Radiat Environ Biophys (2005) 44: 169–181
DOI 10.1007/s00411-005-0015-2

ORIGINAL PAPER

N. I. Ryabokon · I. I. Smolich · V. P. Kudryashov
R. I. Goncharova

Long-term development of the radionuclide exposure of murine rodent populations in Belarus after the Chernobyl accident

Received: 7 February 2005 / Accepted: 9 September 2005 / Published online: 8 October 2005

Abstract As a determinant of the associated health risks, the behavior of radionuclides in natural ecosystems needs to be better understood. Therefore, the activity concentration of various long-lived radionuclides released due to the Chernobyl accident, and the corresponding contributions to the whole-body dose rate, was studied as a function of time in mammalian indicator species inhabiting the natural forest ecosystems of Belarus, the bank vole (*Clethrionomys glareolus*) and the yellow-necked mouse (*Apodemus flavicollus*). The activity concentrations of ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{238}Pu, ^{239,240}Pu, ^{241}Pu and ^{241}Am in soil and in animals were measured at five monitoring sites with different ground deposition of radionuclides at different distances from the destroyed reactor. The observed temporal pattern of the radionuclide activity concentration in the studied animal populations reflects the changes in biological availability of these isotopes for biota, mostly due to fuel particle destruction and appearance of dissolved and exchangeable forms of radionuclides. The time course of $^{134+137}$Cs activity concentrations in animal populations appeared as a sequence of increase, peak and decrease. Maximal levels of radiocesium occurred 1–2 years after deposition, followed by an exponential decrease. Concentrations of incorporated ^{90}Sr increased up to the tenth year after deposition. The activity concentrations of transuranic elements (^{238}Pu, ^{239,240}Pu, ^{241}Pu and ^{241}Am) were much lower than those of the other radionuclides, in the studied animals. A considerable activity of ^{241}Am in animals from areas with high levels of contamination was firstly detected 5 years after deposition, it increased up to the tenth year and is expected to increase further in the future. Maximal values of the whole-body absorbed dose rates occurred during the year of deposition, followed by a decrease in the subsequent period. Generally, this decrease was monotonic, mainly determined by the decrease of the external γ-ray dose rate, but there were exceptions due to the delayed maximum of internal exposure. The inter-individual distributions of radionuclide concentrations and lifetime whole-body absorbed doses were asymmetric and close to log-normal, including concentrations and doses considerably higher than the population mean values.

N. I. Ryabokon (✉) · I. I. Smolich · R. I. Goncharova
Institute of Genetics and Cytology, National Academy of Sciences of Belarus, 220090 Minsk, Republic of Belarus
E-mail: nrabakon@yahoo.com

V. P. Kudryashov
Institute of Radiobiology,
National Academy of Sciences of Belarus,
Gomel, Republic of Belarus

Introduction

The Chernobyl accident gave rise to a widespread contamination of land throughout Europe by various radionuclides, including long-lived ones. Subsequently, the distribution pattern of the radionuclides in soil changed due to various factors, such as the destruction of nuclear fuel particles and the physical decay of the radionuclides involved, their migration in soil and their physicochemical reactions with soil components. The changeable composition and concentration of radionuclides in different components of the biota affected the level and type of radiation exposure. For example, due to the decay of ^{241}Pu, the content of the soluble ^{241}Am in the soil is increasing and is predicted to be maximal in the year 2058 [1]. Thus, the accumulation of ^{241}Am in plants via root uptake may elevate the concentrations of this element in trophic chains and, in this way, increase the radiation risk [2].

It is therefore important to investigate the main peculiarities of radionuclide behavior and dose accumulation in different components of biota, in order to support the long-term forecast of the radioecological situation and to conduct protective countermeasures in areas suffering from the Chernobyl accident or from radiation contamination of any other origin. Such investigations should be primarily carried out in

populations of plants and animals, which are used as biological indicators of environmental quality and may represent different components of trophic chains.

Among wildlife species, murine rodents are known as indicators or "sentinels" representing the herbivorous group of the trophic chains. Radioecological investigations in natural rodent populations have shown their usefulness not only for the indication of low radio active contamination of the environment [3, 4], but also for revealing the time behavior of radionuclides from global fallout [5]. Our previous research has allowed us to detect increased levels of ^{137}Cs and ^{134}Cs in populations of wild rodents inhabiting forest ecosystems in regions of Belarus with low levels of contamination due to the Chernobyl accident, and to find some common features of cesium dynamics in populations at different levels of radionuclide contamination [6]. These observations and the data of our further assessment of the radioecological situation in rodent populations, now extended to the other relevant radionuclides, are summarized in this paper. The overall aim of our research has been to study the time course of the concentrations of various long-lived radionuclides, their contributions to whole-body dose rates and to lifetime whole-body doses, in populations of murine rodents inhabiting forest ecosystems with different ground deposition of radionuclides after the Chernobyl accident. This study is part of a complex radioecological and genetic monitoring work carried out in contaminated areas since the accident in the Chernobyl nuclear power plant (ChNPP) in April 1986 [6].

Materials and methods

Monitoring sites and animal trapping

Five monitoring sites were selected throughout Belarus in regions located at different distances and directions from the ChNPP:

- Site 1: the Minsk region, 330 km NW (Priluksky reserve);
- Site 2: the Vitebsk region, about 400 km NNW (Berezinsky biosphere reserve);
- Site 3: the vicinity of the Majsk village, Bragin district, Gomel region, 60 km N;
- Site 4: the vicinity of the Babchin village, Khoiniki district, Gomel region, 40 km NNW, exclusion zone;
- Site 5: the vicinity of the Radin village, Khoiniki district, Gomel region, about 18 km N, exclusion zone.All sites are located in regions with restricted people activity, far from settlements, roads and possible sources of industrial or domestic pollution, in the large forestry areas typical of Belarus. They show very similar natural biotopes of mixed forests including pine-trees and deciduous species, as well as rich undergrowth.

Investigations at sites 1, 3 and 4 were started in the year of the accident (1986); at sites 2 and 5 they began in 1991 and 1996, i.e. 5 and 10 years later, respectively. The rodents were usually captured in August and September using live traps. The first animals were captured 4 months after the accident, i.e. after the period of acute irradiation by short- and long-lived radionuclides. Two indicator species were selected to represent the herbivorous species typical for the ecosystems studied: the bank vole (*Clethrionomys glareolus, Schreber*), known for its dominant population density among small mammals at the monitoring sites, and the yellow-necked mouse (*Apodemus flavicollus, Melchior*), known for its subdominant density. The animals were killed by ethyl ether and cervical dislocation and frozen at −20°C after sampling small pieces of tissues for genetic tests. By the end of this study, about 1,500 specimens of bank vole and yellow-necked mice had been trapped and analyzed.

Assessment of radionuclide deposition in soil and animal bodies

Samples of soil were collected at the trapping sites using two standard sampling cylinders made of steel (one with a diameter of 150 mm and a length of 50 mm, the other with a diameter of 45 mm and a length of 200 mm), following the protocol of the Hydrometeorological Centre of Belarus, Minsk. The ^{134}Cs, ^{137}Cs, ^{106}Ru and ^{144}Ce activities of the soil samples were detected with the gamma-spectrometer ADCAM-300, equipped with a high-purity germanium (HPGe) detector GEM-30185 (EG&G Ortec, USA), by the staff of the Hydrometeorological Centre of Belarus.

Whole-body measurements of animals captured in 1986–1988 were performed using the gamma-coincidence spectrometer ARGUS (32-crystal NaI(Tl) detectors of 150×100 mm^2) developed at the Institute of Physics, National Academy of Sciences of Belarus, Minsk, which allows the total γ-activity of samples to be measured with high sensitivity. In the range of detected energies 0.2–3.0 MeV, the detection limit was 1 Bq [7]. Since 1988, the activity concentrations of radionuclides in animal samples were analyzed using the ADCAM-300 gamma-spectrometer. Good agreement between total γ-activities measured by the two different spectrometers was obtained, in animals captured in 1988. Generally, for all radionuclides the detection time was chosen large enough so that the uncertainty of the measured activity did not exceed 30%. Radionuclides for which this was not possible were not taken into consideration.

