCLIDES IN THE SOIL

The persistence of the fallout particles to weathering is one of the principal factors influencing the radionuclide release into the soil environment and initial radionuclide chemical forms at the earliest stage of the accident. The same important at this stage is soil ability for radionuclide absorption (reversible and irreversible).

The Chernobyl-derived fallout is known to be very different from the global fallout [221]. The first has much higher proportion of unexchangeable and unextractable components* [31]. The differences are most pronounced in the near zone (5–10 km from the accidental unit) where about 90% of ^{90}Sr and 75% of ^{137}Cs are represented by the fuel component. The differences between the global and Chernobyl-derived fallouts are smoothed as the distance from ChNPP increases. In the Russian Federation and other CIS countries, except for the exclusion zone, the radionuclide mobility in the initial fallout varied within wide limits [126].

* A common method to identify basic physicochemical groups of radionuclides is sequential treatment of the fallout (or contaminated soil) with several extragents as follows (sample / extragent weight ratio): (1) water-soluble (H_2O, 1:10, 20° C); (2) exchangeable (1M CH_3COONH_4, 1:10, pH = 7, 20° C); (3) associated with carbonate/oxide/hydroxide/organic matter (1M HCl, 1:10, 20° C); (4) unexchangeable (6M HCl, 1:10, 100° C), but potentially available for plant roots, mycelium, etc.; (5) inextractable (residues), practically immobile and unavailable under natural conditions [31].

Table 66. Chemical forms of ^{137}Cs and ^{90}Sr in the soils of the exclusion zone in 1987 [15, 130, 188]

Radionuclide	Extragent			Unextractable residue
	H_2O	CH_3COONH_4	6M HCl	
^{137}Cs	0.3–2.9	2.2–17.7	15.0–65.8	14.2–78.5
^{90}Sr	0.8–3.0	6.7–24.7	27.7–68.6	31.8–54.1

Immediately after the accident, chemical fractionation of the radionuclides depended primarily on the direction and distance from the accidental unit [130]. Some sporadic data suggest that in the first months after the accident, the proportion of water-soluble ^{137}Cs could be up to 10% in the forest litter and reach 22–53% in the upper mineral layers [80]. In this case, the availability of ^{137}Cs in this period was higher than ^{90}Sr. By 1987, however, the content of water-soluble ^{137}Cs in the soil samples decreased to 0.1–1% and was somewhat lower than the content of soluble ^{90}Sr (Table 66). The most deposition in the soil is represented by unexchangeable and unextractable (residue) forms. The radionuclide mobility at this stage may be ranked as follows: $^{144}Ce = {}^{106}Ru << {}^{137}Cs < {}^{90}Sr$. In general, the proportion of mobile (water-soluble + exchangeable) fractions of ^{137}Cs and ^{90}Sr did not exceed 25% [22, 94, 129, 182]. The share of unextractable residue decreases away from the accidental unit, whereas the share of mobile and potentially mobile radionuclides increases. Intensive radionuclide mobilisation from the fallout particles enlarges the proportion of mobile and exchangeable forms, particularly of ^{90}Sr (Fig. 65). This process runs most intensively during the first five years after the fallout, followed by stabilization of the mobile ^{90}Sr at the level of 50–60% of its total deposition in the soil [129, 130].

Five years after the accident, the proportion of exchangeable ^{90}Sr in various soils reaches 50–70%, which exceeds the corresponding value for ^{137}Cs by a factor of 10–15 (Table 67). A higher proportion of the unextractable residue of ^{137}Cs suggests that its considerable proportion is irreversibly fixed by clay minerals or still contained in the fallout particles. The latter is confirmed by a higher content of this fraction of ^{137}Cs in the A0l and A0f horizons of the forest litter practically devoid of clay particles (Table 68).

The absolute content of all radionuclide fractions decreases down the soil profile, however, the relative proportion of mobile (potentially mobile) forms in deeper layers increases. For example, the total content of mobile ^{137}Cs is 3–14% of its deposition in the 0–1 cm layer and 17-27% in 5–10-cm layer [73]. In general, the proportion of mobile ^{137}Cs increases with depth by a factor of 2–6. The corresponding value for ^{90}Sr is somewhat less [153].

As mentioned, in the first 1–2 years after the fallout, the radionuclide availability and mobility in the soil depended primarily on the distance from ChNPP. In the remote zone (Bryansk region, the Russian Federation), the share of available ^{137}Cs and ^{90}Sr in 1986–1988 exceeded the corresponding values in the near

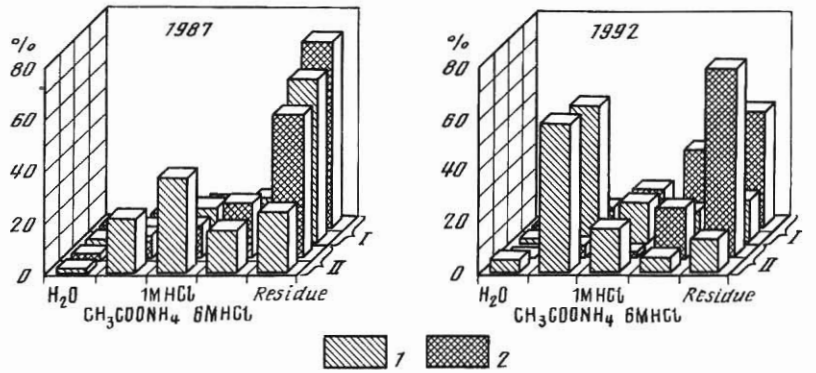

Fig. 65. Forms of ^{90}Sr (a) and ^{137}Cs (b) in the investigated automorphic soils: (1) 30 km exclusion zone; (2) "remote zone" (Bryansk region, Russian Federation)

zone by a factor of 5–10 (see Fig. 65). The differences smoothed in time because of a faster increase in the proportion of exchangeable ^{90}Sr in the exclusion zone [43, 130, 168]. In the ensuing years, the type of soil and ecosystem became more influential on the radionuclide mobility. There is a clear tendency to an increase in the proportion of both the soluble and exchangeable fractions in the mixed forests [135] and a fast decrease of these fractions in all forest types on hydromorphic soils [11]. The latter seems to be contradictory to the main features of a long-term dynamics of TF to the vegetation (see Chapter 3.1). A possible explanation is that the formal classification of the radionuclides by the form of mobility does not necessarily correspond to their actual biological availability. The latter is particularly probable in the near zone to be known enriched with the fuel component that serves as a source of continuous radionuclide release into the soil [92].

Table 67. Average content of ^{137}Cs and ^{90}Sr chemical forms in forest soils (0–20 cm layer, % of total deposition, 1992)

Radionuclide	Fraction of solubility		
	1N CH_3COONH_4	6M HCl	Unextractable residue
Mixed broad-leaved-pine forest (key site D-1)			
^{137}Cs	15.8	35.5	48.7
^{90}Sr	45.4	29.2	25.4
Pine forest (key site K-2)			
^{137}Cs	3.9	54.6	42.5
^{90}Sr	64.8	25.3	9.9

Table 68. Vertical distribution of ^{137}Cs chemical forms in the soils (the exclusion zone, 1992 [135])

Depth, cm	1N CH_3COONH_4		6M HCl		Unextractable residue	
	1*	2	1	2	1	2
Mixed, broad-leaved-pine forest (key site D-1)						
A0l + A0f	0.2	3.0	0.4	50.0	0.4	47.0
A0h	11.0	7.4	22.0	47.9	26.6	44.7
0–1	1.2	8.6	5.1	31.3	10.0	60.1
1–2	0.3	18.4	1.1	31.4	2.1	50.2
2–20	3.1	16.3	6.9	35.2	9.7	49.2
Pine forest (key site K-2)						
A0l + A0f	0.4	2.8	2.9	52.7	1.8	44.5
A0h	2.4	5.6	43.7	61.1	36.8	33.3
0-1	0.4	12.5	14.4	62.5	2.4	25.0
1-2	2.2	15.6	1.0	50.0	0.4	34.4
2-20	0.5	13.9	1.6	50.6	11.0	36.5

*1 – % of total ^{137}Cs content in the soil profile; 2 – % of total ^{137}Cs content in each layer

Radionuclide distribution by various chemical fractions is different from the distribution of the corresponding isotopic carriers (Fig. 66). For example, the proportions of the exchangeable and unexchangeable ^{90}Sr are lower and higher, respectively, compared to the corresponding values characteristic of stable strontium. The forms of stable strontium may be ranked by its relative content as follows: exchangeable > water-soluble > acid-soluble-1 (1M HCl) > acid-soluble-2 (6M HCl). The corresponding range for ^{90}Sr is different: exchangeable > acid-soluble 2 (6M HCl) > acid-soluble 1 (1M HCl) > water soluble. For ^{137}Cs, the proportion of exchangeable fractions is higher and the proportions of acid-soluble and mobile fractions are lower than these of stable caesium isotope .

Thus, 6 years after the accident the radionuclide distribution among the chemical fractions of different mobility in the soils still differs from that of their isotopic carriers. The above-discussed differences between the radionuclide and stable isotope fate suggest that an additional amount of ^{90}Sr may be mobilised and more ^{137}Cs may be irreversibly adsorbed by the soil in the near decade [130].

The proportion of water-soluble and exchangeable (mobile) forms of radionuclides in upper organic horizons (A0 or Ad) is, as a rule, much less than the corresponding value in the underlying organomineral layers (0–5 cm) (Table 69). A low relative content of mobile radionuclides in the upper soil layers is due to a higher proportion of insoluble fallout particles in these layers [141]. As to the deeper layers, their radionuclide composition is apparently constituted by

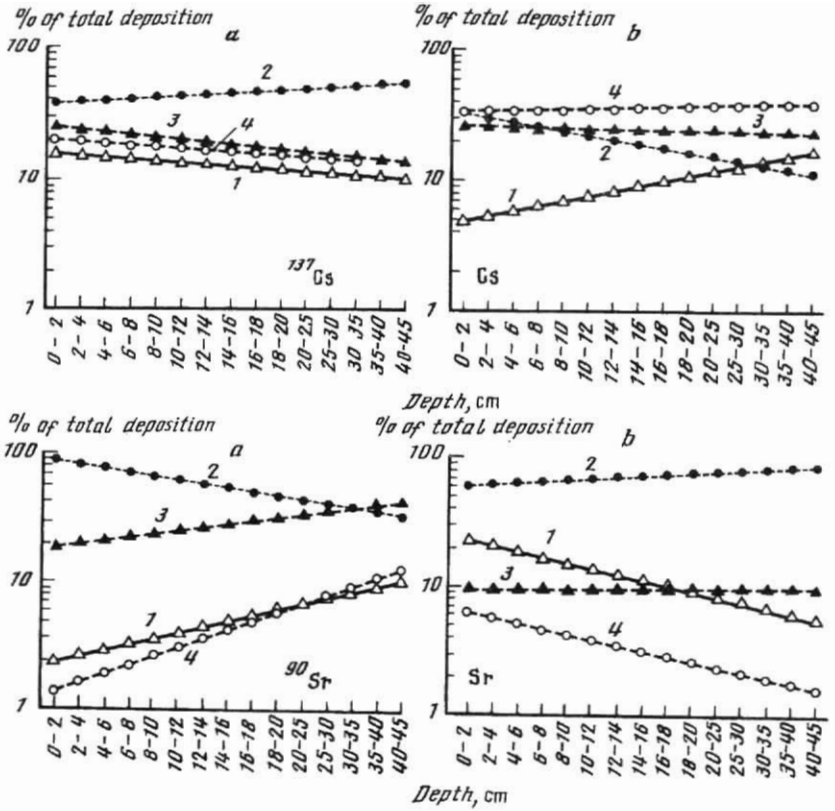

Fig. 66. Exponential approximation of vertical distribution of (a) radioactive and (b) stable isotopes of cesium and strontium in the peat soils (Bryansk Region, Russian Federation, [168]):

(1) H_2O; (2) 1M CH_3COONH_4; (3) 1M HCl; (4) 6M Hcl

the mobile forms arrived from the upper layer. Even there, however, the share of mobile ^{137}Cs and ^{134}Cs varies from 1.3 to 14% of caesium inventory, while the rest 86–99% of radiocaesium is either irreversibly adsorbed by the soil or entered the layer in an insoluble form (*lessivage*, biogenic replacement, etc.).

The proportion of exchangeable radionuclides in the A0 horizon and 5–10 cm layer considerably exceeds the proportion of water-soluble fraction, and radionuclide composition of the water and acetate extracts is completely different. The water extract reflects a wide spectrum of the radionuclides contained in the soil. The acetate extract is almost completely represented by caesium isotopes. The same picture is characteristic of all samples irrespectively of the absolute radionuclide content. The absence of any other radionuclide but caesium in the acetate extract (i.e., among the exchangeable forms) is apparently due to the peculiarities of the "soil–radionuclide" interaction. The so-called "fuel-attributed" radionuclides, such as ^{144}Ce and ^{106}Ru, are likely to be extract-

Table 69. Radionuclides in the acetate and water extracts (% of total content in the sample)

Sample number and depth*	Water extract					Acetate extract			
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{125}Sb	^{134}Cs	^{137}Cs	^{106}Ru	^{125}Sb
Mixed, broad-leaved/pine forest (D-1)									
(1) 0–5	bd**	bd	0.68	bd	bd	1.84	2.12	bd	bd
(2) 5–10	bd	bd	1.25	bd	bd	6.33	7.00	bd	bd
Pine forest with mixed, herb/moss cover (K-2)									
(3) 0–5	0.27	0.07	0.09	0.49	bd	1.11	1.16	bd	bd
(4) 5–10	bd	2.26	2.02	bd	bd	6.82	7.56	bd	bd
Floodplain boggy meadow (K-4)									
(5) 0–5	bd	0.07	0.08	bd	bd	0.26	0.36	bd	bd
(6) 5–10	bd	0.20	0.22	bd	bd	1.21	0.98	bd	bd
Sphagnum bog with grass cover (K-7)									
(7) 0–5	bd	0.11	0.13	0.57	4.69	4.28	4.47	0.47	bd
(8) 5–10	bd	0.53	0.61	bd	bd	3.86	4.18	bd	bd
Mixed, broad-leaved-pine forest (Sh-1)									
(9) 0–5	0.16	0.04	0.05	0.12	0.81	1.70	1.85	bd	bd
(10) 5–10	bd	0.07	0.44	bd	bd	6.49	6.64	bd	bd
Pine plantation (Sh-2)									
(11) 0–5	0.07	0.07	0.07	0.14	0.82	2.17	2.23	bd	0.56
(12) 5–10	bd	0.54	0.64	bd	bd	13.15	14.78	bd	bd

* (1)....(12) – individual number of the sample (see the text): 0–5 cm layer is represented by forest litter in D-1, K-2, Sh-1, and Sh-2, sphagnum residues in K-7; and root mat in K-4; 5–10 cm layer is represented by mineral soil in D-1, K-2, K-4, Sh-1, and Sh-2, and peat layer in K-7

** Below detectable

ed directly from the fallout particles rather than the soil adsorption complex (SAC). They are either irreversibly bound to the soil humus, or leave the soil layer of 5–10 cm with the infiltration flow. The latter is confirmed by the chemical properties of both cerium and ruthenium: their mobile forms may exist under natural conditions only as anions or/and radionuclide-organic compounds, both are not readily adsorbed by soil [1, 132, 133]. Lysimetric studies carried out on the same objects revealed preferable sorption of ^{144}Ce in the podzolic horizon (10–20 cm), which indicates prevalent migration of ^{106}Ru with the infiltration flow compared to other radionuclides [236].

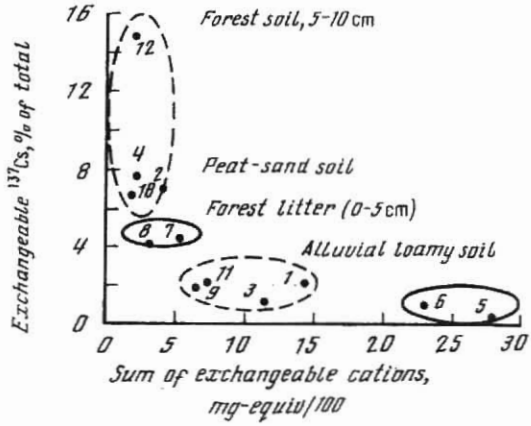

Fig. 67. Exchangeable ^{137}Cs versus the total exchangeable cations in the soil samples (see Table 67)

An inverse correlation was revealed between the relative content of exchangeable radiocaesium and a sum of other exchangeable cations in the soil (Fig. 67). It is of particular interest that such a regularity is characteristic of all investigated soils and does not depend on horizon, organic matter content, or soil type. A possible explanation is prompted by the distribution of the sample types given in Fig. 68. The even-numbered points (2–12) corresponding to the mineral layers are combined more to the left and upper sides of the figure in comparison with organic ones represented by the odd numbers (1–11). The points corresponding to the loamy soil (5, 6) are located to the far right. Sandy-peat soil (points 7 and 8) occupies positions between the mineral soil layers and forest litter. The differences between the relative content of exchangeable ^{137}Cs in the upper and lower layers have been considered above. The lowest content of exchangeable ^{137}Cs in both horizons of the loamy soil is explained by the well-known ability of clay minerals for strong fixation of caesium ions.