Radiochemical analysis of soil and animal samples was applied in order to determine the transuranic elements (TUE) and ^{90}Sr contents. To increase the accuracy of the analysis of TUE in animals, three or six animals of the same sex, age and maturation stage were included into the samples from the sites 2–4. All samples were homogenized, ashed in a muffle furnace at 550 ± 50°C and dissolved completely using a mixture of

concentrated HNO_3 and H_2O_2. Radioactive ^{242}Pu and ^{252}Cf carriers, and the ^{85}Sr-tracer were added to the samples. The main steps of the radiochemical analysis were based on co-precipitation of strontium, plutonium and americium isotopes as oxalates, oxidation of oxalates, followed by co-precipitation of $^{238,239+240}Pu$ and ^{241}Am with iron hydroxide [8, 9], and isolation of $^{238,239+240}Pu$ and ^{241}Am by ion-exchange methods using AV-17 anionite and KU-2 cationite. The precipitates containing TUE were filtered under vacuum on a membrane filter (hole diameter 0.15 mm) and counted by α-spectrometry with the silicon surface barrier detector Ortec 576A (EG&G Ortec, USA). The activity of ^{90}Sr in the samples was determined by measurements of the daughter radionuclide ^{90}Y. For this, ^{90}Y was permitted to grow in the rest of the sample solution by use of yttrium nitrate, and was then separated as hydroxide and finally transferred as an oxalate on to stainless steel disks, and measured by a gas-flow counter (Tesla Automat, Slovakia). ^{241}Pu was detected with the liquid scintillation counter Tricarb 2700TR (Packard Company). The total uncertainty did not exceed 25% for all radionuclides measured.

Dosimetry

Whole-body absorbed dose rates due to the chronic exposure to external γ- and internal α-, β- and γ-radiation of incorporated radionuclides were estimated following a handbook of Moiseev and Ivanov [10]. For this purpose, local measurements of external γ-dose rates at a height of 3–5 cm above the ground surface were performed using the dosimeters SRP-68-01 (including a NaI(Tl) detector) and DBG-06T (including a Geiger counter) manufactured in the Former Soviet Union. These instruments were calibrated to measure exposure rates in mR/h (1 $R = 2.58 \times 10^{-4}$ C/kg of air). For the conversion to absorbed dose rate in soft tissues of small animals at photon energies of about 0.5–3 MeV, we used the factor 9.65 μGy/mR [11]. It is assumed that the animals were uniformly irradiated on the ground surface and in the holes in the upper soil layers.

Individual values of incorporated radionuclide activity concentrations were used for the assessment of internal irradiation in captured specimens of bank vole and yellow-necked mice. In 1986–1987, when spectral analyses of animal samples were not available, the data on the total γ-activity of incorporated radionuclides were used for the assessment of absorbed dose rate. We assumed that short-lived γ-emitting radionuclides did not give significant contributions to the irradiation of animals captured 5 months after the accident or later and that ^{137}Cs and ^{134}Cs were the prevailing γ-emitters contributing to the total γ-activity of animal samples. Thereafter we used the fact that the ratio of the activity concentrations of ^{137}Cs and ^{134}Cs in the animals is approximately equal to the well-known ratio of these concentrations in the soil at the same time. This was used as the basis for the assessment of the contributions of these isotopes to the measured total γ-activity concentration in the animals.

The whole-body absorbed dose rate from incorporated γ-emitting radionuclides was calculated according to the absorbed fraction model [12] under the assumption that the emitters were uniformly distributed in a cylindrical body of 3 cm diameter and 5 cm length having the density of skeletal muscle (1 g/cm^3) [13]. For the whole-body absorbed dose rate attributable to incorporated α- and β-emitters we used the local absorption model [12], applying the average particle energies according to the International Commission on Radiological Protection [14]. As strontium is known to be mostly accumulated in bone tissue, the absorbed dose rate in the bone tissue of the animals was estimated using the known approximate value of 6.5 for the ratio of the ^{90}Sr activity concentrations accumulated in bone tissue and in the whole-body, as observed in voles inhabiting areas contaminated by the Chernobyl accident [15] and the radioactive trace in the Urals [16].

The "lifetime whole-body absorbed dose", i.e. the product of the individual whole-body absorbed dose rate and the age of the animal at the time of capture, was also calculated for every animal. For age determination, the root, cusp and height of the first mandibular molar (M_2) were measured according to N.V. Bashenina [17]. The specimens were divided into seven age groups, which corresponded to ages of approximately 2 weeks, 1, 2, 3 and 4 months and 1 and 1.5 years, respectively.

Statistical evaluation

Several statistical methods were employed—such as the Chi-square, Kolmogorov-Smirnov and *U*-tests, as well as regression and correlation analyses. These algorithms were part of the STATISTICA software package (StatSoft Inc., USA).

Results

Soil contamination

Before the accident, the contamination of soil by ^{137}Cs and ^{90}Sr due to nuclear weapons tests had been in the range of 1.5–3.7 and 0.74–2.59 kBq/m^2 in Belarus, respectively [18, 19]. The contamination by $^{238,239,240}Pu$ had been in the level of 0.037–0.059 kBq/m^2 [20]. The densities of contamination with radionuclides released from the destroyed Chernobyl reactor and detected in soil at the monitoring sites are presented in Table 1. All sites show increasing levels of contamination in comparison with pre-accident data, with decreasing distance from the ChNPP.

Table 1 Radionuclide contamination of soil (kBq/m^2) at five monitoring sites

Site	Number of samples analyzed	April–May 1986				August 1996				
		^{137}Cs	^{134}Cs	^{106}Ru	^{144}Ce	^{90}Sr	^{238}Pu	^{239,240}Pu	^{241}Pu	^{241}Am
1	5	8 ± 0.9	4 ± 0.6	5 ± 0.6	0	3.81 ± 0.26	0.04 ± 0.01	0.10 ± 0.01	2.98 ± 0.37	0.14 ± 0.02
2	5	18 ± 1.4	9 ± 0.9	12 ± 1.1	0	5.12 ± 0.46	0.07 ± 0.01	0.14 ± 0.02	5.10 ± 0.53	0.19 ± 0.02
3	5	220 ± 18	140 ± 11	150 ± 12	440 ± 37	38.6 ± 3.3	0.62 ± 0.07	1.28 ± 0.13	48.8 ± 5.1	1.81 ± 0.17
4	5	1,530 ± 240	1,020 ± 241	1,090 ± 172	3,050 ± 485	117 ± 7	1.17 ± 0.07	2.35 ± 0.09	86.7 ± 4.5	3.21 ± 0.23
5	5	8,500 ± 1,340	5,650 ± 879	5,790 ± 1,023	17,200 ± 3,105	1,200 ± 72	4.90 ± 0.69	10.66 ± 1.17	420 ± 46	15.00 ± 1.79

Data of April–May 1986 for 137Cs, 134Cs, 106Ru, 144Ce; data as of August 1996 for 90Sr and transuranic radionuclides. Mean values and standard deviations are shown

External radiation monitoring

The time course of the γ-dose rate at the monitoring sites is presented in Fig. 1. After a fast decrease in the first days after the accident (Fig. 1a), the dose rate decreased more gradually, and at the weakly contaminated sites 1 and 2 closely approached the background level of 8–9 μR/h [21] by the tenth year after radionuclide deposition (Fig. 1b, c). The time dependence of the γ-dose rate can be approximated by power functions.

Incorporated cesium isotopes

Among the long-lived radionuclides, which were of principal interest, ^{137}Cs and ^{134}Cs attracted special attention because of their high concentrations in the fallout released from the destroyed reactor. Other isotopes found in soil samples, i.e. ^{106}Ru (half-life 1.02 years) and ^{144}Ce (half-life 284.9 days), showed very low concentrations with high uncertainties in animal bodies 2 years after the deposition, and were difficult to be detected later.