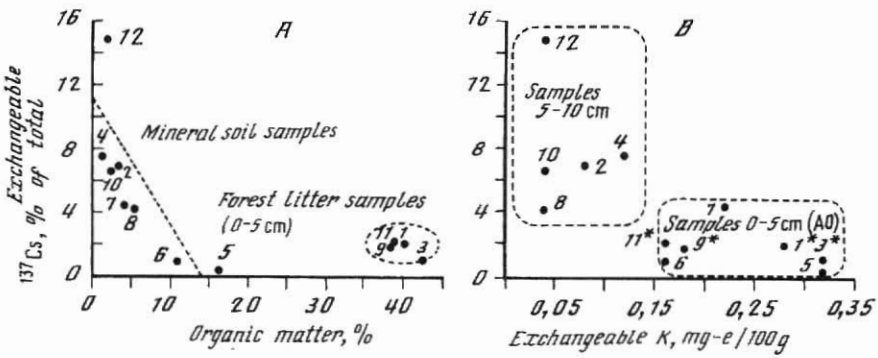

Fig. 68. Exchangeable ^{137}Cs versus (A) organic matter content and (B) exchangeable potassium (^{39}K) in the investigated soil samples (see Table 67)

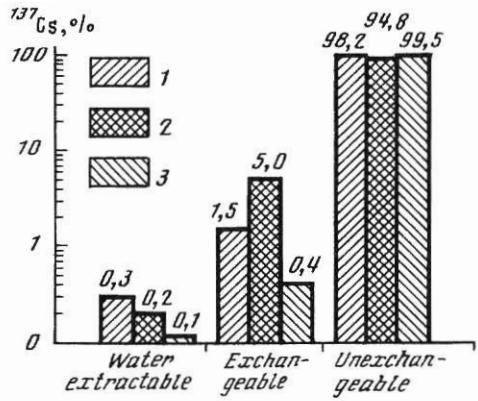

Fig. 69. Chemical forms of ^{137}Cs in the 0-10 cm layer of various soils (1993):

(1) podzolic, sandy soil; (2) peat-gley soil; (3) alluvial-meadow soil

Those components of SAC capable of irreversible caesium absorption have a more manifested effect on its mobility [129]. Our data suggest that the minimum relative content of exchangeable ^{134}Cs and ^{137}Cs should be characteristic of clayey alluvial soils, and the corresponding maximum is attributed to mineral horizons of podzolic sandy soils.

A share of the total mobile caesium is inversely proportional to both the organic matter and the total exchange cation content (Fig. 68). The organic matter content is known to be in line with soil adsorption capacity, and the organic matter may, therefore, cause no direct effect on caesium mobility in the soil. Nevertheless, the organic matter content and composition are both influential on the soil microbiota that, in turn, may effectively immobilise casium by its accumulation in living cells [355].

There is no correlation between the exchangeable ^{137}Cs and the potassium content in the same samples (Fig 68). This phenomenon indicates the differences between radiocaesium and its stable non-isotopic carrier (potassium). This may be explained by (1) differences in the chemical nature of both elements, and (2) age-long equilibrium of potassium (and not ^{137}Cs) in the solid-liquid soil system. The latter is confirmed by a practically stable infiltration flow of potassium coupled with a progressive absorption of ^{137}Cs from the infiltration flow (see Chapter 6.3) [114].

The total reserves of the available radiocaesium (water-soluble plus exchangeable) in the investigated forest soils vary from 0.3 to 5% of the deposition (Fig. 69). The reserves are minimal in the meadow alluvial soil, which is apparently due to a high content of clay minerals. The highest proportion of mobile radionuclides (particularly $^{134,137}Cs$) is characteristic of hydromorphic peat soils, which reflects a low clay content and a high concentration of low-molecular soluble organic compounds [2–6]. The same general regularity is observed for potentially available radiocaesium as well (water-soluble plus exchangeable plus HCl-extractable) [170, 171].

Thus, the content of mobile radionuclides in the 0–5 cm layer amounts to 0.4–4% of the radionuclide inventory in this layer depending on the radionuclide and the soil properties. The proportion of exchangeable caesium in the soil exceeds the proportion of water-soluble caesium by a factor of 3–5.

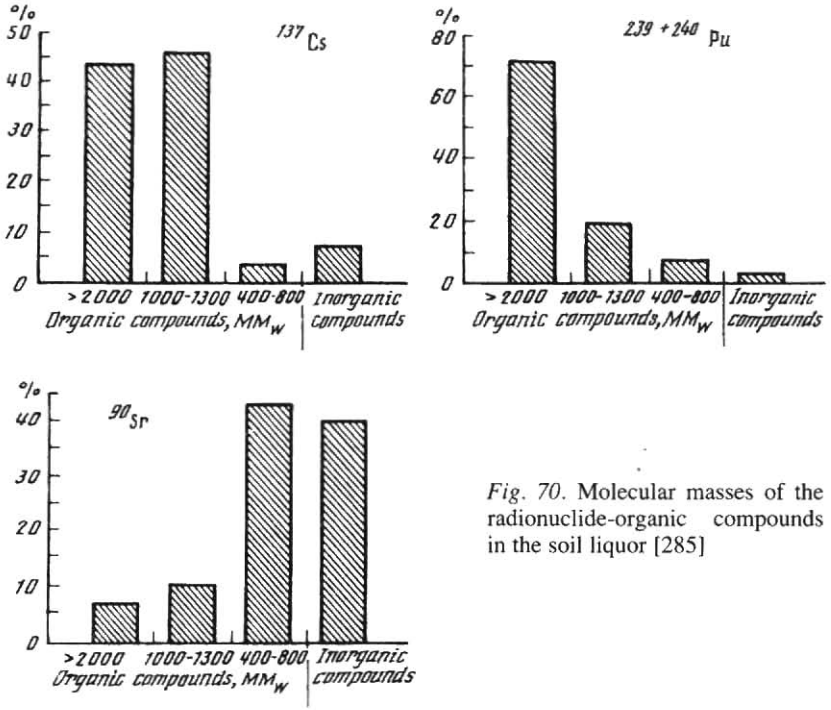

Fig. 70. Molecular masses of the radionuclide-organic compounds in the soil liquor [285]

Radiocaesium is practically the only exchangeable gamma-emitting radionuclide, whereas the water extract is represented by other radionuclides as well. A share of exchangeable radiocaesium is inversely proportional to the organic matter content.

The quantitative and qualitative radionuclide composition of the soil liquid phase varies down the soil profile depending on the chemical nature of radionuclide and the type of ecosystem. The radionuclide content in the soil solution decreases in the range as follows: $^{90}Sr > ^{137}Cs > ^{239,240}Pu$ (^{238}Pu) > ^{241}Am [2–6]. Radionuclides present in the soil solution mostly as radionuclide-organic compounds. $^{239,240}Pu$ (^{238}Pu) and ^{241}Am are associated primarily with high-molecular fractions (MMw > 2000). Highest proportion of ^{137}Cs is associated with medium- and high-molecular organic compounds (MMw over 1000). ^{90}Sr is associated with the low-molecular organic and inorganic compounds (MMw = 350–500) (Fig. 70) [285].

5.2. EXPERIMENTAL SIMULATION OF RADIONUCLIDE MIGRATION USING SOIL COLUMNS

The general parameters of radionuclide migration in the "soil–plant" system depend on the ecosystem type and various environmental factors, which is crucial to assess and forecast the dynamics of the radionuclide content in the ecosystem components. Soil is a complex, multi-component system, and radionuclide migration in the soil is determined by several simultaneous

processes, such as diffusion of free and exchangeable ions, infiltration of radionuclides in the form of ions, colloids, and fine solid particles (*lessivage*), radionuclide upward and downward transport by plant roots, soil fauna and anthropogenic activities, etc.

^{137}Cs migration in the soil "solid–liquid" system. Two basic coefficients, the coefficient of selection (*Ks**) and the coefficient of distribution (*Kd***), are used normally to assess the migration ability of radionuclides in the soil [166]. The first characterises the ionexchange processes (radionuclide behaviour in the surface area of soil colloids), and the second reflects the total distribution of the radionuclides among the solid and liquid phases independently on the type of the radionuclide association with the solid phase. In other words, *Kd* reflects both exchangeable and irreversible adsorption (fixation) of the radionuclides by the soil solid matter. Therefore, both coefficients characterize thermodynamics of the radionuclide migration in the soil column.

In our study, both *Kd* and *Ks* were determined in the simulative experiments using the columns of soddy podzolic forest soils sampled in the exclusion zone [198]. The corresponding data are shown in Table 70. The coefficients values vary depending on moisture and temperature as follows: a) increase in temperature at a stable moisture promotes the soil selectivity for Ca and Cs; b) increase in moisture at a stable temperature decreases the soil selectivity for Ca and Cs. Thus, a higher *Ks* for Cs/K pairs is observed at a lower temperature, and release of exchangeable adsorbed ^{137}Cs to the soil solution is maximum under conditions of a low temperature and a high moisture. The similar tendency is observed at the increased soil moisture. At the same time, Kd^{137}Cs increases as the temperature decreases and moisture increases, which suggests an increased unexchangeable absorption of ^{137}Cs by clay minerals.

Kinetic of exchangeable and unexchangeable sorption of ^{137}Cs by sandy soils. Experiments on the sorption kinetics under static conditions were conducted using columns of non-contaminated soil. The undisturbed columns were treated under various temperature and moisture conditions by the solutions extracted from the contaminated forest litter sampled in the exclusion zone [198]. The soil was incubated during various time intervals (from 10 seconds to 1 month), followed by separation of the solid and liquid matter by vacuum ultrafiltration. The soil residues in the filter were immediately treated by 0.1 M solution of NH_4COOH to separate exchangeable radionuclides. The content of irreversibly adsorbed radionuclides in the sample was calculated from the difference between the specific activity of the residues after the acetate treatment.

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* Coefficient of selection is calculated by the following equation: $Ks = [(M_1)Am_2] / [(M_2)Am_1]$, where M_1, M_2 are the cation (radionuclide) content in the soil solid phase, and Am_1, Am_2 are the cation (radionuclide) activity in the liquid phase.

** Coefficient of distribution is calculated by the following equation: $Kd = [SR]/[R]^+_w$, where [SR] is the equilibrium content of the cation (radionuclide) absorbed in the soil solid phase and $[R]^+_w$ is the equilibrium cation (radionuclide) concentration in the liquid phase.

Table 70. Coefficients of ^{137}Cs selectivity (K_s) and distribution (K_d) in the peat-podzolic soils (0–15cm layer)

Experimental conditions (T°C/W, %)	K_s for various ion pairs						K_d
	Mg/Ca	K/Ca	Cs/Ca	K/Mg	Cs/Mg	Cs/K	^{137}Cs
25/7	0.88	1.52	0.64	1.72	0.72	0.42	2.84
5/7	0.41	0.39	0.72	0.96	1.77	1.84	4.95
5/14	0.65	0.64	0.74	0.98	1.13	1.15	14.96
25/14	0.32	0.34	0.46	1.09	1.46	1.34	5.35

Under conditions of stable moisture and full field capacity (30 weight percent), the exchangeable sorption gets completed in 6–10 min (Fig. 71). Since that moment, i.e. on completion of ^{137}Cs transfer to the solid phase, the radionuclide equilibrium is shifting toward the irreversibly adsorbed (fixed) forms. The proportion of fixed forms at the initial moment is about 20% of the total radionuclide content, and increases up to 90% after 24 hours of the incubation. Under the given conditions, the irreversible adsorption requires several days to be completed. A higher moisture considerably slows down the sorption: up to 5% of ^{137}Cs ions still present in the soil solution for several months after the accident (Fig. 72). Unexchangeable absorption of ^{137}Cs is also limited by 50–60% of its total content in the soil. The proportion of exchangeable ^{137}Cs is stabilised at 40–50% of its total content, with some tendency to decrease. It is obvious that, like in the case of low moisture, ^{137}Cs participates in various redistribution processes: (i) between the soil solution and diffusive layers of the soil colloids (exchangeable sorption), and (ii) between the diffusive colloid layers and specific sites in the crystal lattice of the soil minerals (fixation). Under high moisture, both processes, particularly (ii), are not intensive because of thick water films preventing fast migration of ^{137}Cs cation through the diffusive colloid layers or the crystal lattice. Thus, the sorption rate of ^{137}Cs by soil in this case is determined primarily by the rate of the diffusion process, which is confirmed by a weak effect of temperature.

After incubation of the soil columns during 24 hours under 30% moisture and +5 °C temperature, the first portions of the filtrates leaving the soil columns were practically free of radionuclides. The data of static experiments show that under the given conditions, 24 hours are the time sufficient to reach equilibrium for both exchange processes and radionuclide fixation. The consecutive portions of the output solutions contain the maximum concentration of radiocaesium, followed by its decrease. It means that the time interval of 10–15 min, that is necessary for the first portion of the solution extracted from the forest litter to pass through the column, is insufficient for the radionuclide absorption in the mineral soil layers. Freezing of the wet soil column reduces the radionuclide absorption drastically, which results in a sharp increase in the ^{137}Cs concentration in the filtrate leaving the soil column immediately after defrosting (Fig. 72).

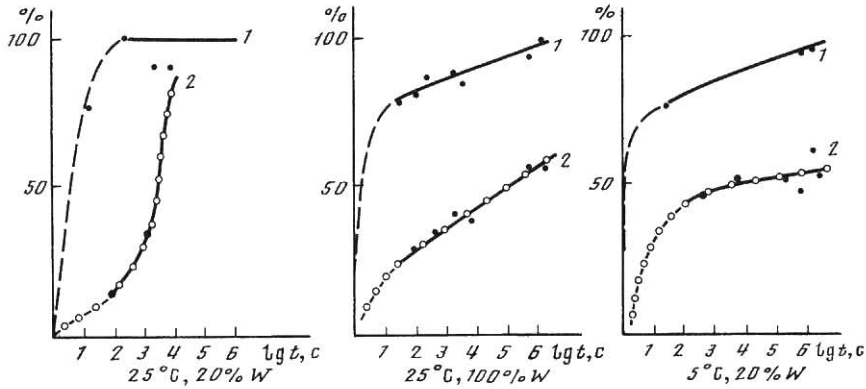

Fig. 71. Proportions of (1) reversibly and (2) irreversibly absorbed radionuclides in the soil samples depending on duration of the soil-water contact

Thus, radionuclide distribution in the "soil–solution" system under conditions of an intensive water transport is controlled primarily by the cation exchange processes. The "solid-liquid" cation ratio depends in this case on the corresponding *Ks* values. On the contrary, when the water flow is weak or absent, ^{137}Cs fixation is not limited by the contact time and may affect the radionuclide distribution, which in this case is characterised by *Kd*.

Estimation of the coefficient of diffusion of ^{137}Cs. It is the practice to express the rate of ion-exchange processes running in the soils by the so-called coefficient of diffusion (D, cm^2/s). Earlier studies show that diffusion is the main factor of the radionuclide distribution in the soil [131, 258]. A more detailed research has revealed that when the concentration of the adsorbed ions is below 0.01 g-equiv/l (which is the case), the rate of ion exchange is limited by the so-called external (film) diffusion [24]. The same was shown to be true for various pairs of ions [334, 346]. There are numerous data, however, which indicate the so-called "inner diffusion" mechanism when the absorption depends on a direct cation interaction with the soil solid phase including the crystal lattice of clay minerals [257]. Therefore, the kinetics of radionuclide (particularly ^{137}Cs) sorption includes two basic processes: (i) ion exchange in itself limited by the external diffusion, and (ii) irreversible adsorption (fixation) limited by a direct radionuclide interaction with the solid matter. Diffusion is of prime importance for the processes of radionuclide migration after the Chernobyl accident: 60 to 99% of ^{137}Cs is believed to be distributed in the soil due to diffusion [138, 172], which is somewhat lower than the corresponding average values for "weapon" caesium of global fallout (Table 71).

This index varies in a wide range depending on the soil properties, moisture regime, particle-size and mineralogical composition, and time. The highest values of D are registered in hydromorphic and semi-hydromorphic soils. In all cases, D decreases in time. A set of special experiments has been conducted to

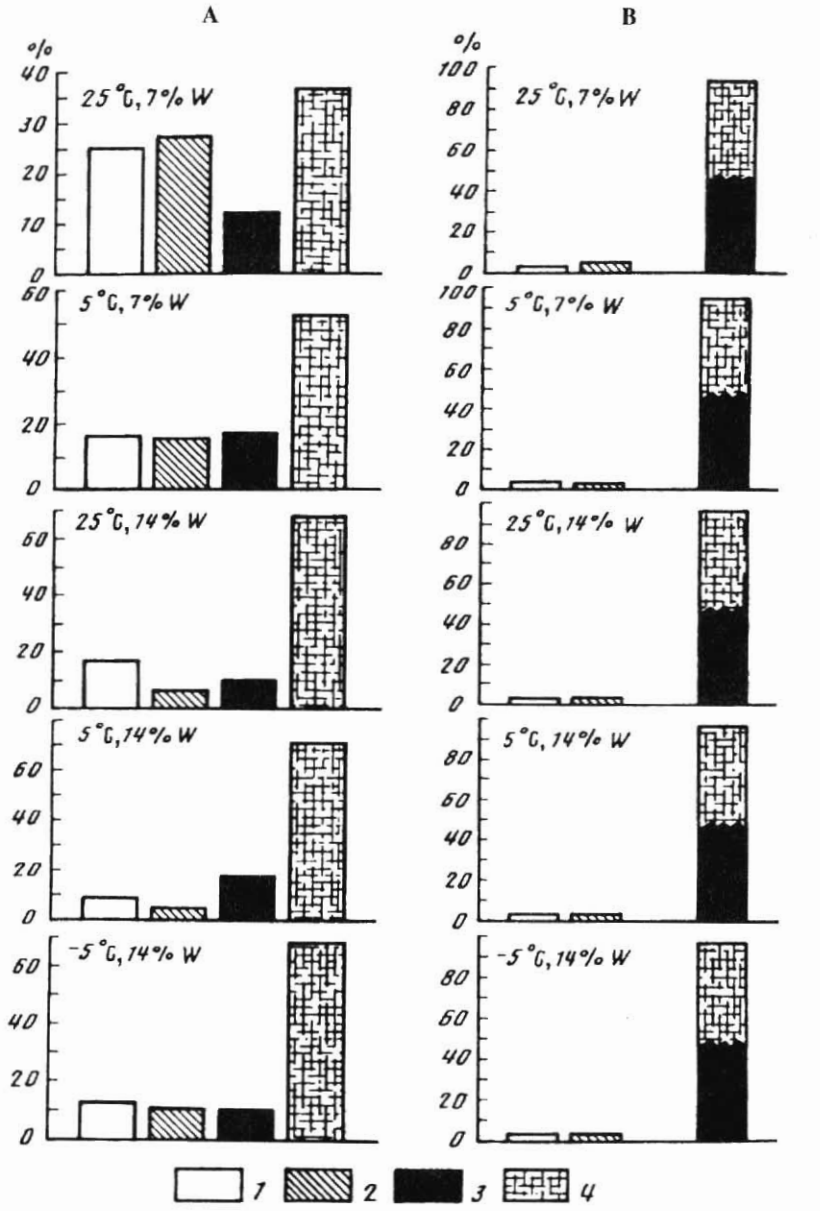

Fig. 72. Radionuclide forms in the soil columns under various hydrothermal regimes: (A) one months of incubation, (B) 10 months of incubation:

(1) water extractable; (2) exchangeable; (3) unexchangeable-1, bound with the soil mineral component, (4) unexchangeable-2, bound with the soil organic component (extractable by hot H_2O_2)

Table 71. The coefficient of diffusion of ^{137}Cs in various soils

Soil (substrate)	D, cm^2/s	Reference
Podzolic, sandy	1.0E-5	[351]
Hydromorphic sandy podzol	1.0E-7	[351]
Podzolic, sand-loam	(2.0–7.0)E-7	[351]
Podzolic, sand-clayey	5.0E-7	[351]
Podzolic, loamy	2.0E-7	[351]
Humus-gley, loamy	5.0E-7	[351]
Semi-bog, silt-loam	7.0E-7	[351]
Podzolic, clay	(2.0–3.0)E-7	[349]
Sand-loam	1.6E-7	[308]
Carbonate sand-loam	2.0E-7	[308]
Carbonate loam	2.0E-7	[308]
Peat	2.1E-7	[308]
Old-tilled (30 km exclusion zone)	(0.2–4.5)E-8	[28, 30]
Soddy-podzolic, sandy, "near zone" of ChNPP	(2.0–3.3)E-8	[213]
Soddy-podzolic, sand-loam, 30 km exclusion zone ChNPP (Belarus')	2.0E-8	[83]
Soddy-podzolic, sand-loam	(1.4–4,9)E-8	[138]
Peat-bog	(1.2–4.1)E-7	[138]
Soddy-podzolic, virgin (Bryansk region, Russia)	(0.6–5.9)E-8	[275]
Sod, soddy-podzolic, peat-bog (Belarus')	(0.4–10.0)E-8	[180, 183]
Automorphic, 30 km exclusion zone (Ukraine)	(1.1–1.7)E-8	[12, 13]
Hydromorphic, 30 km exclusion zone (Ukraine)	(1,8–2,3)E-8	[12, 13]

determine D in forest soils (calculations were made using the Romer and Shilling equation [340]. D varies down the forest soil profiles as follows [198]:

Substrates	D (cm^2/s)
(a) Forest litter–forest litter	$2 \cdot 10^{-11}$
(b) Forest litter–mineral soil	$1 \cdot 10^{-11}$
(c) Mineral soil–mineral soil	$4.5 \cdot 10^{-8}$

Comparison of the data on D obtained by different authors is difficult because of different experimental approaches and methods of calculation, and it may be done correctly if we compare only soil-to-soil values for the same soil types (Table 71). The values of (c) variant are within the range reported in the literature, but we found no published data on the variants (a) and (b). The data by Oscarson (private communication) suggest that the organic soil horizons have a lower D compared to the mineral layers. It seems to be true, since the pore space of the organic horizons is more developed, and wettability of the soil organic matter in itself is relatively low.

Release of ^{137}Cs from the "hot" particles to soil solution. The most specific feature of the Chernobyl-derived fallout in the exclusion zone is a considerable proportion of the "hot" particles consisting of the reactor core fragments enriched with the nuclear fuel components. Mobility of the radionuclides incorporated in these particles depends on the size and composition of the latter, soil properties and regimes, and nature of each radionuclide [91, 129, 130].

The following experiment has been conducted to estimate the rate of ^{137}Cs release from the "hot" particles to the soil solution. The samples of forest litter taken in the near zone were repeatedly treated with NH_4COOH and distilled water. The ^{137}Cs content in the filtrate became stable (the same as the radionuclide concentration in the water extract) after a triple treatment with ammonium acetate. In total, 2.5 to 4.4% of the initial ^{137}Cs depositions were lost from the treated samples, which indicates that the treated samples were completely depleted of mobile ^{137}Cs. The samples were incubated for 30 days under 25 °C and moisture of 30%, followed by the same extraction procedures. This has resulted in the extraction of additional 0.8% of the total radionuclide deposition. Organic matter is known to adsorb radiocaesium by the exchange mechanism, and some proportion of ^{137}Cs, irreversibly adsorbed by a negligible amount of mineral admixtures in the forest litter, is unextractable by ammonium acetate. Thus, the additional portion of ^{137}Cs was extracted from the fuel particles apparently due to biochemical processes running in the forest litter.

It may be assumed, as a first approximation, that the rate of ^{137}Cs release from the hot particles is about 0.8% per month (at a temperature of 25 °C and moisture of 30%). Assuming the vegetative period of 4–5 months, an annual rate of ^{137}Cs leaching from the hot particles is about 3.2–4%. From 90 to 95% of caesium released from the particles is retained in the forest litter. Thus, the proportion of mobile (potentially mobile) radionuclides in the soil solution will be directly proportional to the content of the hot particles if the rate of radionuclide leaching from the particles is comparable with the rate of cation dissociation in the "solid-liquid" soil system.

The effect of diffusion, mass-transport, and *lessivage* on ^{137}Cs migration. The integrated effect of diffusion, water transport, and *lessivage* on ^{137}Cs is shown in simulative column experiments. In the first set of experiments, the radionuclide migration was simulated by passing water extracts from the forest litter through the columns of uncontaminated soil (12 cm in length) under various conditions corresponding to the environmental conditions of the region. Three seasons were simulated in the experiments: (i) spring (autumn) period with high temperature and rainfall of 370 mm, (ii) winter period with negative temperature and zero water movement (frozen), and (iii) early spring period with low temperature and rainfall of 230 mm. Three basic time intervals were simulated as the conventional one-year, three-year, and ten-year periods.

During the one-year simulated period, about 0.83% of ^{137}Cs was transported from the forest litter to mineral soil layers. Only 0.03% of the radionuclide has left the column, whereas the largest proportion of mobile ^{137}Cs was absorbed by the soil (Table 72).

Table 72. Vertical distribution of ^{137}Cs in the experimental soil column

Layer, cm	Diffusion, 3 months*		Filtration, 3 years		Diffusion + filtration, 3 years		Filtration, 10 years	
	1	2	1	2	1	2	1	2
Forest litter	99.14		98.94		99.12		98.9	
0–0.5	0.76	88.32	0.67	63.4	0.64	74.74	0.82	72.85
0.5–1	0.07	8.27	0.12	11.11	0.08	8.44	0.09	8.81
1.0–2	0.02	1.79	0.08	8.04	0.05	4.08	0.04	5.54
2.0–3	0	0.08	0.07	6.59	0.01	1.55	0.02	1.6
3.0–5	0.01	1.35	0.03	2.76	0.01	0.98	0.01	1.12
5.0–7	0	0	0.01	0.98	0.01	0.4	0	0.89
7.0–12.0	0	0.22	0.04	3.45	0.02	4.42	0.05	2.02
Filtrate			0.04	3.66	0.06	5.39	0.06	7.17
Total loss from the entire forest litter	0.86		1.06		0.88		1.1	

* 1 - % of total deposition; 2 - % of the loss from the forest litter

For the three-year simulated period, the forest litter has lost 2.33% of ^{137}Cs inventory, and 0.06% of the radionuclide left the column. A reliable increase in the radionuclide content was registered in the upper 2 cm layer of the mineral soil.

For the ten-year period, the radionuclide content increased in deeper layers of the column, although the main proportion of ^{137}Cs was retained by the upper 2 cm mineral layer.

During the first year, the upper 0.5 cm mineral layer of the column absorbed about 65% of the total ^{137}Cs lost from the forest litter. After the three-year simulation, this value increased up to 85%. The intensive accumulation may be caused not only by the migration of leached (mobile) radionuclides, but a direct transport of solid particles as well (*lessivage*).

Freezing of the moistened soil columns reduces the radionuclide absorption. Defrosting results in a dramatic increase in ^{137}Cs concentration in the column drainage (filtrate). Cold filtrate leaving the column immediately after defrosting contains the same amount of radionuclides as the double volume of the warm filtrate leaving the column under normal conditions.

Thus, in soddy-podzolic soils mobility of ^{137}Cs increases in response to temporal hydromorphic conditions, such as abundant rains, intensive snow melting, etc. These periods promote fast ^{137}Cs migration into deeper soil layers. The radionuclide portion transported to deep soil layers (potentially to the aquifers) is in general correlation with the filtration rate and depth of water percolation. The data on radionuclide distribution obtained in the course of simulative column experiments are in a good agreement with the corresponding data obtained from the field observations and experiments.

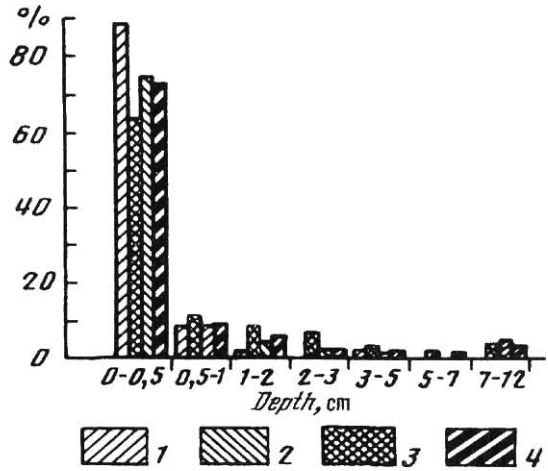

Fig. 73. The effect of various processes on ^{137}Cs distribution in the soil columns (the radionuclide portion replaced from the forest litter is taken as 100%). Simulated processes:

(1) diffusion, 3 months; (2) diffusion + infiltration, 3 years; (3) infiltration, 3 years; (4) infiltration, 10 years

Another set of the column experiment was designed to prevent a possible transport of solid particles down the soil column. For this, a filter paper was placed into the soil columns in such a way to separate the column parts corresponding to the soil horizons. The following conditions and moisture regimes were simulated in the experiment: (i) pure diffusion (no infiltration, 25 °C, 3 months), (ii) infiltration corresponding to the average 3-year summer rainfall (25 °C, 7 days), (iii) regular alternation of stagnant and filtration regimes (3-year summer rainfall for 7 days, followed by 1 month of stagnant regime, 25 °C), and (iv) extreme filtration (10-year summer rainfall for 1 month, 25 °C). The obtained data (Table 70, Fig. 73) prove that diffusion is the main factor of ^{137}Cs migration in the upper soil layer (0–1 cm). All differences in the radiocaesium content in the lower layers were direct in proportion to the infiltration rate. Setting the mechanical barriers to prevent *lessivage* results in a considerable decrease in radionuclide migration from the forest litter to the mineral soil layers. For example, the simulation of 3-year filtration and diffusion results in the accumulation of 2.3% of the total caesium in the upper mineral layer under normal conditions, and only 1% in the case of filter paper inserted between forest litter and mineral layers. The contribution of *lessivage* and other mechanisms of particle migration may be even more manifested under native conditions.

6. BIOGEOCHEMICAL FLUXES OF RADIONUCLIDES IN FOREST LANDSCAPES

Radionuclide migration in the "soil-plant" system is of prime importance for its distribution in terrestrial ecosystems. The contribution of individual components of biogeocenosis to radionuclide migration and accumulation is possible to estimate by analysing individual fluxes in the biological cycle of radionuclides.

6.1. RADIONUCLIDE INFLUX TO THE SOIL SURFACE WITH LITTERFALL

As mentioned, arboreal vegetation was decontaminated from a large proportion of radionuclides as soon as two years after the accident. However, residual contamination of the stand components directly exposed to the fallout (for example, external bark) remained up to the middle 1990s. At the same time, the root uptake has played an increasing role in the stand contamination for 2 or 3 years after the accident, which was expressed in the increased radionuclide content in the inner and annually renewed tree organs (internal bark, wood, leaves, etc.). Thus, there is an additional radionuclide flux from the contaminated stand to the soil surface. The contaminated litterfall serves as a food source for soil fauna (primarily, invertebrates), and incoming radionuclides are readily incorporated in the ecosystem food chains [127, 356]. Forest litter decomposition results in radionuclide mobilisation from the contaminated plant residues and may be redistributed in the "soil-solution" system reutilized by soil biota, etc. The radionuclide flux as a part of litterfall is, therefore, an indispensable component of the forest biogeochemical cycle.

The total weight of annual litterfall in the investigated forests varies from 310 to 560 g/m^2 per year depending on year and ecosystem type (Fig. 74). The litterfall mass is composed primarily by assimilative organs: leaves and needles (Fig. 75). Maximum of this fraction (over 50%) is characteristic of mixed, broad-leaved coniferous forests, and it is about 40% in alder forest. The least weight proportion is contributed by generative organs (cones) and external bark (cork) fragments.

The radionuclide flux depends, however, not only on the weight of the contaminated components, but the radionuclide content in each component as well. In this context, the picture is almost opposite: the radionuclide content is the highest in the external bark and the least in the leaves and nee-

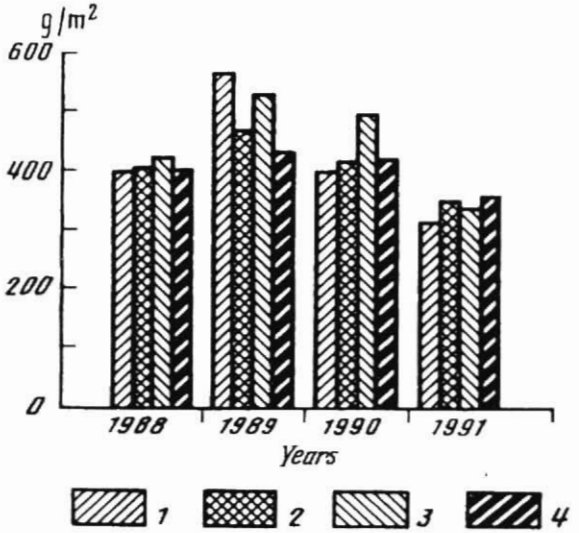

Fig. 74. Multiyear dynamics of the litterfall coming to the soil surface in various key sites (30 km exclusion zone, dry weight):
(1) D-1; (2) D-3; (3) K-2; (4) Sh-1

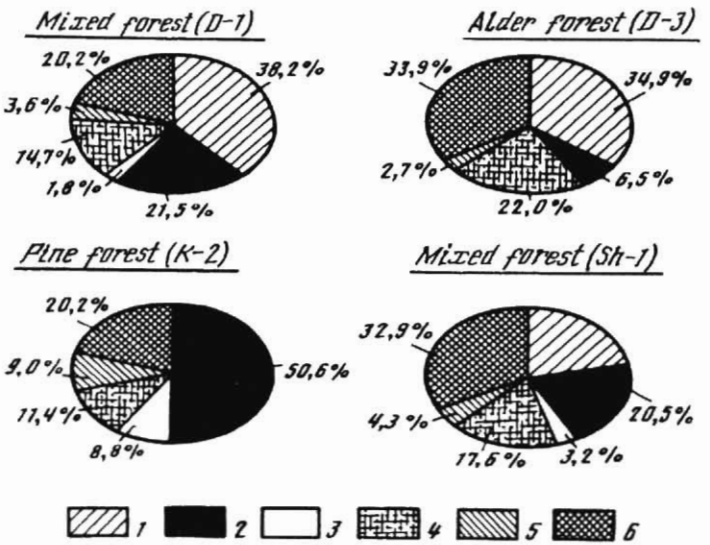

Fig. 75. Fractional composition of the litterfall (% of total, dry weight, weighted-average for 2 years):
(1) leaves; (2) needles; (3) cones; (4) branches; (5) external bark; (6) other

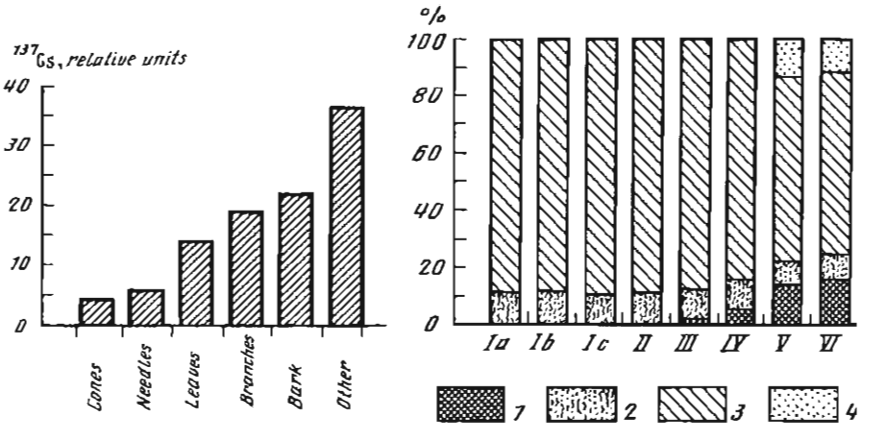

Fig. 76. Relative content of ^{137}Cs in the structural fractions of the litterfall

Fig. 77. Radionuclide composition of various fractions of the litterfall (1990): (I) leaves (a - oak, b - birch, c - aspen); (II) cones; (III) needles; (IV) branches; (V) bark; (VI) other. Radionuclides: (1) ^{144}Ce; (2) ^{134}Cs ; (3) ^{137}Cs; (4) ^{106}Ru

dles (Fig. 76). In other words, in the early 1990s, the most contaminated components were still those directly exposed to the initial fallout or secondary contaminated due to re-suspension processes. This is confirmed by presence of ^{144}Ce and ^{106}Ru in the radionuclide composition of the litterfall (Fig. 77). Since the radionuclide content in the fresh litterfall is by 2–3 orders of magnitude less than in the soil (forest litter), even the smallest admixture of soil in the litterfall may cause a significant change in its radionuclide composition.

The radionuclide content in the litterfall components is, as a rule, low compared to the corresponding alive organs. Manifestation of the difference depends on species: it is minimal for oak leaves and maximal for pine and birch leaves. Although the litterfall contains some proportion of ^{144}Ce and ^{106}Ru, it is enriched with ^{137}Cs compared to the soil upper layer, which is quite explainable by the selective root uptake of the latter (Fig. 78).

An absolute radionuclide content in the litterfall is in proportion with the deposition, whereas the relative concentration (normalised to deposition) is site-specific (Fig. 79). The highest absolute ^{137}Cs concentrations were registered in the "near zone", and the highest relative concentration is characteristic of hydromorphic areas with highest $TF^{137}Cs$ to the vegetation.

Thus, the specific activity and radionuclide composition of fresh litterfall depends on radionuclide deposition in the soil, species composition of the stand, soil-ecological conditions, and weather parameters controlling dust transport in the forest. Seasonal and long-term dynamics of the radionuclide content also depends on all these factors.

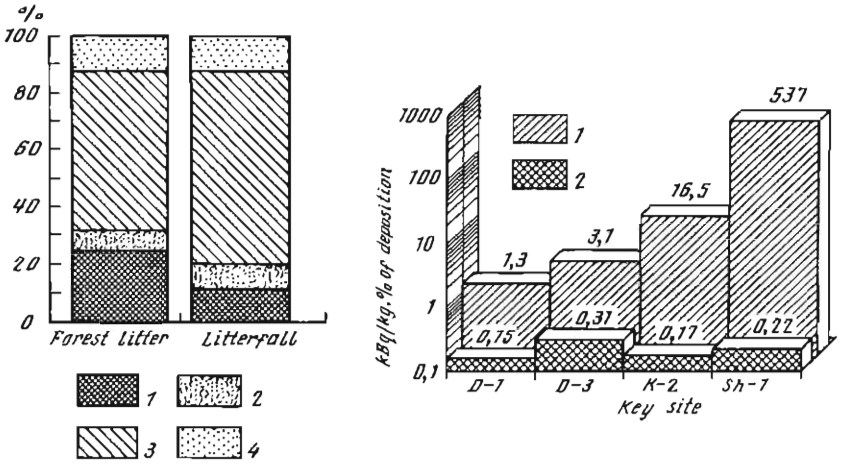

Fig. 78. Average radionuclide composition of the forest litter and fresh litterfall in the exclusion zone (1990):
(1) ^{144}Ce; (2) ^{134}Cs ; (3) ^{137}Cs; (4) ^{106}Ru

Fig. 79. Total radionuclide content of the litterfall (1) and annual radionuclide incoming to the soil (2) in 1990

Seasonal dynamics of the litterfall incoming to the soil surface exhibits (particularly in coniferous cenoses) two maximums (Fig. 80). ^{137}Cs content in the litterfall is inversely proportional to its mass, which is likely due to a well-known phenomenon of withdrawal of organic and mineral nutrient from the assimilative organs shortly before a massive litterfall [352–354]. ^{137}Cs transport to the soil surface by litterfall depends on both deposition in the site and litterfall fraction (Fig. 81).

Long-term dynamics of ^{137}Cs in the forest cenoses varies depending on the ecosystem type (Table 73, Fig. 82). The radionuclide content in the litterfall of

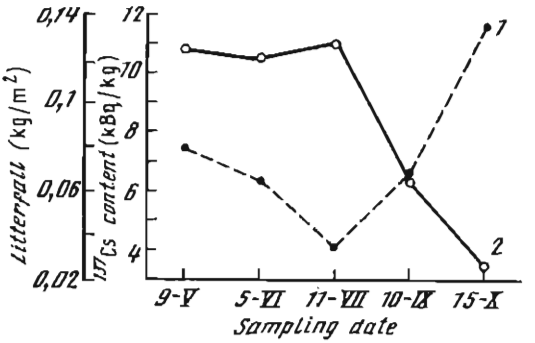

Fig. 80. Seasonal dynamics of (1) litterfall incoming and (2) ^{137}Cs content in the litterfall (1994, pine forest, key site K-2)

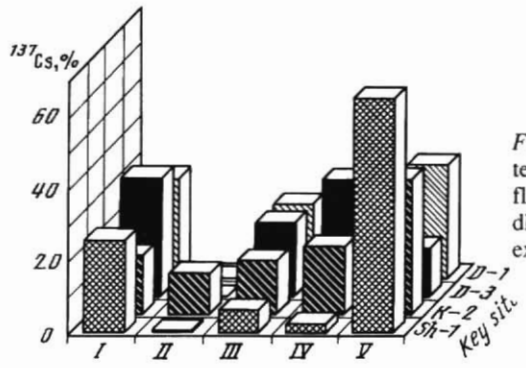

Fig. 81. Contribution of various litterfall fractions to ^{137}Cs annual flux to the soil: (I) leaves and needles; (II) cones; (III) branches; (IV) external bark (cork); (V) other

all ecosystems decreased gradually for the first 3–4 years after the accident. Later on, the decrease continues in the mixed and pine ecosystems of the "remote zone" and automorphic areas. In the "near zone" and hydromorphic areas, the radionuclide content in the litterfall tends to increase in line with $TF^{137}Cs$ to the overstorey.

In general, annual addition of ^{137}Cs with litterfall to the soil surface varies from 1.5 to 6.2% of its inventory in the tree canopy and from 0.2 to 0.7% of its total deposition (soil included). In the first two years after the accident, however, these values were apparently much higher because of intensive natural decontamination processes running in the tree canopy at that time [277]. Thus, litterfall contributes a lot to the biogeochemical cycle of ^{137}Cs in the first years after the accident (due to intensive natural decontamination) and in the subsequent years (due to root uptake). The corresponding radionuclide flux depends on deposition, fallout and soil properties, stand, and weather conditions. The intensity of ^{137}Cs flux is

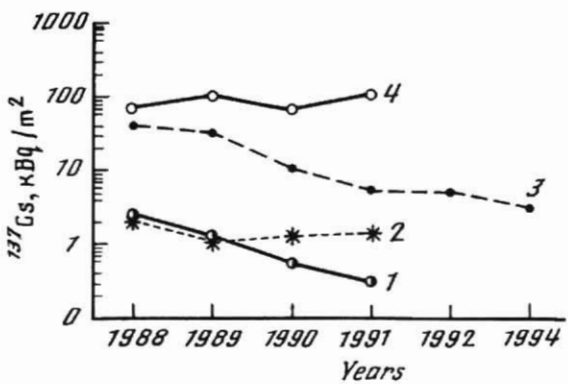

Fig. 82. Long-term dynamics of ^{137}Cs influx to the soil surface with litterfall in various parts of the exclusion zone; key sites:
(1) D-1; (2) D-3; (3) K-2; (4) Sh-1

Table 73. Multiyear dynamics of the ^{137}Cs content in litterfall, (kBq/kg, dry weight, average of n = 5–10)

Year	Key site			
	D-1	D-2	K-2	Sh-4
1988	6.41	5.34	99.2	175.79
1989	2.93	3.1	59.81	221.44
1990	1.37	3.06	21.07	152.96
1991	0.97	3.91	19.59	291.52
1993	nd*	nd	12.18	nd
1994	nd	nd	7.41	nd

* No data

in direct proportion with $TF^{137}Cs$ to arboreal vegetation: it is most intensive in the hydromorphic areas and in the "near zone" of ChNPP.

6.2. RELEASE OF RADIONUCLIDES FROM THE PLANT DEBRIS AND FOREST LITTER

Transformation of plant residues as components of forest litter and radioactive mobilisation within forest litter are the key processes influencing radionuclide migration in forest ecosystems. Firstly, forest litter is the main radionuclide sink in the contaminated forest ecosystems [242, 277]. Secondly, a large portion of soluble organic compounds (SOC) is discharged from the soil organic horizons as a result of transformation and decay of plant residues. SOC are known to promote radionuclide mobilisation from the fallout particles by formation of the so-called radionuclide-organic complexes (ROC) [2–6, 333]. In addition, fresh litterfall in itself contains a significant amount of mobile and readily mobilised radiocaesium.

Direct data suggest that 30 to 70% of radionuclides are released from the fresh litterfall in the autumn-spring months (Table 74). The minimum and maximum rates of radionuclide leaching were observed in pine and mixed forests, respectively. These data are in agreement with the known concepts and mechanisms of organic matter transformation and element leaching in various forest ecosystems [208, 209]. The completeness of leaching is high (about 50%), but ^{106}Ru exhibits somewhat higher mobility compared to other radionuclides, especially in pine cenoses. Leaves lose radionuclides much faster than the pine needles.

In general, radionuclides incoming to the soil with fresh litterfall have a considerably higher mobility compared to the radionuclides incorporated in the fallout particles. In other words, participation of radionuclides in the biological cycle increases their migration potential.

Table 74. Radionuclide leaching from the litterfall components in autumn and winter of 1989-1990 (% of initial content, average of n = 20)

Litterfall component	Radionuclide				
	Sg	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru
Pine needles	38.6	42.4	40.8	32.2	45.1
Small branches	55.8	71.1	59.2	51.8	68.6
External bark	58.2	65.5	57.2	49.3	71.7
Aspen leaves	51.3	63.2	52.5	44.1	61.3

Numerous data suggest that microbial activities (and, respectively, the rate of decomposition of organic matter) in forest soils are most manifested in the forest litter subhorizon A0f, and least manifested in deep mineral horizons (Fig. 83) [61, 63, 338]. The only exclusion is peat-gley hydromorphic soils (site D-3) under alder forest where maximum biological activity is attributed to the topmost subhorizon (A0l). This is due to the most favourable air and moisture regimes in this horizon of peat soils [114]. Comparison of the decomposition of the test material (cellulose) in the soils of various cenoses suggests that the decomposition processes are more intensive in the forest cenoses compared to the meadows, particularly during the vegetative interlude (from the late autumn until the middle spring) (Fig. 83). This is apparently due to "softer" microclimatic conditions under the forest cover compared to the meadow areas exposed to any weather change, severe winter frosting and summer desiccation [33].

In general, decomposition of the test material obeys the same regularities as the decomposition of fresh litterfall. The annual weight loss of the latter varied from 38 to 65% (Fig. 84). The maximum loss was characteristic of the test material in the automorphic areas under mixed forests (site D-1) and the minimum weight loss was typical of hydromorphic areas under alder forest (site D-3). In all cenoses, fresh litterfall decays faster than the transformed material of A0f (60 and 30%, respectively). The maximum rate of decomposition falls on summer and early autumn, which is due to the most favourable moisture-temperature balance and presence of a fresh organic substrate, favourable for microbiological activities (Fig. 85).

Significant weight loss of the litterfall samples during decomposition is not necessarily in correlation with loss of radionuclides. This is true only in the case of a highly contaminated fresh material placed on the "clean" or slightly contaminated sites (Fig. 84-II). In contrast with it, when the experimental litterfall samples have been sampled and established in the same site, practically no loss of ^{137}Cs was registered (Fig. 84-I). This phenomenon is due to much higher (up to two orders of magnitude) radionuclide content in the forest litter layers A0f and A0h compared to A0l and fresh litterfall. Radionuclides leached from the samples in parallel with their upward transport from the deeper layers of the forest litter (A0f and A0h)

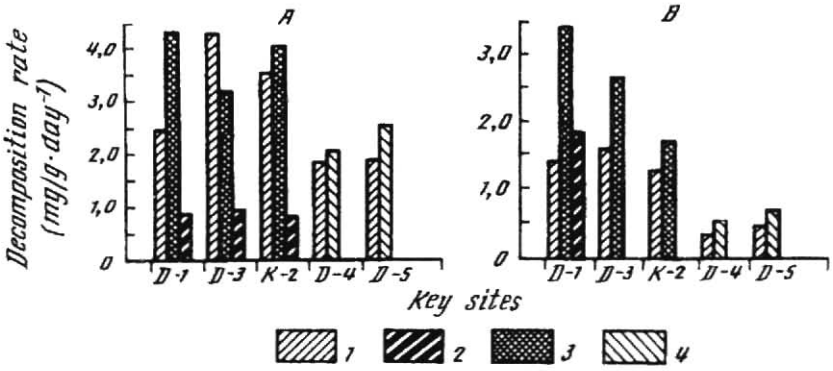

Fig. 83. Decomposition of cellulose in the investigated soils during (A) vegetative and (B) interim period:

(1) on the soil surface; (2) at the depth of 150 cm; (3) within the forest litter layer A0f; (4) within the mineral layer 0-5 cm

to its surface (A0l) by soil fauna (mechanical transport) and/or mycelium (physiological transport) [294]. Thus, radionuclide loss from the most upper layer of the forest litter is often overcompensated by the upward transport from more contaminated layers. The same experiment conducted with more contaminated samples placed on the soil surface at relatively "clean" sites resulted in average annual leaching of 40 to 70% of ^{137}Cs from the litterfall. The variation depended on year (weather conditions) and the site type (the radionuclide leaching was more intensive in hydromorphic areas).

The content and reserves of stable macroelements (K, Na, Ca, etc.) never increase in the samples as a result of their transformation, but some increase was observed in A0f, especially by the end of the vegetative season (Fig. 86). The latter is likely to be caused by the element redistribution within forest litter. The differences in the behaviour of ^{137}Cs and its stable carriers confirm the presence of an upward transport of this radionuclide in forest litter. A pronounced concentration gradient of ^{137}Cs is not characteristic of stable elements, and their upward transport (if exists) is not observable.

Interesting results were obtained from the experimental simulation of a temporal storage of highly contaminated forest vegetation (the so-called "red forest") eliminated and buried in 1986 in the close vicinity of the accidental unit. The fallen trees were buried in 1986–1987 at the floodplain of the Pripyat river. In the experiment, the contaminated samples of forest litter and cellulose were buried at the depth of 1.5 m, along with a massive (several kilograms) sample of fresh uncontaminated litterfall (Fig. 87). In the eluvial (dry) landscapes, the microbiological activity was found to be very intensive: about 90% of cellulose and 70% of forest litter were decomposed (Table 75). This is apparently due to favourable conditions of moisture and aeration in the eluvial sandy soils. In the hydromorphic areas, the rate of decomposition was about 40–50%. The radionuclide loss from the

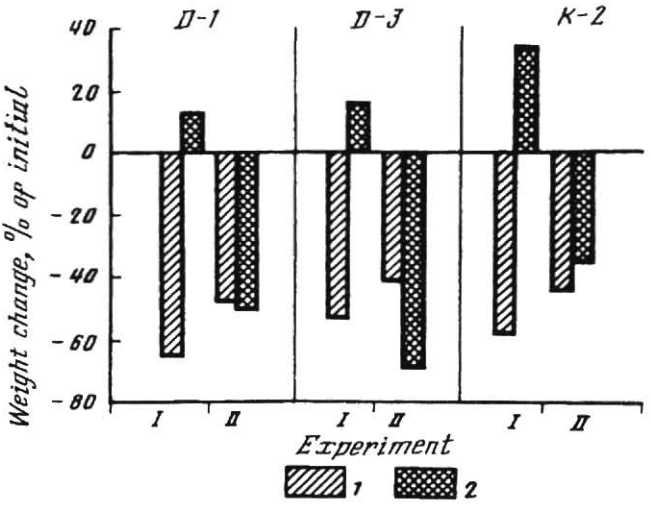

Fig. 84. Decomposition of A0l (1) and release of ^{137}Cs (2) from the experimental samples. Experiment design:

(I) A0l was sampled from and placed to the same site (K-2); (II) sample A0l was sampled from K-2 (more contaminated site) and placed to D-1 and D-3 (less contaminated sites)

contaminated samples occurred in line with the decomposition (70% and 40–60%, respectively). In both landscapes, the maximum loss was characteristic of ^{106}Ru. Thus, radionuclides are very mobile in massive underground storage of contaminated plant residues both under aerobic or anaerobic conditions. A high concentration of an organic matter provides

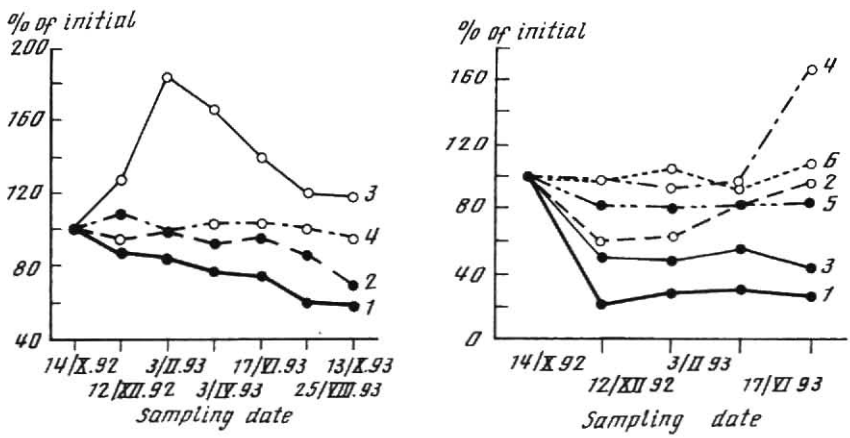

Fig. 85. Seasonal dynamics of mass and ^{137}Cs content in the samples, % of initial: (1) mass of A0l; (2) mass of A0f; (3) ^{137}Cs in A0l; (4) ^{137}Cs in A0f

Fig. 86. Seasonal dynamics of some stable elements in the experimental samples: (1) K in A0l; (2) K in A0f; (3) Mg in A01; (4) Mg in A0f; (5) Ca in A0l; (6) Ca in A0f

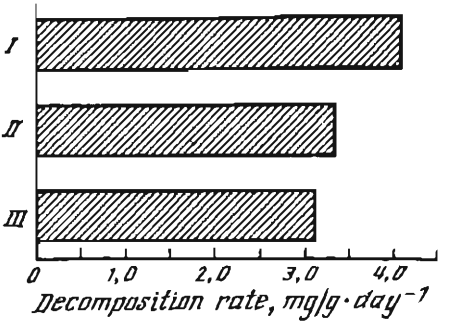

Fig. 87. Decomposition of the experimental samples (mg/g · day^{-1}) in the soil at the depth of 150 cm in the presence of fresh plant residues:
(I) cellulose; (II) needles; (III) leaves

microorganisms with a favourable substrate, which results in the intensive decomposition of the material and intensive mobilisation of the radionuclides as soluble radionuclide-organic compounds. The highest mobility under these conditions is characteristic of ^{90}Sr and ^{106}Ru. In this case, special measures shall be undertaken to suppress biological activities and stop radionuclide leaching from the temporal storage.