Since the second post-accident year, the total γ-activity in the animals was mainly due to ^{137}Cs and ^{134}Cs. By the tenth year of investigation, ^{134}Cs (half-life 2.06 years) had decayed to a large extent, and was therefore not detected in animals trapped in areas with low (sites 1, 2) and moderate contamination (site 3). Thus, ^{137}Cs (half-life 30.0 years) became the main contributor to the total γ-activity of animals in these areas.

Data on the time course of the total activity concentration of incorporated γ-emitting radionuclides in populations of the two studied species are presented in Fig. 2. Considerable levels of γ-activities existed over the whole monitoring period. With the exception of the first year at the least contaminated site 1, the yellow-necked mouse accumulated significantly lower activity concentrations of ^{137}Cs and ^{134}Cs than the bank vole (U-test, $P < 0.01$). In spite of these differences between the two species, common characteristics in the behavior of radiocesium were identified, namely a significant increase in the value of γ-activity concentration during 1 or 2 years after the deposition, a maximum value and a subsequent decrease (Fig. 2). This decrease had an

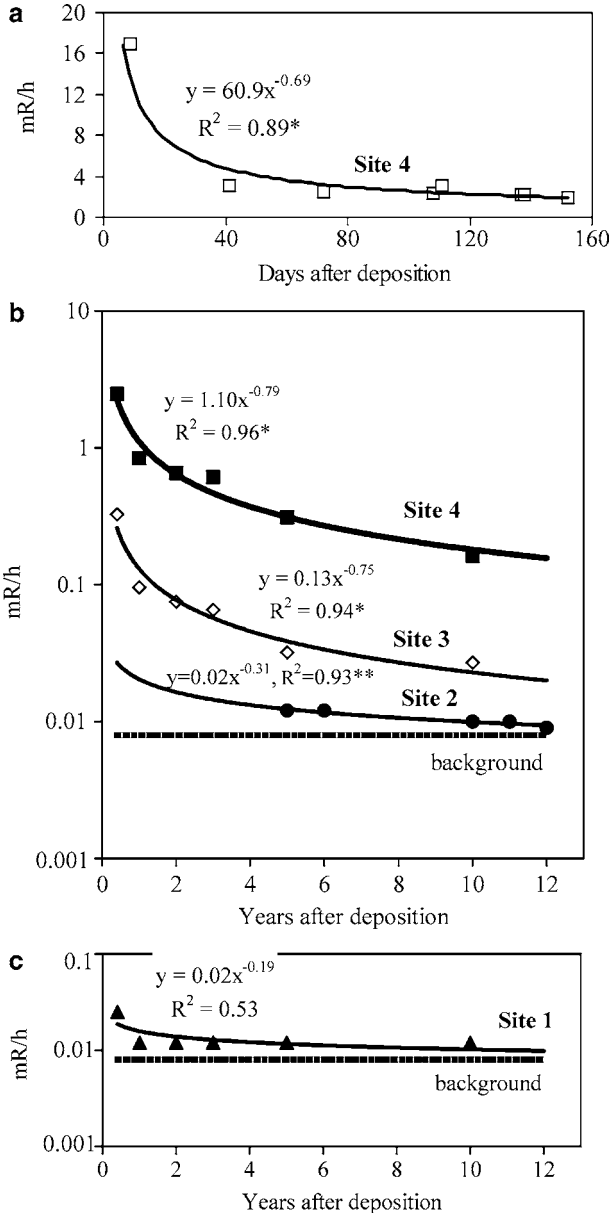

Fig. 1 Time course of the mean external γ-dose rate on the ground surface at sites 1–4 as a function of time after the accident. **a** Short-time behavior, **b** and **c** long-time behavior. Fitting curves were obtained by regression analysis: $*P < 0.05$ and $**P < 0.01$. The background level which existed before the accident is presented according to Hardy et al. [20]

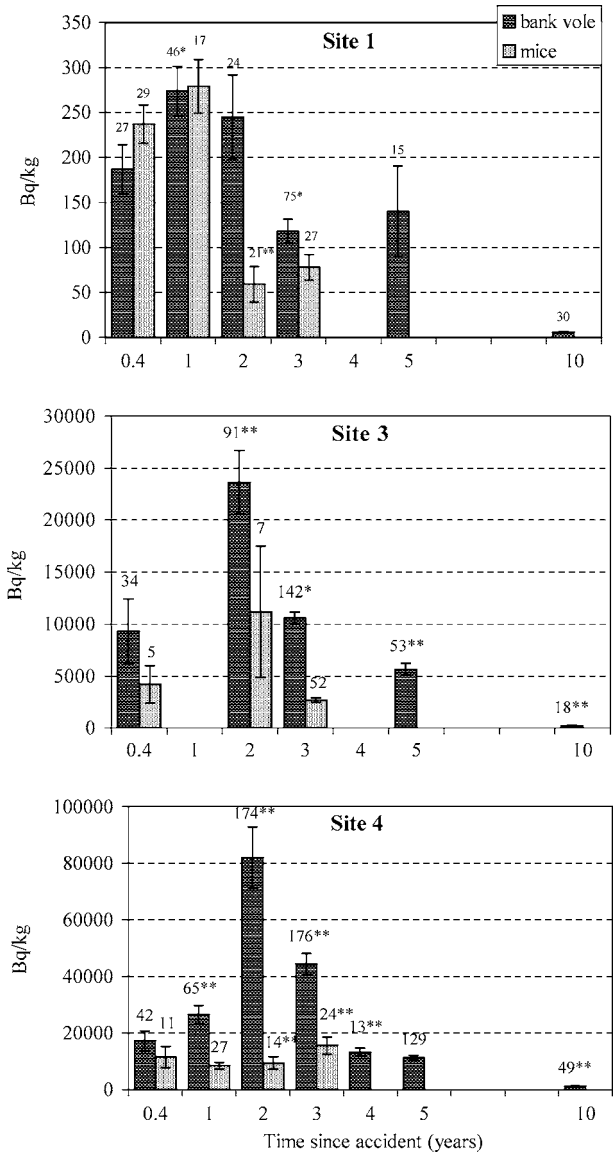

Fig. 2 Time course of mean activity concentration of γ-emitting radionuclides in populations of bank vole and yellow-necked mice, at the investigated monitoring sites. Standard deviations of the mean values are indicated. The *numbers above the bars* represent the number of animals analyzed: $*P<0.05$ and $**P<0.01$ in comparison with previous year (U-test)

approximately exponential shape for ^{137}Cs (Fig. 3a) and ^{134}Cs (Fig. 3b) as observed in bank vole populations.

The activity concentration of radiocesium did not depend on the age of the animals, and it significantly differed between the sites, showing a direct correlation with soil contamination (Fig. 4).

Incorporated ^{90}Sr

In contrast to the cesium isotopes, an increase of the ^{90}Sr activity concentration in bank vole populations occurred later. The concentration of incorporated radiostrontium was approximately doubled (U-test, $P<0.01$) at the contaminated sites in the period from 5 to 10 years after deposition (Fig. 5). With the exception of site 3, the populations at the other monitoring sites had significantly lower activity concentrations of ^{90}Sr in comparison with ^{137}Cs (U-test, $P<0.01$) (Table 2). The activity concentration of ^{90}Sr in the vole populations correlated with the ^{90}Sr contamination of soil (Fig. 4), but did not correlate with the age of the animals.

Incorporated TUE

The activity concentrations of the TUE in animal bodies were much less than those of the other detected radionuclides (Table 2). Like the other long-lived radionuclides studied, the TUE correlated with the levels of soil contamination by these isotopes (Fig. 4). Spectrometric analyses showed that significant amounts of ^{241}Am firstly appeared in some specimens of bank vole in the highly contaminated area 5 years after the deposition. A decade after the accident, ^{241}Am was detected in all populations analysed (Table 2). ^{241}Pu was dominant and its daughter product ^{241}Am was subdominant among TUE accumulated in animals.

Frequency distribution of accumulated radionuclides

High variability in individual concentrations of radionuclides was observed in all populations over the whole monitoring period, sometimes ranging over more than one order of magnitude. In most cases, distributions of individual concentrations were asymmetrical with skewness to the high concentrations (Fig. 6). The underlying type of distribution appears to be log-normal, but in many cases there was only a rough similarity of the observed distributions with this theoretical shape. The high variability in individual concentrations and the non-normal type of distribution requires a large number of animals to be collected and used for statistical analysis. As the density of yellow-necked mouse populations was often very low, we stopped collecting yellow-necked mice by the end of the 3-years' monitoring.

Time course of absorbed dose rate

The whole-body absorbed dose rate in rodents before the accident did not exceed 2 μGy/day and was mostly due to external γ-radiation, as one can derive from the data on rural populations of people living in Belarus and the neighboring Poland at that time [10, 21]. For the time after the accident, whole-body absorbed dose rates from external γ-irradiation and internal α-, β- and γ-irradiation from the Chernobyl fallout, and external exposure from the natural background were considered. External β-contribution, assumed to be mostly self-absorbed, was not taken into consideration.

The whole-body dose rate due to external and internal exposure was found to be maximal in the year of the

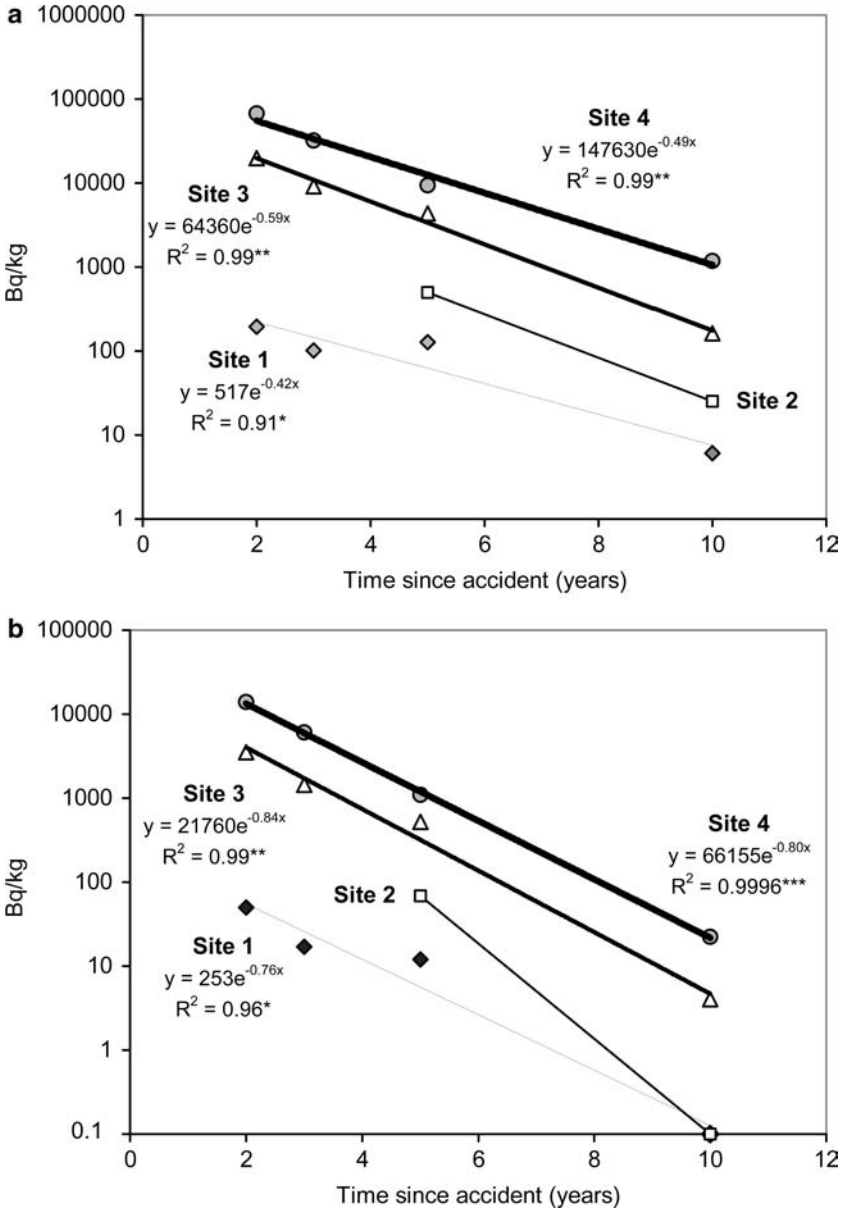

Fig. 3 Time course of mean **a** ^{137}Cs and **b** ^{134}Cs activity concentration in bank vole populations after reaching the maximal concentrations 1–2 years after the accident: $*P < 0.05$, $**P < 0.01$ and $***P < 0.001$

accident. Over the subsequent period it decreased—with some exceptions—in an overall monotonic fashion (Fig. 7). Ten years after the radionuclide deposition, the dose rate was close to but did not reach the background level even in populations inhabiting areas with low contamination (sites 1 and 2). As can be seen in Fig. 7, the populations of bank vole generally had significantly higher doses from external than from internal irradiation over the period of investigation, but with some exceptions. In years when the concentration of incorporated radiocesium reached its peak (Fig. 2), the contribution of internal exposure to the whole-body dose rate became significantly higher than the external one in animals in the areas with moderate and high contamination, and this affected the monotonic decrease of the total absorbed dose rate at site 4 (Fig. 7).

The contribution of incorporated radionuclides to the whole-body dose rate is of particular interest in the remote period after the deposition, when external γ-exposure has significantly decreased and the long-lived isotopes with α- and β-activities remain in the contaminated ecosystems (Table 3). Our estimates show that the internal exposure of animals from incorporated radionuclides 10 years after the accident was in the range of 0.1–18 % of the total dose rate, while the external γ-irradiation contributed 82–99.9 %. The incorporated isotopes of cesium had the maximal part, from 0.1 to 16%, which increased from the less to the higher contaminated sites. ^{90}Sr and the TUE were the next; their contributions to dose rate were 0–8 and 0–3%, respectively. The time course of the dose rate from ^{90}Sr revealed that its contribution to dose rate in bank voles