Table 75. Loss of organic matter and radionuclides for decomposition from the buried organic material April–October 1992 (% of initial radionuclide content or weight)

Material	Total loss, %		Loss of individual radionuclides			
	Weight	Total radionuclide content	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru
Eluvial landscape (automorphic soil)						
Mixed, fresh leaf–needle litterfall	70.6	71	62.5	67.5	66.8	100
Accumulative landscape (hydromorphic soil)						
Fresh needle litterfall	58.8	43.8	49.5	42	41.3	44
Fresh leaf litterfall	45.7	59.2	100	49.4	44.5	100

6.3. RADIONUCLIDE TRANSPORT BY INFILTRATION FLOW

Radionuclides are mobilised and come into the soil solution in the course of natural destruction of the fallout particles. Their further migration depends primarily on diffusion and mass transport (vertical infiltration) mechanisms [37, 51]. The rate of both processes depends on many factors, such as soil properties, rainfall rate and intensity, content and composition of a soluble organic matter in the soil, etc. As the soil solution percolates through the soil profile, it may be either enriched with or depleted of mobile

radionuclides due to interaction with soil solid components and biota (plant roots, fungi, and microorganisms) [144, 298, 353].

Lysimeters are widely used to study soil water mobility and composition under the native conditions [47, 107, 109, 270, 290], however, their application in native radioecological studies was limited by an extremely low concentration of radionuclides in the soil solution coupled with a relatively small volume of the lysimetric samples. That is why the studies of radionuclide behaviour in the soil solution are performed, as a rule, with the use of the "equilibrium" laboratory techniques, and the obtained data are extrapolated to the natural "dynamic" conditions [2–6, 132, 133, 198, 200, 317, 332].

The Chernobyl accident has led to severe radionuclide contamination of vast native areas, which made it possible to use lysimeters in a series of field experimental study of the radionuclide concentration and budget in various forest ecosystems [114, 190, 198].

The obtained direct data suggest that the radionuclide transport by the soil infiltration flow depends primarily on (i) the rate of the radionuclide release to the soil solution, and (ii) soil capability for radionuclide absorption from the soil solution. Radionuclide behaviour in the infiltration flow is, therefore, controlled by the same basic factors determining the behaviour of a soil nutrient and other stable elements. The most important in this context is forest litter that serves as a main sink of mobile radionuclides in the contaminated forest environments (Table 76) [241]. In our study, the solution passed through the forest litter is considered as the initial solution. The radionuclide concentration and composition of the solution from deeper soil layers reflect a relation between sorption-desorption processes in these horizons.

As the solution leaves forest litter and filters down to the mineral soil horizons, the radionuclide content in the lysimetric water decreases. Therefore, no more than 10–15% of the radionuclides contained in the initial solution penetrate deeper than 20–30 cm even in the most permeable sandy soils. The most intensive "interception" of the radionuclides occurs in the topmost organomineral layer of 5–10 cm. The rate of absorption in deeper layers decreases. Thus, a thin organomineral stratum lying immediately beneath the forest litter (the so-called A0/A1 transient subhorizon) is, in fact, the only significant biogeochemical barrier for the radionuclides leaching down the soil profile and potentially threatening the water bearing horizons (aquifers).

While leaching through A0f and A0h layers of forest litter, the solution is enriched with ^{137}Cs, and radionuclide absorption from the solution within forest litter is insignificant (Table 77). In this case, even small admixture of mineral components may increase the absorption drastically, and the radiocaesium concentration in the soil solution passed through the topmost organomineral layer A0/A1 (i.e., deeper than 1 cm beneath the forest litter) drops twofold compared to A0h. These data confirm that ^{137}Cs practically is not absorbed in the forest litter, but strongly absorbed in A0/A1 layer enriched with mineral components capable of irreversible adsorption of ^{137}Cs (see Chapter 1.3).

The coefficient of relative caesium content ($Kd = (Bq/l_{lysimetric\ water}) : (Bq/kg_{soil\ layer})$) in the investigated soils is low, which reflects low solubility of the fallout particles and a high rate of radionuclide absorption in the mineral horizons [236, 242, 277].

Table 76. Radionuclide content in the lysimetrical water from various soil layers (weighted average for 1991)

Layer, cm	Radionuclides, Bq/l					
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{90}Sr	Total
			Key site D–1			
0–5	bd*	0.07	0.5	0.32	0.85	1.74
0–10	bd	0.02	0.28	0.3	0.22	0.81
0–20	bd	0.07	0.23	bd	0.22	0.52
			Key site D-3			
0–5	bd	0.06	1.16	bd	0.62	1.84
0–10	bd	0.06	1.08	bd	0.15	1.29
0–20	bd	0.09	0.75	bd	0.1	0.94
			Key site K-2			
0–5	3.02	1.08	11.4	6.34	91.97	113.81
0–10	1.78	0.78	8.62	11.54	78.04	100.75
0–20	0.19	0.12	2.18	2.91	29.54	34.93
			Key site Sh-1			
0–5	19.79	10.91	116.22	11.42	108.04	266.39
0–10	1.34	4.29	45.09	6.52	46.25	103.49
0–20	0.43	0.56	5.59	3.98	58.09	68.65
0–30	bd	0,42	5.05	3.9	42.18	51.55

*Below detectable

Table 77. $^{134,137}Cs$ content in the lysimetrical water from various layers of the forest litter (pine forest, key site K–2, weighted average for 1993–94)

Sublayer	Depth, cm	^{134}Cs, Bq/l	^{137}Cs, Bq/l	^{137}Cs, Kd
A0l+A0f	0–3	0.64	12.692	0.004
A0l+A0f+A0h	0–4	0.65	15.913	0.006
$A0_{total}$/A1	0–5	0.4	8.487	0.003

Lysimetric water always contains some proportion of a fine solid matter transported through the soil by the infiltration flow. These particles were separated by passing the native solutions through paper filters (pore diameter of 0.5 μm). The particle content was low: 0.01–0.02 g/l in the water from upper (0–5–10 cm) layers and up to 0.07 g/l in the water from 10–20 cm layer (Table 78). The proportion of ^{137}Cs attributed to the particles varies from 0.45% (in the

Table 78. ^{137}Cs and ^{40}K content in the liquid and solid components of the infiltration flow (key site K–2, weighted average for 1994)

Depth, cm	Transport of solid material, g/l	^{137}Cs			^{40}K		
		Total transport, Bq/l	Content in the solid material, % of total	Content in solid material, Bq/kg	Total transport, Bq/l	Content in the solid material, % of total	Content in solid material, Bq/kg
0–5	0.02	38	0.45	8500	2.59	49.81	64500
0–10	0.01	3.1	6.03	19000	2.27	44.93	102000
0–20	0.07	2.8	5.71	2286	3.25	47.69	22143

organic layer) to 6% (in deep mineral layers). The latter suggests that *lessivage* contributes to the radionuclide migration and redistribution in sandy soils. The autoradiography and radionuclide composition of the soil deep layers also confirms active migration of the radioactive particles down the soil profile [65, 66, 72]. This process is likely to involve not only migration of "hot particles" as themselves, but also the transport of ^{137}Cs irreversibly incorporated in the clay particles. The latter is confirmed by the data on ^{137}Cs and ^{40}K content in the "mineral residue" after treatment of the isolated solid matter by a hot mixture of HCl/HNO_3. The treated particles contained, therefore, only caesium irreversibly adsorbed within the crystal lattice. The ^{40}K content in the soil is known to be in equilibrium with stable potassium, a high content of which is an indicator of clay minerals. Therefore, a high proportion of both ^{137}Cs and ^{40}K in the solid matter after treatment suggests that caesium in the "solid" intrasoil flow is attributed to the clay minerals.

In this case, the radionuclide content in the infiltration flow from different layers varies in a wide range depending on the radionuclide nature, vegetation cover, and soil type.

Radionuclide mobility in the infiltration flow is reflected in both the content and radionuclide composition of the lysimetric water from various soil layers. In general, the investigated radionuclides may be arranged into the following series by migration ability: $^{90}Sr > {}^{106}Ru > {}^{137}Cs > {}^{144}Ce$.

The weighted average mobility of ^{90}Sr in the infiltration flow is about 1.5–2 times higher than ^{137}Cs, and under pure pine stand the corresponding difference reaches a factor of 5. The latter suggests that the radionuclide infiltration depends on type of ecosystem (phytocenosis). This is likely due to some specific features of pine forest where forest litter is enriched with a low-molecular soluble organic matter that promotes mobility of ^{90}Sr [2–6, 78]. In contrast with it, high-molecular humus components are known to reduce mobility of ^{90}Sr, ^{144}Ce, and ^{106}Ru in the soil. These components are more abundant in the soils under a mixed stand [221, 245, 315, 322, 326]. A high mobility of ^{106}Ru may depend also on its ability to form anionic and cationic complexes with sol-

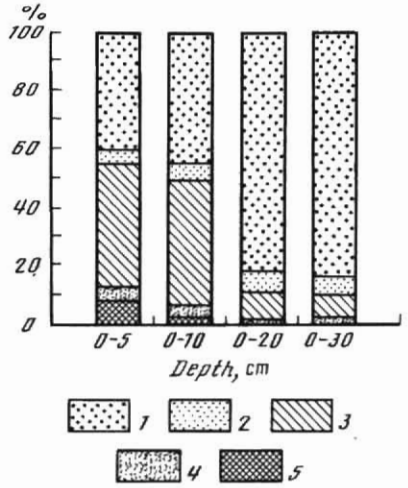

Fig. 88. Radionuclide composition of lysimetrical waters from various soil layers (weighted average for 1991, "near zone"): (1) ^{90}Sr; (2) ^{106}Ru; (3) ^{137}Cs; (4) ^{134}Cs; (5) ^{144}Ce

uble soil components [211]. Anionic forms of ruthenium are highly mobile: in the presence of fulvic acids its solubility increases by 2–3 orders of magnitude [42]. In addition, ruthenium is more sensitive to acidification compared to other radionuclides [211, 317]. Thus, the environmental conditions in a pine forest promote the mobility of ^{106}Ru and ^{90}Sr (fulvic humus, low pH, etc.).

Adsorption capacity of the investigated soils for ^{137}Cs and ^{90}Sr is different (Table 76). The maximum difference is observed between peat (site D-3) and mineral soils (other sites). In mineral soils, ^{137}Cs content decreases dramatically in the lysimetric water from mineral layers of 5–10 and 10–20 cm (by a factor of 1.5–2 and 2–20, respectively). ^{137}Cs practically is not absorbed in the layer of 5–10 cm of peat-gley soil, and the farther increase in absorption is not so pronounced [151, 171, 300, 325]. ^{90}Sr is absorbed from the infiltration flow primarily in the upper horizons of peat soil.

Specific physicochemical properties of the initial fallout have a long-lasting effect on the radionuclide content in the soil solutions. The radionuclide content (particularly, ^{144}Ce, ^{106}Ru, and ^{90}Sr) is the lowest in the "near zone" of ChNPP (site Sh-1). In this case, low solubility of the fallout is likely to dominate over all other factors (phytocenosis, soil properties, etc.), since the soils in the area are enriched with the low soluble fuel components [110, 111]. The behaviour of ^{106}Ru is less clear, but there are all reasons to suppose that in this territory both ^{106}Ru and ^{90}Sr are attributed to the fuel component of the fallout. In the remote zone, a considerable proportion of ^{106}Ru was represented by volatile RuO4 condensed on fine fallout particles (less than 0.2 μm) [56, 217].

The radionuclide composition of the lysimetric waters is very different from the radionuclide composition of the soil as a whole. The difference is more manifested for deeper soil layers (Table 52, Fig. 44, 88). The proportion of ^{106}Ru and ^{90}Sr in the radionuclide composition increases and the proportion of ^{144}Ce decreases. The share of ^{137}Cs also decreases with depth, but smoother than ^{144}Ce. The above-discussed differences in the radionuclide composition are most characteristic of pure pine cenoses.

6.3.1. Radionuclide Dynamics in the Infiltration Flow

Dynamics of the radionuclide composition in lysimetric water depends basically on the radioactive decay of relatively short-lived radionuclides, such as ^{144}Ce and ^{106}Ru. As a result, the proportion of these radionuclides in the infiltration water became negligible five years after the accident. Currently, the radionuclide composition is almost completely represented by ^{137}Cs, ^{90}Sr, ^{238}Pu, and $^{239+240}Pu$.

Seasonal dynamics of the radionuclide content in the infiltration water is most manifested in the A0 horizon (forest litter as a whole). In deeper layers, the variation range decreases, since radionuclides may appear in the mineral soil profile only due to their migration from the forest litter. Mineral soil layers intensively absorb radionuclides in proportion to the initial content (Fig. 89). This is especially characteristic of ^{137}Cs because of high irreversible adsorption of this radionuclide in the soil. The radionuclide equilibrium in the "solid-liquid" system is shifted to the "solid" side, as the radionuclide concentration in the liquid phase increases [28]. Thus, a larger soil thickness smoothes the seasonal variation of the radionuclide content in the infiltration solution from deeper layers (0–20 and 0–30 cm).

There are some common features in the seasonal dynamics of all radionuclides: their concentration in the soil solutions increases in the early spring and late summer (or the early autumn). This is likely due to the dynamics of biological activity in the soil, which is known to promote radionuclide release from the fallout particles [108, 126, 292, 333]. In addition, increased biological activity promotes the transformation processes in the forest litter and increases the soluble organic matter (SOM) content in the soil solution. This intensifies the radionuclide mobilisation from the forest litter and mineral soil horizons.

In spring, the soil temperature increases, which, along with a high proportion of fresh plant residues in the forest litter, pushes on the microbiological activities and increases the radionuclide mobilisation by microbiota and SOM.

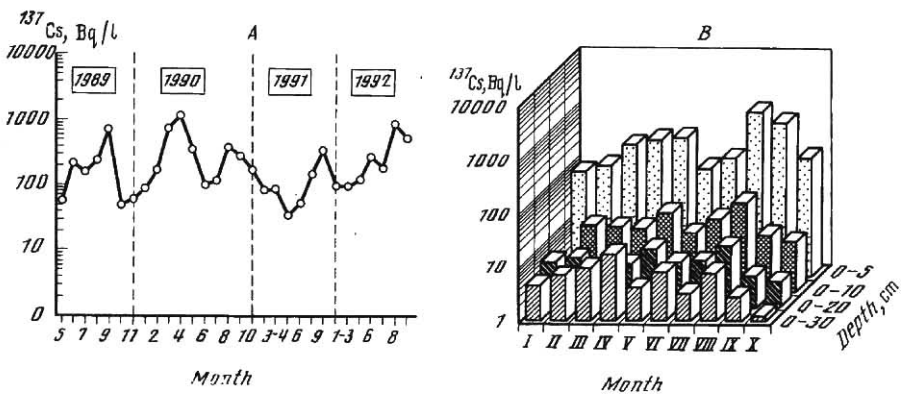

Fig. 89. Seasonal and multiyear dynamics of ^{137}Cs in the lysimetrical waters from (A) forest litter and (B) mineral soil layers

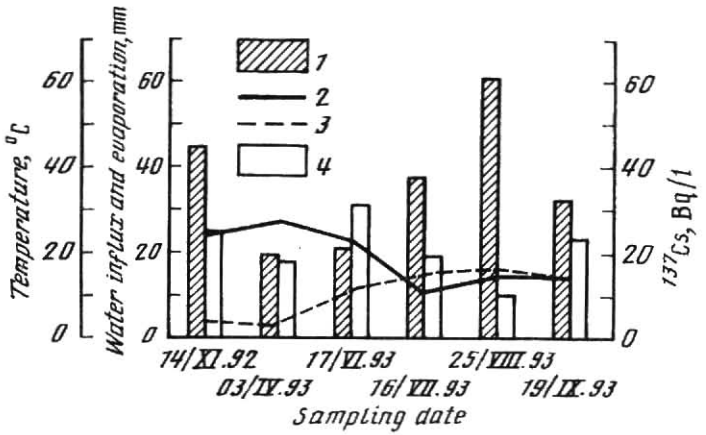

Fig. 90. Seasonal dynamics of water influx to the lysimeters, ^{137}Cs content, evaporation, and surface air temperature (30 km exclusion zone, key site K-2):
(1) water volume; (2) ^{137}Cs content; (3) temperature; (4) evaporation

As the fresh substrate is depleted and soil surface desiccated, the microbiological activity is damped out, which causes the corresponding decrease in the radionuclide content in the lysimetric water. By the late summer or early autumn, the addition of fresh litterfall to the soil intensifies the microbial activity again, which results in a new increase in the concentration of mobile radionuclides. In winter or late autumn, the biological activity is damped and the radionuclide concentration in the soil solution decreases in line with the soil temperature.