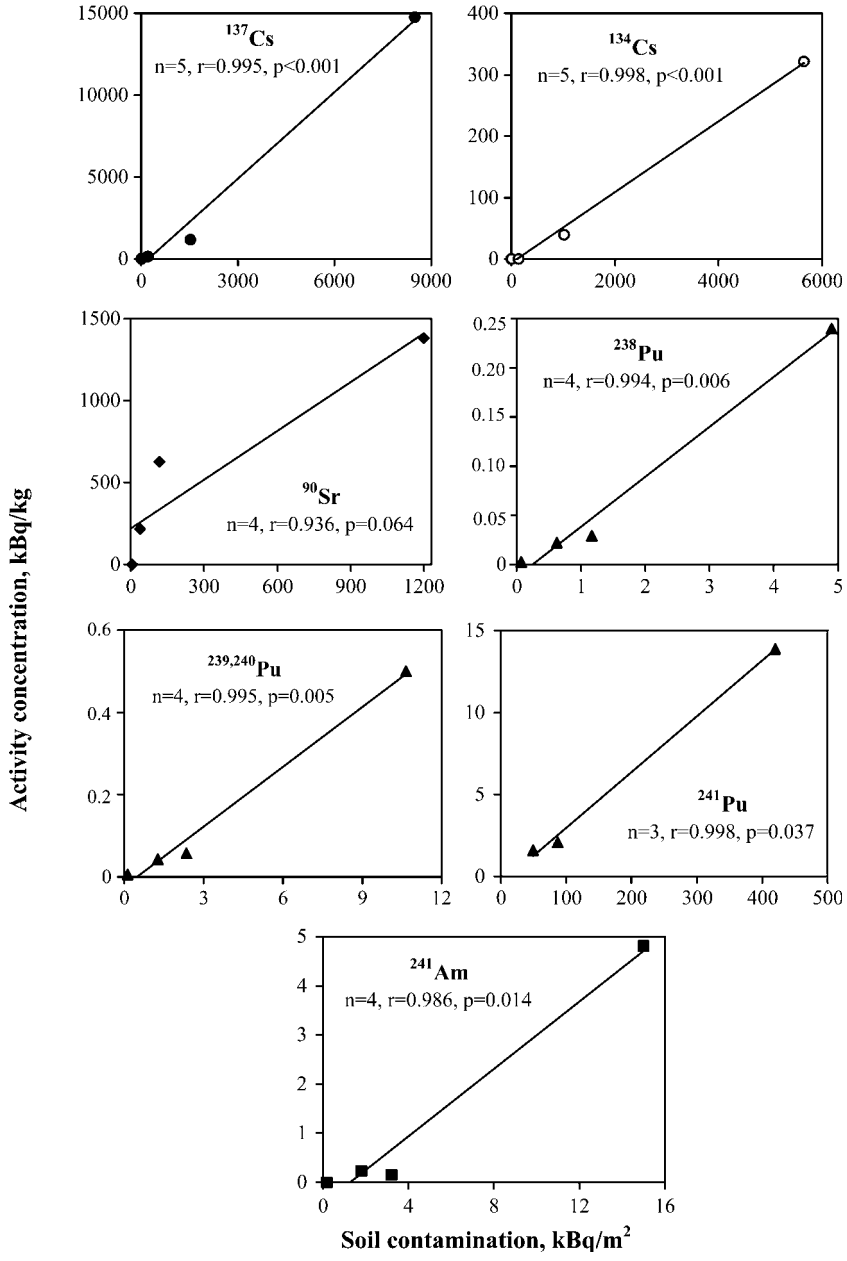

Fig. 4 Correlation of the activity concentration of long-lived radionuclides in bank voles with the contamination of soil, at five monitoring sites and 10 years after the accident

increased from 1 to 8% at site 3 and from less than 1 to 3% in bank voles at site 4, in the period from 5 to 10 years after deposition.

In particular, Table 3 shows that the dose rate values in bone tissue from β-radiation of ^{90}Sr were much less than from the external γ-irradiation and, in most cases, were less than the internal β-irradiation from incorporated cesium isotopes. However, the situation changed when the concentration of ^{137}Cs and ^{134}Cs had significantly decreased and the content of ^{90}Sr in animals living in areas with high levels of ^{90}Sr contamination (sites 3–5) had increased 10 years after the deposition. At that time, the dose rate attributable to β-irradiation of bone tissue by strontium became close to or larger than the dose rate due to β-irradiation by the cesium isotopes. Thus, in the late period after the radionuclide deposition the local dose rates from β-particles of incorporated ^{90}Sr can be considerable in comparison with those from other radionuclides.

Lifetime whole-body doses

The average age of the voles in the trapping seasons was about 3–4 months. The average lifetime whole-body doses in all populations studied were maximal in the year of the accident. At the highly contaminated site 4, they were 55 and 88 mGy for populations of bank vole and yellow-necked mouse, respectively, 4 months after the accident. However, the highly asymmetrical distribution of individual radionuclide content (Fig. 6) suggests that a considerable number of animals have

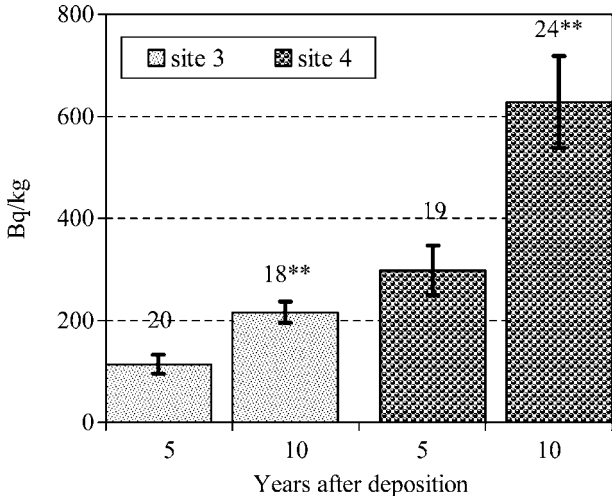

Fig. 5 Time course of ^{90}Sr activity concentration in two populations of bank vole. Standard deviations of the mean values are indicated. The *numbers above the bars* represent the number of animals analyzed: ** $P < 0.01$ in comparison with previous year (*U*-test)

received doses largely exceeding the population mean. Figure 8 illustrates the change of the distribution of the individual doses in the course of the investigation at sites 3 and 4 over 10 years.

Discussion

The bank vole has limited individual home ranges up to 400 m and migration ranges up to 1.2 km in the reproduction period [17, 22]. Similar distances can be assumed for the yellow-necked mouse. Thus, the vole and mouse well represent the habitant areas. This is supported by the close correlation found between the concentrations of incorporated radionuclides in bank vole and the environmental contamination, in this work and elsewhere [3]. Due to the short life span and the high reproduction capacity of the investigated species, the populations studied in a certain year in the trapping season (summer–autumn) almost completely consisted of individuals born at least two generations after the population investigated in the previous year. Thus, the renewed populations represent a radionuclide accumulation during the very short period of their life span (about 3–4 months or somewhat longer).

It is well known that rodents accumulate radionuclides mainly through plant digestion [23]. Some fraction of the bank vole diet consists of radionuclide accumulators such as lichens, mosses and mushrooms [17]. On the other hand, a diet of plant seeds is more preferred by the yellow-necked mouse than by the vole. As the seeds accumulate cesium isotopes less than the other parts of plants [24], (which by the way also leads to a lower radioactivity of the seeding species in comparison with grazers [25]), it becomes clear why the concentrations of incorporated cesium in mice in this work have been found to be less than in bank voles.

The bank vole is known to obtain about 9% of its ^{137}Cs uptake from abiotic sources (soil, water, air) [26]. During the first year after the accident, 35% of the ^{137}Cs and ^{134}Cs contained in the dust occurring on the plant surfaces were biologically unavailable for absorption in the digestion tract of animals [27]. The time course of the cesium isotope activity in bank vole and yellow-necked mouse after the accident, that we observed and described as a sequence of increase, peak and decrease, suggests changes in the concentrations of these isotopes incorporated in the basic diet of the studied animals, due to changes in the bioavailability of these radionuclides for trophic chains in herbivorous animals.

The most important reasons for this phenomenon are (a) the destruction of fuel particles and the appearance of dissolved and exchangeable forms of radiocesium available for plants and mushrooms [28], (b) the vertical migration of ^{137}Cs and ^{134}Cs into deeper soil profiles towards plant roots and mushrooms mycelium [29]. Due to these processes, the root uptake of radionuclides by the plants became the dominant process in the subsequent years after the accident, while superficial plant contamination had been dominated in the first period after the accident. The transfer factor for the "soil–plants" pathway, which depends on exchangeable and dissolved fractions of radiocesium in soil [30], was found to increase in some plants 50 km around the ChNPP by the second and third year since the accident [31], and for the pathways "soil–lamb meat" [32] and "plants–goat milk" [33] it was found to be doubled, respectively, 2 and

Table 2 Radionuclide concentration (Bq/kg) in bank vole 10 years after the accident

Site	Number of samples/animals analyzed for		^{137}Cs	^{134}Cs	^{90}Sr	^{238}Pu	^{239,240}Pu	^{241}Pu	^{241}Am
	^{134}Cs, ^{137}Cs	^{90}Sr, TUE							
1	30/30	–	6.06 ± 0.71	0	–	–	–	–	–
2	40/40	5/30	25.25 ± 3.36	0	1.50 ± 0.45	0.0025 ± 0.0004	0.0058 ± 0.0011	–	0.0036 ± 0.0009
3	18/18	6/18	162 ± 18	0	216 ± 38	0.0220 ± 0.0052	0.0435 ± 0.0099	1.60 ± 0.37	0.23 ± 0.08
4	49/49	8/24	1,182 ± 145	39 ± 4	628 ± 176	0.029 ± 0.005	0.057 ± 0.007	2.09 ± 0.30	0.15 ± 0.04
5	28/28	28/28	14,772 ± 1,585	321 ± 35	1,382 ± 40	0.24 ± 0.02	0.50 ± 0.04	13.90 ± 1.33	4.81 ± 0.42

Mean values and standard deviations are shown. Data marked as "–" were not analyzed

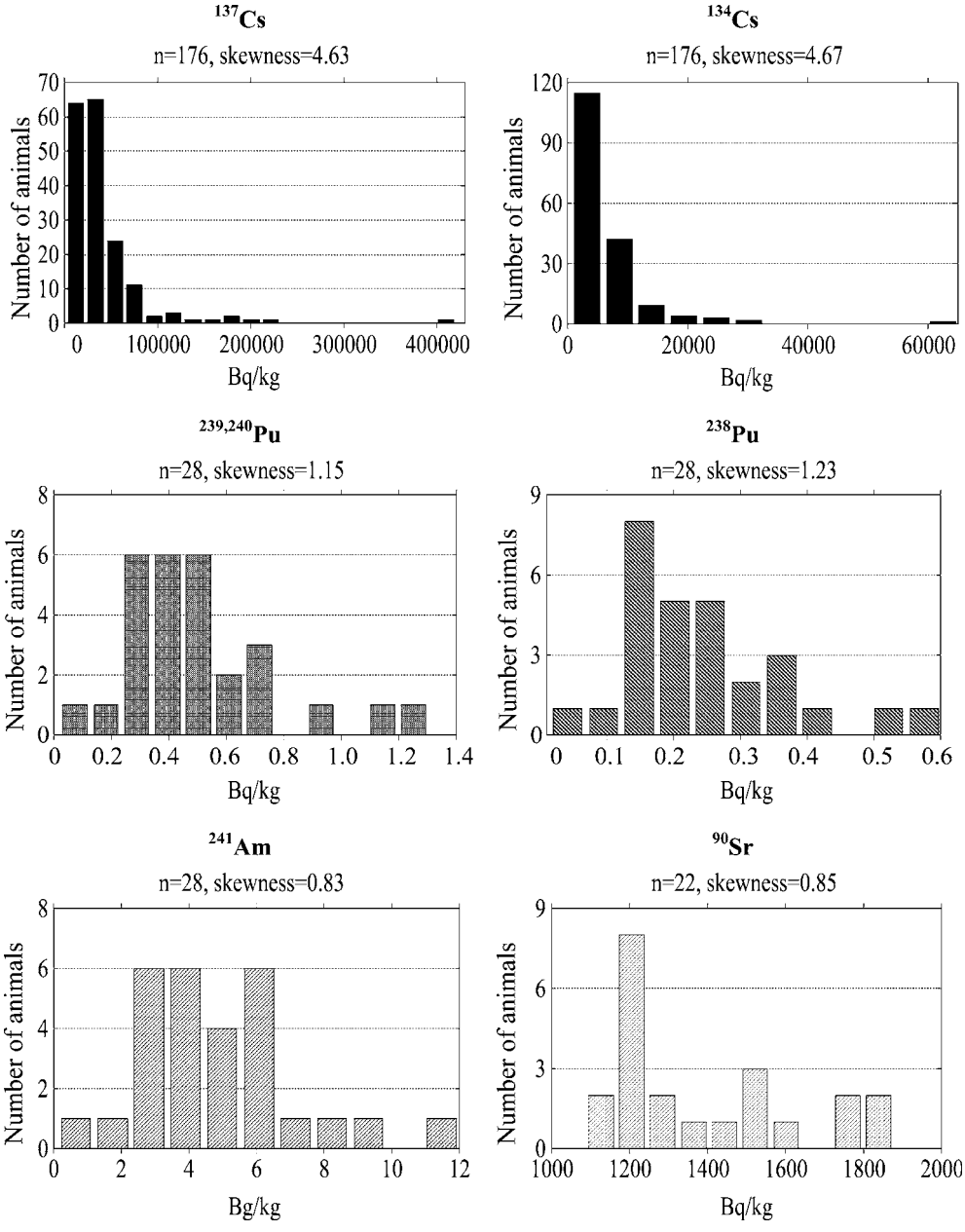

Fig. 6 Typical distributions of individual concentrations of incorporated long-lived radionuclides in bank vole specimens. Data on ^{137}Cs and ^{134}Cs in animals 3 years after deposition at site 4, and on TUE and ^{90}Sr in animals 10 years after deposition at site 5 are shown

3 years after deposition. Maximal values of cesium in various but not all studied species of plants and mushrooms from natural ecosystems in the contaminated areas were observed within 1–3 [32, 34–36] and even 5–6 years [37] after the accident. The concentration of ^{137}Cs in mushrooms, for example, increased by a factor of 4–100 during this period, significantly influencing the cesium content in wild animals [34]. However, in the following period, the process of cesium fixation by soil clay minerals was shown to predominate [38]. Therefore, ^{137}Cs became less available for root uptakes [39], and its concentration in plants decreased up to ten times [40]. The levels of cesium in rodent populations were shown to reflect this decrease in this work.

Radionuclides in a far zone around the ChNPP mainly represent gas-condensed and more mobile (dissolved) forms of fallout, while the fuel particles predominate in a near zone (65–85%) and take time to be destructed [28, 38, 41]. The biological availability of ^{137}Cs in the near zone has been shown to be less than in the far zone by a factor of 1.5–2, during 3–4 years after the accident [42]. These facts are in line with our observation: the nearer the sites, the later the peaks in cesium concentration in animal populations. Specifically, maximum levels of $^{134+137}$Cs content were registered 1–2 years after the accident in populations living at 330 and 60, and 40 km from the ChNPP, respectively (Fig. 2). Maximal concentrations of ^{137}Cs were also detected by other researchers 1 and 3 years after the accident, in bank vole and striped field mouse (*Apodemus silvaticus*) populations living at large distances from the ChNPP like in Great Britain [43] and

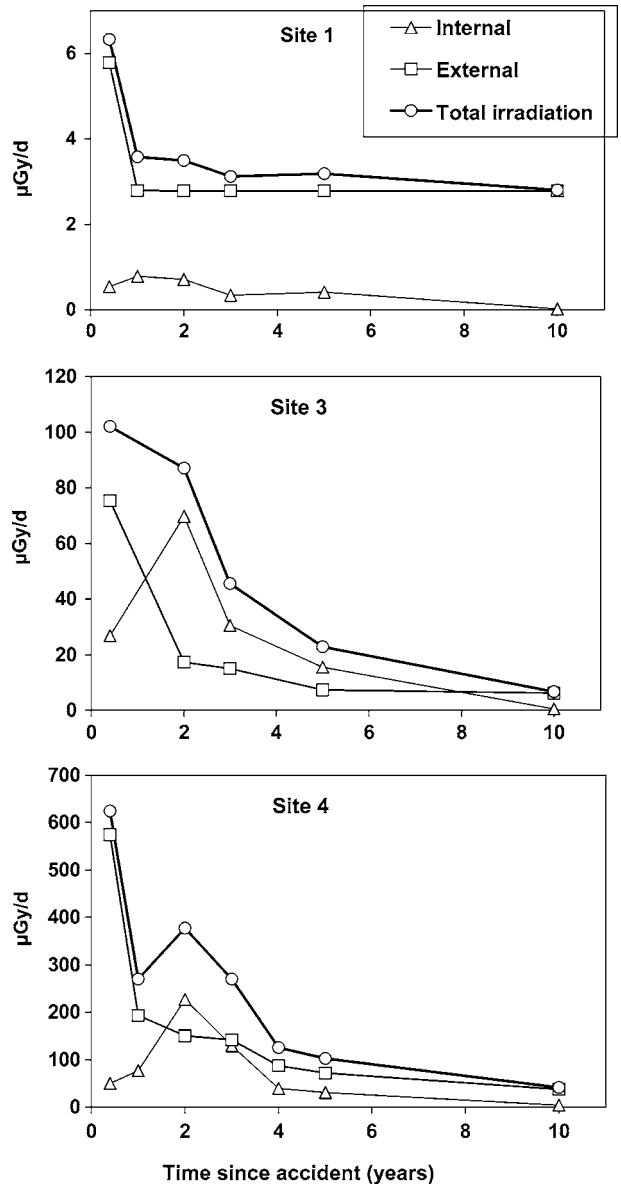

Fig. 7 Time course of mean absorbed dose rate due to external γ- and internal $\gamma + \beta$-irradiation from incorporated ^{137}Cs and ^{134}Cs in bank vole populations