Variation of the radionuclide concentration in the lysimetric water during a vegetative season depends also on the rainfall and rain intensity (Fig. 90). The concentration is inversely proportional to the rainfall, which is most manifested during rain-abundant seasons (the early spring and late autumn). Seasonal peaks of the radionuclide concentration are sometimes shifted in response to the weather conditions.

6.3.2. Radionuclide Loss for Infiltration

The radionuclide loss from various soil layers may be estimated from the radionuclide concentration and the flow rate of each layer.

The average annual volume of the lysimetric water in the 0–30 cm layer of the investigated forest soils is estimated at 30–60% of total annual rainfall and decreases with depth (Table 79).

The monthly averaged water infiltration to the lysimeters varies from 20 to 100% of the monthly rainfall. Such a high variability is caused by superposition of at least three factors: rainfall, temperature, and soil moisture dynamics. Minimum and maximum of the infiltration fall on summer and winter months, respectively. However, such a considerable monthly variation has no visible

Table 79. Average annual output of water from various soil layers (% of annual rainfall)

Depth, cm	D–1		D–3		K–2			Sh–1		
	1989	1990	1989	1990	1989	1990	1991	1989	1990	1991
0–5	44.5	45.5	45.5	42.6	40.3	49.8	44.5	60.5	52.3	64.4
0–10	35.2	35.8	51.3	52.2	32.5	36.5	35.2	39.5	45.2	38.7
0–20	29.5	36.7	40	39.9	22.6	28.3	30.6	36.5	40	39.3
0–30	nd*	nd	nd	nd	nd	nd	nd	37.4	45.2	35.6

* No data

effect on the annual infiltration that is quite stable and varies only slightly by years. Thus, variation in the annual radionuclide loss from the soil layers depends on the radionuclide mobility rather than the total volume (rate) of the intrasoil water flow. The absolute radionuclide loss (Bq/sq.m) is not very informative in terms of biogeochemistry, since this value reflects a trivial "concentration-deposition" dependence. For our purpose, it is more convenient to assess the relative radionuclide loss (A) defined as follows:

$$A = \frac{\text{radionuclide flow from a soil layer (Bd/sq.m)}}{\text{deposition in the layer (Bq/sq.m)}} \cdot 100\%$$

The calculations show that only a negligible proportion of radionuclides leaves the root-abundant soil layer with the infiltration flow. The corresponding average annual loss of radionuclides from the forest litter constitutes from 0.06 to 0.9 % of the deposition in this layer. The corresponding values for the 0–20 cm layer are as low as 0.001–0.2 %. The relative loss of ^{90}Sr and ^{106}Ru from the layers of 0–10 and 0–20 cm is ahead of ^{144}Ce and even ^{137}Cs (Table 80).

All these features are most manifested in pine forests where the total loss of radionuclides due to infiltration is maximal, and the relative loss of ^{90}Sr exceeds that of ^{137}Cs by one order of magnitude. In deciduous forests, the corresponding differences do not exceed a factor of 1.5–2.

In peat soil, the radionuclide transport with infiltration has some specific features. Loss of ^{137}Cs from these soils is higher than that of ^{90}Sr, which is one more evidence of an extremely low capacity of these soils for caesium retention.

Comparison of the final distribution of radionuclides in the soil profile five years after the accident with the average annual radionuclide replacement by infiltration suggests that the total radionuclide flux exceeds its infiltration by 1–1.5 orders of magnitude. It means that the infiltration is not an important factor of radionuclide redistribution in the upper 20–30 cm soil layer.

Transport of plutonium by the infiltration flow is of particular interest because of insufficient data on the plutonium behaviour in native soils. The relative annual flux of plutonium isotopes is about 0.1 % per year, which is comparable with ^{137}Cs (Table 81). The behaviour of mobile plutonium in the soil depends, however, on the ecosystem type. Mineral soil layers under a pine stand practically do not

Table 80. Relative output of various radionuclides with the infiltration flow ([Bq/m$^2_{water}$]:[Bq/m$^2_{soil}$]) .100%; weighted averages for1991)

Depth, cm	Radionuclide					
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{90}Sr	Total
			Site D–1			
0–5	bd*	0.077	0.056	0.364	0.1	0.119
0–10	bd	0.016	0.024	0.265	0.02	0.065
0–20	bd	0.054	0.017	bd	0.02	0.018
			Site D–3			
0–5	bd	0.067	0.128	bd	0.07	0.053
0–10	bd	0.067	0.088	bd	0.02	0.034
0–20	bd	0.093	0.112	bd	0.01	0.043
			Site K–2			
0–5	0.15	0.1	0.11	0.52	0.92	0.36
0–10	0.06	0.05	0.06	0.67	0.57	0.283
0–20	0.01	0.01	0.01	0.14	0.17	0.066
			Site Sh–1			
0–5	0.078	0.108	0.115	0.095	0.11	0.1
0–10	0.003	0.024	0.025	0.031	0.03	0.022
0–20	0.001	0.003	0.003	0.019	0.03	0.012
0–30	bd	0.002	0.002	0.014	0.02	0.007

* Below detectable

Table 81. Relative output of ^{137}Cs and plutonium isotopes from various soil layers (June–August, 1989)

Depth, cm	Relative output, %			
	^{137}Cs	$^{239+240}$Pu	^{238}Pu	$^{238+239+240}$Pu
		Key site K–2		
0–5	0.073	0.071	0.079	0.073
0–10	0.015	0.089	0.096	0.091
0–20	0.03	0.065	0.073	0.068
		Key site Sh–1		
0–5	0.087	0.077	0.074	0.076
0–10	0.004	0.006	0.005	0.005
0–20	0.002	0.003	0.003	0.003
0–30	0.003	0.001	0.002	0.001

absorb plutonium, whereas the same soil layers under a mixed stand absorb this radionuclide very actively (no more than 0.001 % of the total Pu leaves soil profile annually). In other words, plutonium mobility in the soil solution under a pine forest exceeds that under a mixed and pure deciduous stand by an order of magnitude. It is difficult to suggest a clear explanation for this phenomenon, since the soil chemistry of plutonium is poorly investigated. It may be suggested that a high migration ability of Pu in the soil under pine forests is determined by the same causes as ^{106}Ru (see above). The latter is indirectly confirmed by an increased mobility of plutonium isotopes under low pH and in the presence of a soluble organic matter [211, 285]. In spite of a low rate of plutonium infiltration, its chance to reach local aquifers is rather high compared to other ^{137}Cs because of the extremely long half-life and practically unlimited migration time (87.7 years for ^{238}Pu, 24100 years for ^{239}Pu, and 6570 years for ^{240}Pu).

6.3.3. Transport of Some Stable Elements by Infiltration Flow

Potassium, calcium, and iron are believed to be non-isotopic carriers of ^{137}Cs, ^{90}Sr, and ^{106}Ru, respectively [307]. The concentration of these elements in natural objects exceeds, however, the concentration of the corresponding radionuclides by 10–12 orders of magnitude. The question is whether the radionuclide behaviour in natural environments is different from the behaviour of the carriers under the same conditions.

^{137}Cs and ^{90}Sr in the solid-liquid soil system are absorbed more effectively than their non-isotopic carriers, particularly in the 5–10 cm layer (Table 82). It may be supposed that these differences are due to unsteady state of the

Table 82. Absorption of some radionuclides and stable non-isotopic carriers from the infiltration flow (% of content in the lysimetrical water from the 0–5 cm layer)

Depth,cm	Absorption in the layer, (%)					
	^{40}Ca	^{90}Sr	^{39}K	^{137}Cs	^{56}Fe	^{106}Ru
			Key site D–1			
5–10	5.3	53.8	35.0	79.0	10.3	9.6
5–20	0.0	53.8	35.0	85.0	53.8	39.6
			Key site K–2			
5–10	10.1	75	22.8	85.8	5	20
5–20	46.4	82.8	33.6	88.6	65	43
			Key site Sh–1			
5–10	30.5	58.0	40.9	92.6	20.1	50
5–30	39.4	61.5	48.4	96.4	87	93.2

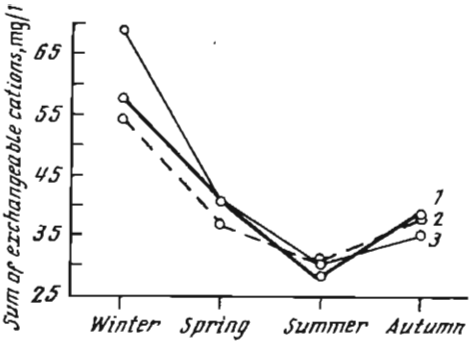

Fig. 91. Seasonal dynamics of stable elements in the infiltration water (the near exclusion zone, weighted averages for 1990-1993):
(1) 0–5 cm; (2) 0–10 cm; (3) 0–20 cm

Chernobyl-derived radionuclides in the ecosystem compared to the age-long equilibrium of the corresponding stable carriers.

The behaviour of radioactive ^{106}Ru and its stable carrier Fe is more agreed compared to the pairs ^{137}Cs/K and ^{90}Sr/Ca, although the main flow of Fe is known to originate in the mineral soil layers and not in the forest litter (like ^{106}Ru, K, and Cs) [33]. This circumstance makes it incorrect to consider Fe to be fully analogous to ^{106}Ru. Besides, the chemical properties of ruthenium are very different from those of iron, although the first may co-precipitate with the latter [1].

The infiltration flow of stable elements also undergoes a considerable seasonal variation, but the dynamics of their concentration is different from that of the radionuclides (Fig. 91). The maximum and minimum contents of stable elements fall on winter and summer months, respectively, whereas the radionuclide content has two maximums: spring and autumn. This is likely to depend on the effect of biota, pH, and eH on stable elements, radioactive fallout, and "free" radionuclides in the soil.

Increase in soil biological activity in spring and early summer is accompanied by active immobilisation of nutrients due to their uptake by the soil organisms. Both stable elements and radionuclides may be equally incorporated to the organisms, but their sources are different. It may be supposed that the most portion of mobile (available) nutrients in the root-abundant layer is depleted by the middle summer. At the same time, the intensity of radionuclide mobilisation from the fallout particles increases in proportion with the soil biological activity (see Chapter 5.2). Therefore, the radionuclide dynamics in the soil solution depends primarily on the rate of radionuclide release from the fallout particles, whereas the dynamics of stable elements depends chiefly on the biological uptake. The differences are expected to be smoothed in the course of time, and, in the future, the radionuclide dynamics is expected to be similar to that of non-isotopic stable carriers.

Thus, the rate of radionuclide redistribution in the soil due to infiltration is very low and constitutes from $n \cdot 0.1$ % to $n \cdot 0.001$ % of the deposition per year depending on soil layer, soil type, forest type, and radionuclide. Radionuclide mobility in the infiltration flow may be expressed as the following series: ^{90}Sr > ^{106}Ru > ^{137}Cs > ^{238}Pu > ^{239,240}Pu > ^{144}Ce. The maximum mobility of ^{90}Sr, ^{106}Ru and plutonium isotopes is characteristic of pine forests, and ^{137}Cs is most

mobile in the organic (peat) soils. The most intensive radionuclide outflux is observed from the forest litter. The radionuclide outflux from the 20–30 cm layer decreases by 1–2 orders of magnitude except for the case of ^{137}Cs in the peat soil, and ^{90}Sr, ^{106}Ru and plutonium isotopes in pine forests. In this case, radionuclides filter down the soil profile without a significant interception by internal soil horizons. Plutonium isotopes are the most interesting in terms of possible contamination of ground water in the future because of an extremely long half-life, low absorption, and low TF to the plants and other soil organisms.

6.4. ^{137}CS IN THE THROUGHFALL AND STEM FLOW

The so-called crown water (throughfall) and the stem flow is reported to play an important role in nutrient migration (including K and Ca) in forest biogeocenoses. No less than a half of the total element flux through the ecosystem is reported to be attributed to throughfall or/and stemflow (47, 102, 103, 148). Table 83 presents the data on ^{137}Cs concentration and output from the tree tissues to the forest litter with throughfall and stem flows.

The average annual ^{137}Cs concentration in the throughfall is 5.3 Bq/l and does not exceed 9.42 Bq/l, whereas in the stem flow these indices are 219 and 244 Bq/l, respectively (Table 83).

Relatively high concentration of ^{137}Cs in the stem flow compared to the throughfall can principally be determined by two processes: (1) its sequential extraction from the external bark, and (2) extraction from the residual particles of the initial surface contamination. The first process is caused by a relatively slow movement of rain water down the stem and gradual enrichment of water with ^{137}Cs. The water falling through the pine crown contacts the needles for too short time to be enriched with radiocaesium to the same extent as the stem flow. The second process may take place simultaneously with the first one, but ^{137}Cs is leached from the fallout particles rather than from the plant tissues. The contribution of this pathway is confirmed by the fact that the external layers of pine bark in the exclusion zone are still one of the most contaminated component of stand [241].

It is of interest that the picture is absolutely different in the forests of remote territories (e.g., in Ireland) where no considerable difference in ^{137}Cs content between the throughfall and stem flows was found [291, 336, 337]. Thus, resid-

Table 83. ^{137}Cs content in the throughfall and stem flow (weighted averages for 1994)

Source	Water, l/m^2	^{137}Cs		
		Bq/l	Bq/m^2	% of deposition (soil included)
Throughfall	173.75	5.31	919.44	0.044
Stem flow	0.65	218.77	142.86	0.007

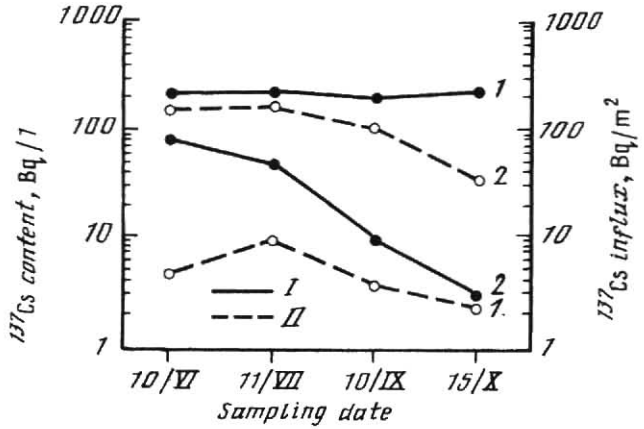

Fig. 92. Seasonal dynamics of the concentration (left scale) and monthly influx (right scale) of ^{137}Cs to the soil surface with (I) stem flow and (II) crown waters (throughfall): (1) ^{137}Cs content; (2) ^{137}Cs influx

ual contamination of the external plant tissues still make a considerable contribution to the radionuclide composition and content of the stem flow and (maybe) throughfall.

An annual influx of ^{137}Cs to the soil by the throughfall and stem flow is 919 and 143 Bq/m^2, respectively. These values were calculated using experimental data on the crown closeness (75%) and stand density (8 trees per 100 m^2) in the key plot. Therefore, the contribution of the throughfall to the downward ^{137}Cs flux from the stand is higher than the corresponding contribution from the stem flow, in spite of high ^{137}Cs concentration in the latter.

At the same time, annual coming of ^{137}Cs with the stem flow directly to the stem-adjacent area is of significance (1850 Bq/tree on the average). This increases the heterogeneity in the distribution of mobile and available forms of ^{137}Cs over the territory and enriches the stem-adjacent soil areas with this nuclide. The latter may influence the local root uptake, and in this case the TF calculated based on the average content of ^{137}Cs in the soil can be significantly underestimated (since the actual TF from the stem-adjacent area is much higher than the average over the territory) [297].

The relative weighted annual influx of ^{137}Cs to the soil with the throughfall and stem flow is now 0.044% and 0.007% of the radionuclide deposition in the ecosystem (soil included), respectively. It is obvious that this value was significantly higher in the first years after the accident because of an extremely high surface contamination of the tree canopy during the initial fallout. When taking into account that the radionuclides leached currently from the plant tissues by throughfall and stem flow are mobile, i.e., readily available for plants, the important role of these fluxes in the biological cycle of ^{137}Cs is obvious.

The dynamics of ^{137}Cs influx to the soil with the throughfall and stem flow depends on seasonal variation of the radionuclide content and rainfall rate during the vegetative season (Fig. 92).

6.5. REDISTRIBUTION OF RADIONUCLIDES IN THE SYSTEM OF GEOCHEMICALLY LINKED LANDSCAPES

According to current biogeochemical concepts, the landscape-geochemical features of any territory have a significant effect on the processes of the primary and secondary distribution of chemical elements in the territory [116, 124, 179]. Eluvial (normally automorphic) landscapes tend to lose elements and nutrients, and accumulative (normally hydromorphic) landscapes tend to accumulate them. At the same time, the rate of the large-scale element redistribution on a landscape scale is very low. It was shown that 10 years after the atmospheric tests, the deposition of "weapon" ^{90}Sr in the automorphic forest landscapes decreased by a factor of two and increased by the same factor in the hydromorphic areas [245–248].