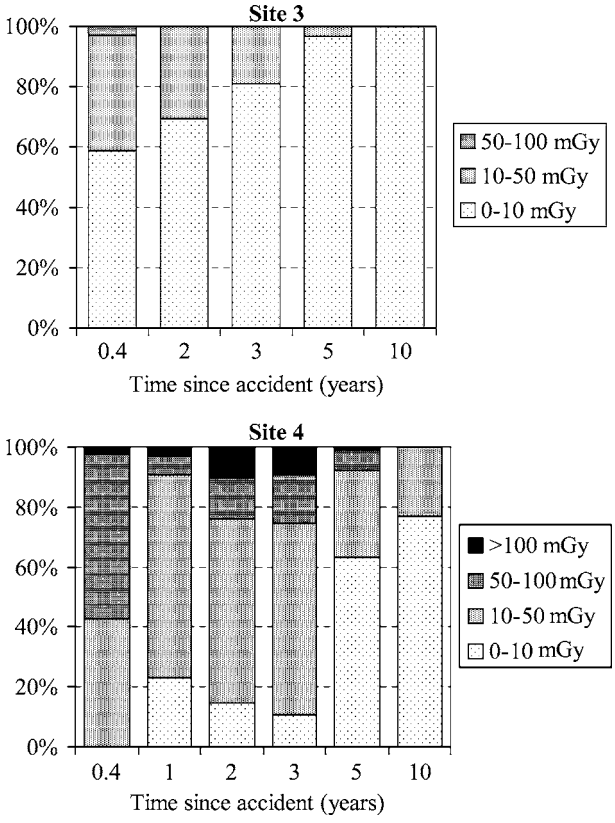

Fig. 8 Time-dependent fractions of bank voles with different values of lifetime whole-body absorbed dose

150–300 km from the ChNPP in Russia [44] respectively. The highest dietary intakes and the highest whole-body measurements of radiocesium were also observed 2–3 years after the accident in critical groups of people (farmers and hunters) with considerable values of meat in the diet, who live in areas of Norway which, according to the Norwegian Radiation Protection Authority, were contaminated with fuel particles and simple cations [32].

Isotopes of strontium and TUE were mostly contained in fuel ("hot") particles of the Chernobyl fallout [38, 45, 46]. During 10–12 years after the accident, the

Table 3 Dose rate values (μGy/d) for external and internal irradiation of whole-body and bone tissue of bank vole

Site	Years after deposition	External whole-body and bone- tissue γ-irradiation (mean data)	Internal irradiation (mean ± SD)					
			$^{137+134}$Cs			^{90}Sr		
			Number of animals	γ-irradiation	β-irradiation	Number of samples/animals	β-irradiation	
				Whole-body and bone tissue			Whole-body	Bone tissue
2	10	2.32	40	0.0062 ± 0.0008	0.0679 ± 0.0090	5/30	0.004 ± 0.001	0.027 ± 0.007
3	5	7.41	53	1.42 ± 0.14	12.76 ± 1.27	20/20	0.31 ± 0.05	2.04 ± 0.34
3	10	6.25	18	0.040 ± 0.004	0.44 ± 0.05	6/18	0.60 ± 0.11	3.87 ± 0.68
4	5	71.80	129	3.07 ± 0.26	27.79 ± 0.47	19/19	0.82 ± 0.13	5.34 ± 0.87
4	10	37.75	49	0.31 ± 0.04	3.23 ± 0.26	8/24	1.73 ± 0.49	11.25 ± 3.15
5	10	222.57	28	3.84 ± 0.41	40.43 ± 4.34	28/28	3.81 ± 0.14	24.78 ± 0.92

particle destruction increased the quantity of mobile forms of ^{90}Sr in the soil [47]. Due to lower fixation of mobile ^{90}Sr on soil components in comparison with the fixation of cesium isotopes, the ^{90}Sr activity concentration in soil pore solutions was equal or even larger than that of ^{137}Cs, which dominated in the fallout [38]. These facts contributed to a significant increase of the content of ^{90}Sr in plants by a factor of 5–10 while the ^{137}Cs concentration decreased [40]. As a result, the concentration of radiostrontium in the studied rodent populations also increased and had a tendency to be higher than the level of incorporated ^{137}Cs. As the value of mobile ^{90}Sr in a different type of soil is predicted to increase up to the year 2006 [47], the same time course of ^{90}Sr can be expected in plants and herbivorous animals.

A temporal delay in the accumulation of maximal concentrations of ^{137}Cs and ^{90}Sr in animal populations was also observed after nuclear weapons tests [5]. Since air-borne fuel particles can hardly migrate to large distances from the source, this type of delay has to be obviously attributed to reasons different from particle destruction, such as the time necessary for the radioactive fallout to reach the plant roots by vertical migration.

The behavior of TUE differs from the above mentioned radionuclides. Detectable levels of ^{241}Am appeared in bank vole populations not before 5 years after the accident, and increased in the subsequent period due to ^{241}Pu decay as well as due to good solubility and availability of its daughter element ^{241}Am for plants [38]. Since maximal levels of ^{241}Am in the soil are predicted for about 70 years after the accident [1], we assume that a similar situation will occur in animal populations inhabiting these regions. The fraction of biologically accessible forms of plutonium isotopes in the contaminated soils were found to be low and constant during the years after the accident, due to their fixation by soil components [38]. This, as well as, respectively, low levels of ^{238}Pu and ^{239,240}Pu detected in bank vole populations, allow us to conclude that the values of these isotopes will not show a further rise in the biota of the contaminated areas.

The study firstly demonstrates that in small mammals during a decade following the radionuclide deposition, the whole-body dose rates generally fall in a monotonic manner, except during the initial phase where parameters like the biological availability of radionuclides are subject to fast changes. We have also found that the distributions of long-lived radionuclide concentrations and of dose rates in animals are highly asymmetrical and can be roughly approximated by log-normal functions. Approximately log-normal radionuclide distributions have been established in different species [5, 48, 49], and they appear to be usual for all biota. For such type of distribution, the individual doses can be considerably higher than the mean population doses and may fluctuate up to about 10% of the LD_{50} in populations of bank vole and yellow-necked mouse, where LD_{50} is 9.7 and 5.4 Gy, respectively [5].

Conclusions

The long-lived radionuclides released from the destroyed reactor of the ChNPP have different half-times, pathways and rates of migration in soil and, consequently, in biota. Probably, these factors determine the time course of radionuclide concentrations and accumulated doses in natural populations of mammals living in contaminated areas, observed and described in the present study. The data obtained can be used as a trend for prediction models of radionuclide uptake into biota, especially for the herbivorous groups of animals in natural ecosystems.

Acknowledgements This work was supported by the Governmental Program on "The overcoming of the consequences of the Chernobyl catastrophe in the Republic of Belarus" (1986–2002). The authors thank the staff of the Antimutagenesis Laboratory, Institute of Genetics and Cytology, National Academy of Belarus, for field and laboratory assistance. We also thank Dr. A. Rozhdestvenskaya and A. Kashtalian, Institute of Zoology, National Academy of Sciences of Belarus, for the analysis of animal age. We are grateful to the staff of the Institute of Physics, National Academy of Sciences of Belarus, and the Hydrometeorological Centre of Belarus for the spectrometric analysis. We gratefully acknowledge support from Dr. M. Malko, Institute of Physical and Radiation Problems, National Academy of Sciences of Belarus, for the recommendations for dose assessment. We especially thank Prof. D. Harder, University of Goettingen, for a critical review of the manuscript and helpful suggestions.