Table 84. Multiyear dynamics of ^{137}Cs and ^{90}Sr deposition in the soils of geochemically linked landscapes (30 km exclusion zone, kBq/m^2)

Radionuclide	Years				
	1986	1987	1988	1989	1990
	Eluvial landscape (key site D–1)				
^{137}Cs	242.3	209.4	192	180.9	172.4
^{90}Sr	nd*	154.3	nd	66.6	40.7
	Accumulative landscape (key site D–3)				
^{137}Cs	237.2	209.4	194.6	185	177.6
^{90}Sr	nd	168.4	nd	84	51.8

Radionuclide	Years				
	1991	1992	1993	1994	1995
	Eluvial landscape (key site D–1)				
^{137}Cs	165.8	160.1	156.1	152.4	148.7
^{90}Sr	nd	31.3	nd	nd	nd
	Accumulative landscape (key site D–3)				
^{137}Cs	172.1	167.2	163.2	159.8	156.9
^{90}Sr	nd	53.8	57.8	nd	nd

* No data

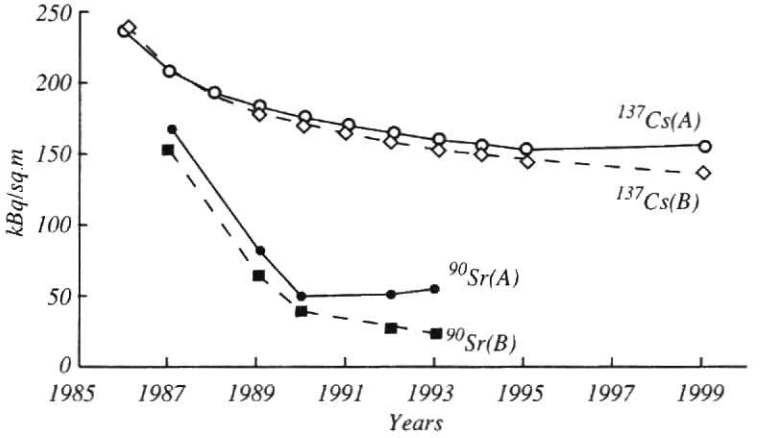

Fig. 93. Multiyear dynamics of ^{137}Cs and ^{90}Sr in the soils of geochemically linked landscapes:
(A) eluvial landscape; (B) accumulative landscape

High deposition over the exclusion zone makes the problem of a large-scale redistribution of radionuclides of high practical significance, especially in terms of their possible concentration in the accumulative landscapes. Some authors suggest that such vertical and lateral migration is already observable, though weakly manifested [268, 269]. Our direct field studies, however, revealed no significant changes in the deposition of ^{137}Cs and ^{90}Sr for 10 years since 1986 (Table 84). By 1996, the difference (Δ) between ^{137}Cs deposition in the soils of adjacent, geochemically linked landscapes varied within a range of statistical variability. Nevertheless, some tendencies indicate that the processes actually take place (Fig. 93).

^{90}Sr redistribution in the system of geochemically linked landscapes is somewhat more pronounced than that of ^{137}Cs, though less manifested compared to "weapon" strontium and also close to the statistical error [246]. The correctness of the opinion in favour of more intensive redistribution of ^{90}Sr is confirmed by the data on its migration to the river network in the exclusion zone. The annual rate is estimated at 0.65% for ^{90}Sr and 0.1–0.2% for ^{137}Cs [278].

More correct information on geochemical redistribution of ^{137}Cs can be obtained taking into account its inventory in the vegetation and in the soil. In this case, the differences between the decay-corrected depositions in the adjacent, geochemically linked eluvial and accumulative landscapes are estimated at 40 kBq/sq.m or 20 % for 10 years. Assuming the same initial deposition in the investigated eluvial and accumulative areas and taking into account the statistical error, the estimated rate of the annual inter-landscape redistribution of ^{137}Cs is about 1% (conservative value).

The future differences between the eluvial and accumulative landscapes in ^{90}Sr deposition may be estimated on the basis of the present differences in the content of stable strontium in the landscapes. Fig. 94 suggests that the deposi-

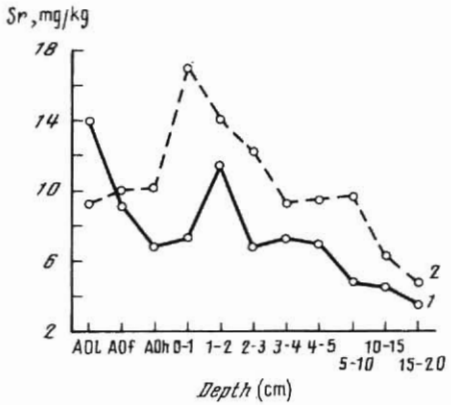

Fig. 94. Vertical distribution of stable strontium in the forest soils of geochemically linked landscapes:
(1) eluvial landscape; (2) accumulative landscape

tion of stable strontium in the accumulative landscapes is two to three times higher compared to adjacent eluvial landscapes. The final differences in the ^{90}Sr content will unlikely exceed this value, since accumulative landscapes slowly lose elements to the river network.

The radionuclide redistribution runs much faster within the elementary landscapes, on a scale of microtopography (meters at the horizontal scale and centimetres at the vertical scale). As soon as 5 years after the accident, the radionuclide content in the dish-shaped depressions, local hollows, etc., was 5-30% higher compared to the convex elements of local topography (Table 85). The difference was most manifested for ^{106}Ru and ^{137}Cs, which confirms a high mobility of these radionuclides (see above). The share of ^{106}Ru in the radionuclide composition in some depressions was 1.5 times higher than that in the adjacent flat areas.

Even more manifested is the radionuclide redistribution within the landscape on a scale of mesotopography (tens meters at the horizontal scale and

Table 85. Deposition and radionuclide composition of the adjacent, geochemically linked forms of microtopography (the exclusion zone, 1991, average of n = 15)

Form of microtopography	Radionuclides				Total
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	
	Deposition, kBq/m²				
Microconvexity	17.02	15.54	157.99	12.95	203.5
Microdepression	17.76	19.98	191.66	19.24	248.64
	Relative units				
Microconvexity	100	100	100	100	100
Microdepression	104.2	122.2	117.6	132.7	118.3
	Radionuclide composition, %				
Microconvexity	8.4	7.7	77.6	6.3	100
Microdepression	7.2	8	77	7.8	100

Table 86. Deposition and radionuclide composition of the adjacent, geochemically linked of mesotopography (30 km exclusion zone, 1991, average of n =15)

Form of micro-topography	Radionuclides					Total
	^{144}Ce	^{134}Cs	^{137}Cs	^{106}Ru	^{90}Sr	
	Deposition, kBq/m²					
The top of a sand belt	0.45	0.3	3.33	0.24	1.12	5.44
Slope of the sand belt	0.49	0.34	3.32	0.38	1.55	6.08
The slope basement	0.63	0.53	5.41	0.38	1.87	8.82
Bottom of the adjacent hollow	0.6	0.41	4.49	0.57	1.28	7.35
	Relative units					
The top of a sand belt	100	100	100	100	100	100
Slope of the sand belt	108.9	113.3	99.7	158.3	138.4	111.8
The slope basement	140	176.7	162.5	100	167	162.1
Bottom of the adjacent hollow	133.3	136.7	134.8	237.5	114.3	135.1
	Radionuclide composition, %					
The top of a sand belt	8.3	5.5	61.2	4.4	20.6	100
Slope of the sand belt	8.1	5.6	54.6	6.2	25.5	100
The slope basement	7.1	6	61.3	4.4	21.2	100
Bottom of the adjacent hollow	8.2	5.6	61.1	7.7	17.4	100

meters at the vertical scale). Within the slopes of 15° and steeper, and the height differences of 2–3 m, the σ values may reach 50–100 % (up to 137 % for ^{106}Ru) (Table 86). The maximum accumulation of radionuclides takes place in the marginal areas of the concave elements of mesotopography, i.e. at footslopes. In general, the most manifested geochemical barriers of various scales are attributed to the marginal areas of the accumulative zones. This phenomenon is in agreement with the data on the large scale redistribution of stable elements and nutrients by different landscapes [32, 179]. Some authors believe, however, that lower radionuclide content in the central areas of the depressions is due to more intensive infiltration losses [26, 269]. In our opinion, this may be true for the case of dish-shaped depressions of tens to hundreds square meters in area rather than large-scale accumulative landscapes.

Thus, the present differences in the radionuclide content at different elements of micro- and mesotopography are determined by the migration processes rather than spatial variability of the initial deposition. Moreover, many

authors suppose that, initially, the dish-shaped depressions were, as a rule, less contaminated than the adjacent convex topographical elements. The difference was smoothed with time, and the deposition in the depressions increased drastically by the 3–4th year after the fallout. The initial increase in deposition was followed by somewhat decrease in the very central area of each depression [14, 32, 268, 269].

Redistribution of the Chernobyl-derived ^{90}Sr by the elements of mesotopography is low-manifested compared to "weapon" ^{90}Sr [246]. This may be explained by (i) specific features of the Chernobyl fallout (see above), and (ii) coarse consistence of the soils in the exclusion zone, which promotes ^{137}Cs and ^{106}Ru migration. The very central areas of local dish-shaped depressions in sandy soils are likely to serve as a vertical pathway for ^{90}Sr migration to the ground water rather than a geochemical barrier [268, 269].

Thus, the process of large-scale redistribution of radionuclides in the system of geochemically linked landscapes is least manifested for ^{137}Cs (currently < 1 % per year). It is better manifested for ^{90}Sr, although its intensity is lower than the rate of the physical decay. The intensity of radionuclide redistribution within the elementary landscapes at the scale of meso- and microtopography is more evident and reaches 10 % per year.

7. CONCEPTUAL MODEL AND PARAMETERS OF BIOGEOCHEMICAL MIGRATION OF ^{137}Cs IN FOREST LANDSCAPES

Calculation of ^{137}Cs inventory and biogeochemical fluxes through living components of forest ecosystems

To assess individual contribution of each ecosystem component to the biological cycle of any chemical element, it is necessary to estimate (i) the total reserves, annual production, and annual return of each component in the ecosystem, and (ii) the average content of the investigated element (radionuclide) in each component.

For herbaceous vegetation, an annual increment was determined based on the multiyear monitoring of the dominant species. The gross annual production was calculated as the total biomass of these species in the maximum production stage. An annual return of the organic matter to the soil was assumed to be 100 %, since almost all dominant herbaceous species were represented by annual plants. The average root biomass was assumed to be 35 % of the above-ground biomass, and an annual production and return of the root biomass was assumed to be 1/3 of the total herbaceous biomass [17]. ^{137}Cs content was assumed to be the same both in the above-ground and the root phytomass, although some researchers believe that the grass roots contain a larger proportion of radionuclides [146].

The total biomass of mosses was determined directly in the field. An annual production of mosses is reported to vary from 10 to 25 % of the total biomass, an annual return of the organic matter from the moss cover to the soil is assumed to vary from 70 to 98 % of an annual production [143]. For the investigated territory, the annual return is assumed to be equal to 98 % of the total annual production [17].

Estimation of the corresponding parameters for soil mycobiota (fungus complex) is quite problematic because of (i) apparent difficulties in quantitative measurements of the underground mycelium biomass, and (ii) variable yield of the fruit bodies and several "generations" of the fruit bodies during a vegetative period. In boreal forest ecosystems, the fruit body biomass may vary from 5 to 100 kg/ha* [38]. As to the mycelium, its total reserves in the soil reaches 200 g/m^2 [335]. The fruit body /mycelium weight ratio varies from 1/62.6 to 1/154 for saprotrophic and symbiotrophic fungi, respectively [38]. The maximum reserves of the total fungus biomass including mycelium (over 200 g/m^2) is characteristic of podzolic and soddy-podzolic soils under pine forests [71, 149]. In some cases, the weight proportion of fungi

* Hereinafter, fungus (mycelium) biomass is reduced to the dry weight (105° C)

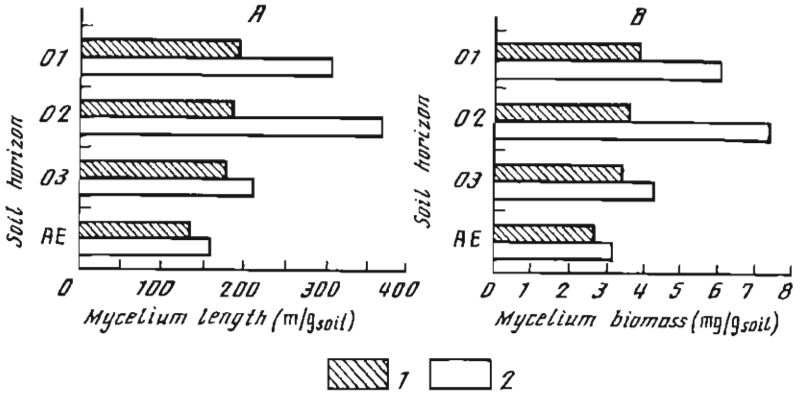

Fig. 95. Mycelium (A) length and (B) biomass in podzolic sandy soils under (1) pine and (2) mixed forest [86]

mycelium may reach 20 % of the total weight of the forest litter [38]. The recent data obtained by the thermoluminescent analysis suggest that mycelium contributes 88 to 99 % of the total microbial biomass in the soil. The latter varies from $n \cdot 10^2$ to $n \cdot 10^3$ g/m^2 depending on ecosystem, soil properties, and weather conditions [185]. In spite of high uncertainty of the information on mycelium and the fruit body biomass in the soil, we tried to estimate the contribution of mycobiota to ^{137}Cs cycle in the investigated forest ecosystems using available literature and our field data.

The average yield of fungus fruit bodies is about 15 kg/ha. In broad-leaved forests of Ukrainian Polessie, on automorphic soils, the highest mycelium content is attributed to the forest litter and decreases down the soil profile (Fig. 95). Nevertheless, the maximum reserves of mycelium is attributed to mineral soil layers because of higher bulk density of the latter [185]. Judging by the mycelium content, the total fungus biomass is estimated at 213 to 290 kg/ha under mixed (deciduous-pine) stand and pure pine stand, respectively [71, 149, 335]. The latter value (290 kg/ha) is assumed to be true for the alder forests on hydromorphic soils as well. We consider it to be correct as a conservative estimation, since fungus biomass in the alder ecosystems is reported to be even higher [185]. We equate the ^{137}Cs concentration in the fruit bodies and mycelium, which is in practice for this type of study [335]. Experimental data on soil fungi suggest that TF^{137}Cs in the fungi mycelium of higher (basidial) fungi exceeds that in the fruit bodies by a factor of 1.5–2 depending on environmental conditions and availability of ^{137}Cs [294]. In general, 10 to 63 % of ^{137}Cs deposition in the forest soils are reported to be accumulated in the fungi mycelium [312, 335]. Some authors believe that ^{137}Cs and nutrients accumulated in the mycelium may be replaced to the fruit body in due time [304, 357]. All these facts prove that the above-discussed assumptions and estimations regarding the contribution of fungi to caesium retention are reasonable, especially taking into account that the calculations were made using the average (not the maximum reported) concentrations of ^{137}Cs in the fruit bodies of various species.

Table 87. The proportions of the light and dark mycelium in the investigated forest soils, %

Horizon	Pine forest (K–2)		Mixed, broad–leaved–pine forest (Sh–1)	
	Light mycelium	Dark mycelium	Light mycelium	Dark mycelium
A0l	70.8	29.2	54.7	45.3
A0f	56.4	43.6	56.3	43.7
A0h	52.9	47.1	58.4	41.6
$AE_{podzolic}$	50.4	49.6	80.4	19.6
A0+AE	57.8	42.2	61.0	39.0

Microbial biomass in the soil exhibits large seasonal and multiyear fluctuations, which indicates a high intensity and reactivity of the production processes. For example, the mycelium biomass may vary by a factor of 2–3 during a growing season [185], which makes it difficult to estimate the corresponding annual production and a die back of the mycelium. We assumed the annual mycelium production to be equal to 50 % of the total fungus biomass. This value was taken using the following considerations. A range of publications suggests that 50 % to 90 % of the mycelium regenerate every year [17, 86, 185]. The proportion of the so-called "light" (actively regenerating) mycelium in the investigated sites is about 60 % of the total mycelium biomass [86] (Table 87). The average life cycle of mycelium in the soil is about two years [327]. Thus, the assumed values reflect the minimum annual production and die back (return) of mycelium the soil.

We would like to emphasise that the above-discussed contribution of various ecosystem components to the biological cycle of ^{137}Cs is based on "a conservative scenario", i.e., it assumes the minimum estimated values of the biomass and ^{137}Cs content in each ecosystem component.

Conceptual model of ^{137}Cs migration in forest ecosystems

The model consists of two main units, namely soil and biota, which are subdivided into more specific subunits (Fig. 96, 97). The contribution of each unit to the total contamination of biogeocenosis changes with time. It will be remembered that about 90 % of the total deposition were initially retained in the overstorey, and (at least formally) belonged to biota during several months after the accident. Currently, from 6.5 to 43.9 % of the total deposition in forest environments are attributed to biota depending on ecosystem and landscape conditions. Taking into account that these radionuclides are involved in the biological cycle, the contribution of biota to the radionuclide retention is at least as high as it was in 1986. Therewith, the role of the tree canopy decreases in spite of its considerable phytomass, while the contribution of the understorey (especially the moss cover) and mycobiota increases. The moss cover accumulates from 0.08 to 5.85 % of the total deposition. In some cases, it is comparable with or

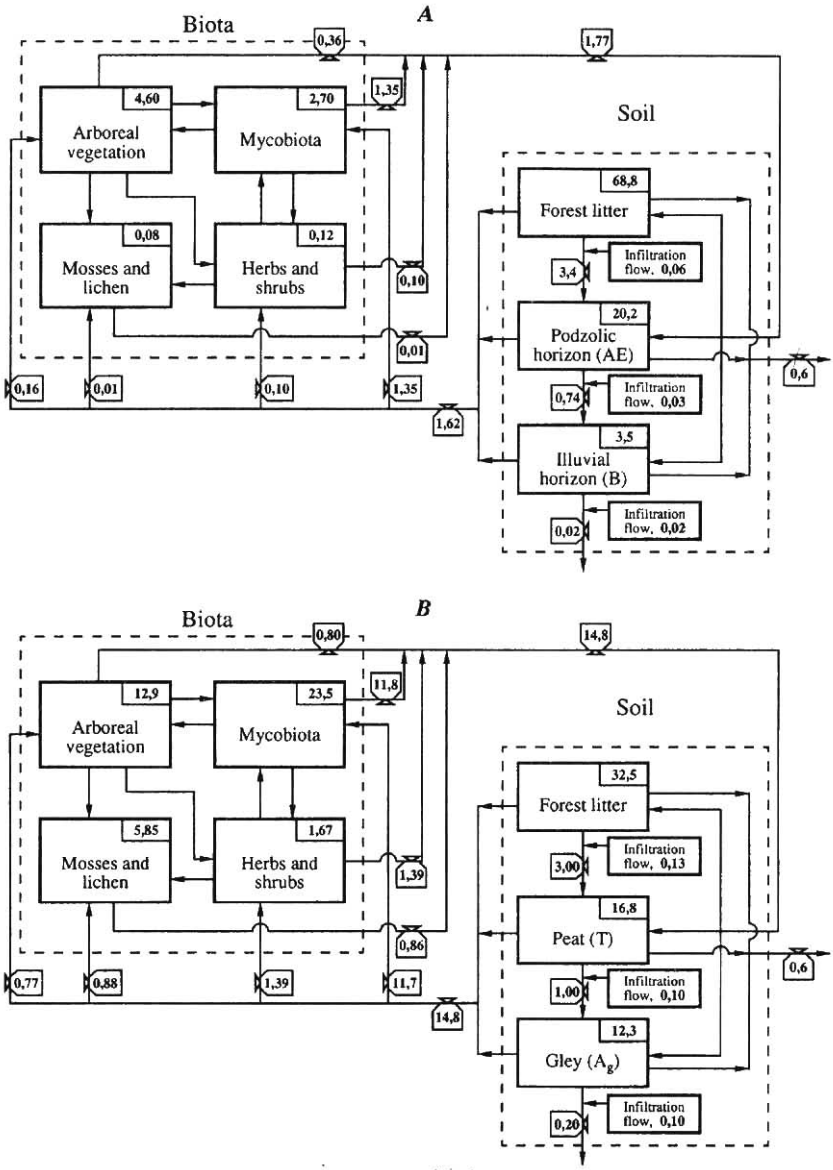

Fig. 96. Biogeochemical cycle of ^{137}Cs in forest ecosystems (the marginal exclusion zone, annual flux is expressed as a per cent of total deposition):

(A) eluvial landscape, mixed forest on soddy-podzolic soil; (B) accumulative landscape, alder forest on peat-gley soil

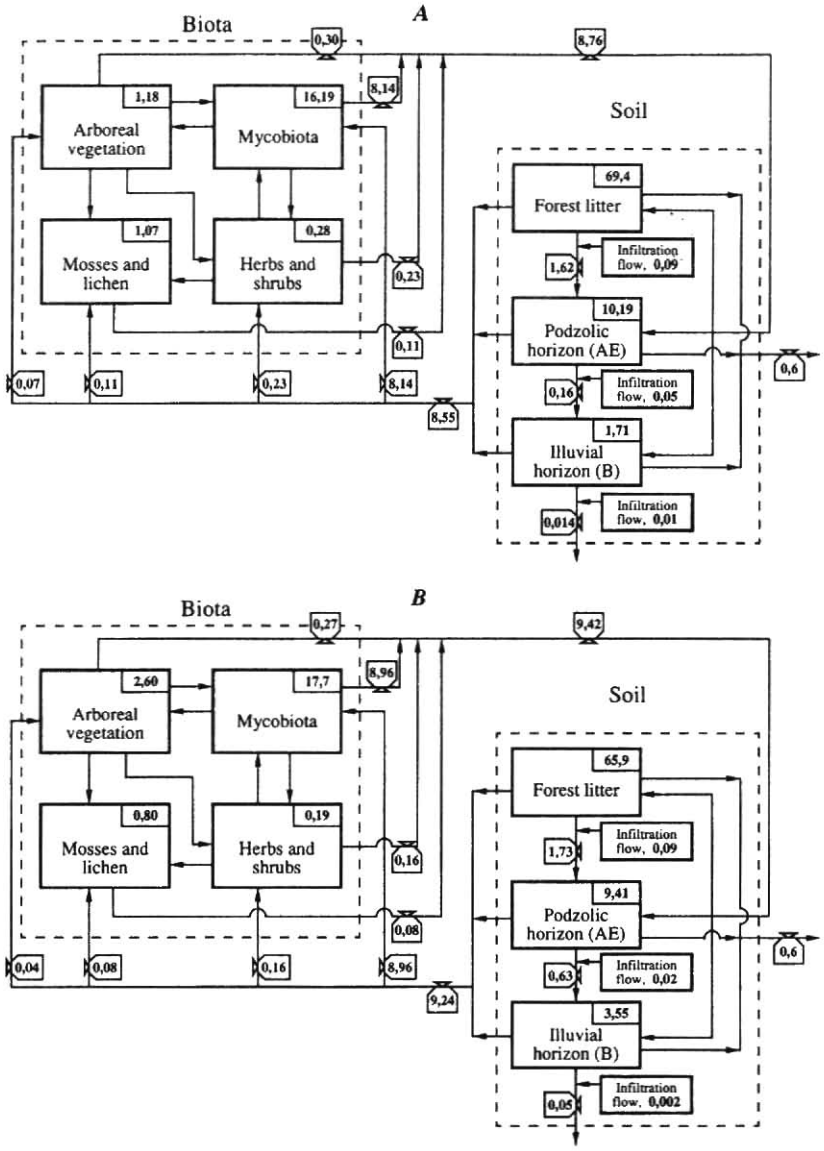

Fig. 97. Biogeochemical cycle of ^{137}Cs in forest ecosystems (the exclusion zone, eluvial landscapes):

(A) pine forest on podzolic soil; (B) mixed forest on soddy-podzolic soil (annual flux is expressed as a per cent of total deposition)

even exceeds (in hydromorphic areas) the contribution of the overstorey. The corresponding variation suggested by other authors is even higher: from 1 to 12 % of the total deposition [133, 193]. In the biota unit, the highest contribution to the radionuclide inventory (2.7–23.5 %) is made by mycobiota. The upper limit of this variation range seems to be unbelievably high and deserves to be discussed in more detail.

In spite of the fact that fungi are known to be effective concentrators of radionuclides, their contribution to the radionuclide pattern in the contaminated forests was neglected because of the presumed low biomass (based apparently on the estimation of the above-ground biomass only) [89]. The suggested values are based on estimation of the total fungus biomass (including the underground mycelium) and experimental data on ^{137}Cs accumulation in the mycelium (10–63 % of deposition) [294, 312, 335].

The contribution of herbs and shrubs to ^{137}Cs accumulation is the lowest of all components of biota (maximum 1.7 %).

Thus, the components of biota may be ranked by their capacity for caesium accumulation as follows: mycobiota > mosses > tree canopy > > herbaceous vegetation (shrubs). The contribution of mycobiota depends on both landscape and ecosystem factors and increases in the range: automorphic areas > hydromorphic areas > pine forests. Mycobiota is, therefore, the most probable factor of radiocaesium retention by forest litter, which is particularly true for pine forests.

Analysing the relative contribution of various ecosystem fluxes to ^{137}Cs redistribution among the forest components, particular attention should be paid to the "coming-return" ratio of the radionuclides to and from various ecosystem components depending on the landscape conditions. In the accumulative landscapes (normally, hydromorphic areas), ^{137}Cs influx to the above-ground vegetation via the root uptake is approximately the same as its return to the soil with litterfall and throughfall. In the eluvial landscapes (normally, automorphic areas), the radionuclide return to the soil currently exceeds its influx to the vegetation by a factor of 2–5. This seemingly paradoxical conclusion is, however, in a good agreement with the long-term dynamics of ^{137}Cs in the overstorey. $TF^{137}Cs$ exhibits a pronounced tendency to decrease in the eluvial landscapes and to increase in the accumulative landscapes. It follows that $TF^{137}Cs$ and the "coming-return" ratio are closely related, i.e., the ratio increases as the $TF^{137}Cs$ decreases, because of an increased contribution of the surface contamination to the total radionuclide inventory in the overstorey. The only exclusion is the area adjacent to ChNPP (the near zone) where the long-term dynamics of ^{137}Cs influx to the tree canopy lags considerably behind its return, in spite of a clear increase in transfer factor and caesium inventory in arboreal vegetation. The reason of this phenomenon is not completely clear yet, and an explanation may be such that the transfer factors in the near zone are still very low compared to other areas [278].

The above-discussed features of ^{137}Cs cycle in the forest ecosystems make it considerably different from the cycles of nutrient and other stable elements. Firstly, radionuclides are initially accumulated in the external components of the plant exposed to the fallout. Therefore, surface decontamination dominates over the root uptake for a long time after the fallout. Secondly, the amount of

radionuclides added to the ecosystem is, as a rule, limited, and the pool of available radiocaesium in the soil decreases in time because of its permanent irreversible adsorption. Unlike stable natural elements, the available radionuclide inventory cannot be resupplied at the expense of their potential reserves in the soil minerals and organic matter.

Thus, in eluvial forest landscapes, return of ^{137}Cs to the soil dominates over its root uptake, whereas in the accumulative landscapes, the "coming/return" ratio is close to 1, which is characteristic of stable elements as well.

Throughfall and stem flow do not contribute significantly to ^{137}Cs cycling in the ecosystem as a whole (the corresponding annual flux constitutes about 0.05 % of the total deposition), but may be of importance in some cases. For example, the above value is comparable with an annual rate of ^{137}Cs infiltration from the forest litter and (in the remote zone) an annual root uptake to the overstorey.

In the soil unit, the main flux of ^{137}Cs (1.6–3.4 % per year) occurs from the forest litter to the topmost mineral layer. Down the soil profile, the radionuclide flux becomes as small as $n \cdot 0.1$or $n \cdot 0.01$ % of the total deposition, i.e., almost all radionuclides which left the forest litter are accumulated in the upper few centimetres of the mineral soil. No more than hundredths of a per cent of the total deposition leave annually the soil layer of 0.5 m. The only exclusion is hydromorphic soils of accumulative landscapes where the infiltration flow is most manifested and almost stable down the soil profile. These soils exhibit the maximum relative loss of radiocaesium from the profile, and in these environments the radionuclide influx to the ground water is most pronounced. It means that the role of the so-called "fast component" in ^{137}Cs vertical migration is much more manifested in the peat soils compared to soddy-podzolic soils [223]. Comparison of actual distribution of radionuclides down the soil profile with the infiltration rate suggests that the contribution of infiltration to the radionuclide vertical migration and redistribution is insignificant in the upper 20–30 cm soil layer, but is of a higher importance for deeper layers.

In general, the rate of the radionuclide involvement in the biological cycle is comparable with its annual loss from the soil thickness. It means that the biological cycle is a powerful factor preventing radionuclide from intensive migration to the ground water. In accumulative landscapes, the effect of biota on ^{137}Cs accumulation and migration increases almost tenfold. This is due to low capacity of organic, peat-bog soils for caesium irreversible adsorption, high TF, and long-term transport of ^{137}Cs from neighbouring areas by the lateral intrasoil flow and surface transport. The total annual increment of ^{137}Cs in the accumulative landscapes due to its large-scale lateral migration normally does not exceed 1 % per year.

SUMMARY

Forest ecosystems subjected to aerial contamination (radioactive fallout) serve as a long-term sink for radionuclides, which effectively slows down further radionuclide migration in the environment. The long-term radionuclide dynamics in the tree canopy is subdivided into three principal periods: (1) intensive mechanical decontamination (1–3 months); (2) biological decontamination and increasing root uptake (2 to 3 years); (3) prevalent root uptake and gradual approach to quasi-equilibrium in the "soil-plant" system (3–10 years and longer).

On the automorphic, particularly, clay-rich soils, contamination of the tree canopy is determined primarily by the tree organs exposed to the initial fallout (such as external bark). On hydromorphic and semi-hydromorphic soils, and in the close vicinity of the accidental unit where the fallout was enriched with coarse, low soluble fuel particles, the main contribution to the contamination is made by wood, internal bark, and assimilative organs (leaves and needles).

The transfer factors of ^{137}Cs and ^{90}Sr ($TF^{137}Cs$ and $TF^{90}Sr$) to the tree organs and components vary by 2–3 orders of magnitude. The range of variation depends on the survey scale as follows: ecosystem (biogeocenosis) < natural climatic zone < entire contaminated territory. In general, the dynamics of $TF^{137}Cs$ to forest vegetation can be classified into 3 basic categories: (1) a pronounced trend to decrease (in eluvial areas); (2) a pronounced trend to increase (accumulative areas and the near zone); (3) unpronounced trend (transit-accumulative areas and the middle part of the exclusion zone). The accumulative capacity of basic ecosystem components for ^{137}Cs is expressed by the following series: fungi complex > mosses and lichen > arboreal vegetation > herbs and shrubs. Biological availability of radionuclides and their accumulation by plants depend on (i) soil type and moisture regime; (ii) stand age; (iii) species composition; (iv) fallout properties. Significance of the latter factor (fallout properties) increases dramatically in the close vicinity of ChNPP (the near zone). The effect of all factors on the biological availability of ^{90}Sr is less manifested compared to ^{137}Cs..

Spatial distribution of radionuclides is characterised by a set of specific features. The radionuclide distribution in soils has a pronounced spot-like character that increases on approaching the accidental unit. On the average, the variation of the radionuclide content in soil is about 30 %, which is similar to other technogenic pollutants. The variation of the radionuclide content in arboreal vegetation is twice as much as in soil, because of different contamination of var-

ious tree organs. In this connection, leaves and needles of the current year are the most appropriate components for monitoring ^{137}Cs dynamics in the stand, while the external bark and "old" needles are most appropriate for monitoring of the behaviour ^{90}Sr.

In general, forest soils in temperate climatic zones (such as the central part of the East-European Plain) provide favourable conditions for radionuclide migration. These soils (except for chernozems) are characterised by a low pH, a low base saturation, and a low humus content and exchange capacity. Mineralogical composition of these soils is poor of weatherable materials and clay minerals. All these features result in a low soil capacity for both exchangeable and irreversible sorption of ^{137}Cs except for the forest litter and a thin (1–2 cm) mineral layer beneath it.

Forest litter serves as the most important biogeochemical barrier that considerably slows down the vertical migration of radionuclides. The sorption capacity of forest litter depends on its thickness and structure. Moss cover increases the retention capacity. Soil mycobiota (fungi complex) is among the key factors determining the soil sorption capacity for ^{137}Cs.

The amount of radionuclides that penetrate into the mineral soil layers beneath the forest litter horizon is the reciprocal of the sorption capacity of the forest litter. The vertical redistribution of radionuclides in soil depends on the soil properties and processes, and (to a lesser degree) the features of the initial fallout. The intensity of radionuclide migration through the mineral soil layers depends on the soil type as follows: hydromorphic forest soils > hydromorphic alluvial meadow soils > automorphic forest soils > automorphic old-tilled soils.

The radionuclide content in the topmost layer of the forest litter (A0l) depends primarily on (i) the annual input of relatively "clean" litter, and (ii) the intensity of the organic matter decomposition. In the other layers of the forest litter and in mineral horizons, the radionuclide content depends on the intensity of various migration processes, i.e., diffusion, infiltration with water, and biogenic migration. In the automorphic soils, the radionuclide migration is most manifested within the limited "active migration zones" (attributed, as a rule, to microdepressions), and the boundary of the radionuclide distribution down the soil profile has a tongue-like shape. In hydromorphic (particularly, peat) soils, the radionuclide migration runs more uniformly, and the boundary of the significant penetration of radionuclides has a "frontal" shape.

The total content of potentially mobile radionuclides in the investigated forest soils does not exceed 5 % of the deposition. The composition of the liquid soil phase is represented by a full radionuclide spectrum, which is, however, not identical to the radionuclide composition of the soil as a whole. The main part of radionuclides in soil solutions is represented by radionuclide-organic compounds. $^{239+240}Pu$ (^{238}Pu) and ^{241}Am are associated primarily with high-molecular fractions of a soluble organic matter (MMw > 2000 Da). The highest proportion of ^{137}Cs is associated with relatively low-molecular organic compounds (MMw > 1000 Da), and ^{90}Sr is associated with the least massive fractions (MMw = 350–500 Da). The radionuclide mobility in the soil liquid phase decreases as follows: $^{90}Sr >> ^{106}Ru > ^{134,137}Cs > ^{239+240}Pu$ (^{238}Pu) $> ^{144}Ce$.

The contribution of various components of biota to the total deposition changes with time. At the present (quasi-equilibrium) stage, when the radionuclide pattern in the "soil-plant" system depends primarily on the root uptake, biota may store from 6.5 to 44 % of total deposition in the biogeocenosis. The contribution of arboreal vegetation (in spite of its dominant biomass) has decreased dramatically since 1986, while the contribution of the moss cover and especially, mycobiota has increased considerably (from 0.08 to 5.85 % and from 2.7 to 23,5 %, respectively).

Annual biogeochemical fluxes of ^{137}Cs in forest ecosystems are characterised by a set of specific features. In the eluvial landscapes, an annual return of ^{137}Cs to the soil with the litterfall, throughfall, and stem flow still exceeds its annual accumulation, and is inversely proportional to $TF^{137}Cs$. In the accumulative landscapes, the "influx" is approximately the same as the "outflux", and both are close to the corresponding fluxes of stable macro-carriers (nutrients). The throughfall and stem flow make a negligible, though measurable, contribution to the ecosystem cycle of ^{137}Cs (about 0.05 % of the total deposition). The most intensive soil flux of ^{137}Cs is from the forest litter to upper mineral horizons (1.6–3.4 % per year). In the mineral layers, the corresponding rate is tenths and hundredths of a per cent. The radionuclide transport with the infiltration flow has a negligible effect on the final radionuclide distribution in the upper soil layers, but the radionuclide penetration into the deeper layers (below 20–30 cm) may be due to this mechanism. Annual rate of ^{137}Cs and ^{90}Sr involvement in the biological cycle is much higher than the annual loss of these radionuclides from the biogeocenosis (vertical or lateral); the rate of biogenic uptake of radionuclides is much higher in accumulative landscapes compared to eluvial ones.

The large-scale lateral redistribution of radionuclides in the system of adjacent, geochemically linked landscapes is hardly observable even 10 years after the accident. It does not exceed 1 % per year for ^{137}Cs, and is somewhat higher for ^{90}Sr. Lateral radionuclide migration is, however, well manifested at the scale of mesotopography (meters to tens meters) and microtopography (centimetres to meters).

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