References

1. Yaroshevich OI, Zhuk IV, Lomonosova EM (1993) Development of tool methods for controlling the content of plutonium and americium in soil (in Russian). Doklady Academii Nauk Belarusi 37:49–53
2. Yakushev BI, Budkevich TA, Zabolotny AI, Mironov VP, Kudryashov VP (1999) Contamination of vegetation in Belarus by transuranium radionuclides due to Chernobyl NPP accident. In: Goossens LHJ (ed) Proceedings of the 9th Annual Conference "Risk analysis: facing the new millennium", Delft University Press, Rotterdam, the Netherlands, October 10–13, 1999, pp 841–844
3. Cristaldi M, Ieradi LA, Mascanzoni D, Mattei T, Von Bothemer S (1991) Environmental impact of the Chernobyl fallout: mutagenesis in bank vole from Sweden. Int J Radiat Biol 59:31–40
4. Copplestone D, Johnson MS, Jones SR, Total ME, Jackson D (1999) Radionuclide behavior and transport in a coniferous woodland ecosystem: vegetation, invertabrates and wood mice, Apodemus sylvaticus. Sci Total Environ 239:95–109
5. Iljenko AI, Krapivko TP (1989) Ecology of animals in radiological biocenosis (in Russian). Nauka, Moscow
6. Goncharova RI, Ryabokon NI (1995) Dynamics of cytogenetic injuries in natural populations of bank vole in the Republic of Belarus. Radiat Prot Dosim 62:37–40
7. Andrukovich SK, Berestov AV, Antovich N, Vukotich P, Gurinovich AA, Khrutchinsky AA, Meletisa ON (2003) Investigation of orthopositronium 3γ-decay using a multidetector spectrometer. Nucl Instr Meth in Phys Res B 207:219–226
8. Pavlotskaia FI, Myasoedov BF (1996) Determination of transuranic elements in nature components. Radiochimia 38:193–209
9. Yaroshevich OI, Zuk IV, Lomonosova EM, Svetlakova NM, Mironov VP, Kudryashov VP, Bushuev AV (1995) Determination of Pu and Am-241 in soil by instrumental method. Nucl Geophys 9:235–239

10. Moiseev AA, Ivanov VI (1990) Handbook of dosimetry and radiation hygiene (in Russian). Energoatomizdat, Moscow
11. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (1996) Effects of radiation on the environment. Sources and effects of ionizing radiation. Report of the General Assembly. New York
12. International Commission on Radiological Protection (ICRP) (1979) Limits for intakes of radionuclides by workers. ICRP Publication 30. Part 1. Ann. ICRP 3/4. Pergamon Press, Oxford
13. International Commission for Radiation Units and Measurements (ICRU) (1992) Photon, electron, proton and neutron interaction data for body tissues. ICRU Report 46. ICRU Publications, Bethesda
14. International Commission on Radiological Protection (ICRP) (1983) Radionuclide transformations. Energy and intensity of emissions. ICRP Publication 38. Ann. ICRP 11–13. Pergamon Press, Oxford
16. Kudyasheva AG, Shishkina LN, Zagorskaya NG, Naskaev AI (1997) Biochemical mechanisms of radiation affection of natural populations of murine rodents (in Russian). Nauka, Moscow
21. Starichenko VI, Liubashevski NM (1998) The individual characteristics of ^{90}Sr in the body of 2 species of the common voles inhabiting the territory of the eastern Urals radioactive trace (in Russian). Radiats Biol Radioecol 38:375–383
17. Bashenina NV (ed) Bank vole (1981) (in Russian). Soviet Committee for the UNESCO Programme "Man and biosphere". Nauka, Moscow
18. Marei AN, Barkhudarov RM, Chumak VK, Petukhova EV, Tiuriukanova EB (1970) Exposure of the population of the Polecia region to Cs-137 and Sr-90 through food (in Russian). Gig Sanit 35:61–66
19. Shevchouk VE, Gourachevskiy VL (eds) (2001) Fifteen years after the Chernobyl catastrophe: consequences in the Republic of Belarus. National Report Committee on the problems of the consequences of the accident at the Chernobyl NPP, Minsk
20. Hardy EP, Krey PW, Volchok HL (1973) Global inventory and distribution of fallout plutonium. Nature 241:444–445
15. Ternov VI, Kondrat'ev AG (1990) Gamma-dosage rate in BSSR in 1981–1982 (in Russian). Zdravookhranenie Belorussii 6:61
22. Andrzejewski K, Babinska-Verka J (1986) Bank vole populations: are their densities really high and individual home ranges small? Acta Theriol 31:409–422
23. Sokolov VE, Krivolutsky DA, Usachev VL (1989) Wild animals in global radioecological monitoring (in Russian). Nauka, Moscow
24. Gancharova NV, Gaponenka VI, Bagdanau AP, Darashkevich VN (1992) Radionuclide accumlation in plant organs and their intercellular localization (in Belarusan) Vesti Academii Navuk Belarusi. Ser Biyal Navuk 2:53–55
25. Whicker FW (1983) Radionuclide transport processes in terrestrial ecosystems. Rad Res 94:135–150
26. Chesser RK, Rodgers BE, Wickliffe JK, Gaschak S, Chizhevsky I, Philips CJ, Baker RJ (2001) Accumulation of ^{137}Cs and ^{90}Sr from abiotic and biotic sources in rodents at Chornobyl, Ukraine. Environ Toxicol Chem 20:1927–1935
27. Hove K (1994) Critical factors determining the uptake of radionuclides in animals and animal products. In: Russian-Norwegian Satellite Symposium on nuclear accidents, radioecology and health, 27–28 October 1994, Moscow, Part 2:319
28. Petryaev EP, Ovsyannikova SV, Lyubkina Iya, Rubinchik SYa (1990) The forms of radionuclides in soils of contaminated regions of Belarus (in Russian). Vesti Academii Navuk Belarusi Ser Phys Ener Navuk 4:65–69
29. Sanzharova NI, Kotik VA, Arkhipov AN, Sokolik GA, Ivanov YuA, Fesenko SV, Levchuk SE (1996) The quantitive parameters of the vertical migration of radionuclides in the soils in different types of meadows (in Russian). Radiats Biol Radioecol 36:488–497
30. Fesenko SV, Voigt G, Spiridonov SI, Sanzharova NI, Gontarenko IA, Belli M, Sansone U (2000) Analysis of contribution of forest pathways to radiation exposure of different population groups in Bryansk region of Russia. Radiat Environ Biophys 39:291–300
31. Fesenko SV, Spiridonov SI, Sanzharova NI, Aleksakhin RM (1996) Modelling of the biological availability of ^{137}Cs in soils subjected to contamination after the accident at the Chernobyl Atomic Electric Power Station (in Russian). Radiats Biol Radioecol 36:479–489
32. Strand P (1994) Radioactive fallout in Norway from the Chernobyl accident studies on the behaviour of radiocaesiums in the environment and possible health impacts. NRPA Report 1994:2. Norwegian Radiation Protection Authority, Osteras
33. Solheim HH, Hove K (1991) Radiocesium bioavailability: transfer of Chernobyl and tracer radiocesium to gout milk. Health Phys 60(5):665–673
34. Mascanzoni M (1992) Determination of ^{90}Sr and ^{137}Cs in mushrooms following the Chernobyl fallout. J Radioanal Nuclear Chem 161:483–488
35. Yushkov PI, Chueva TA, Karavaeva EN (1993) Content of $^{134+137}$Cs and ^{90}Sr in the crown of the birch in the first years after the accident in the Chernobyl NPP (in Russian). Ecologiya 1:86–91
36. Shcheglov AI, Tikhomirov FA, Tsvetnova OB, Kliashtorin AL, Mamikhin SV (1996) The biogeochemistry of the radionuclides of the Chernobyl release in the forest ecosystems of the European portion of the CIS (in Russian). Radiats Biol Radioecol 36:469–478
37. Fesenko SV, Soukhova NV, Sanzharova NI, Avila R, Spiridonov SI, Klein D, Badot PM (2001) ^{137}Cs availability for soil to understory transfer in different types of forest ecosystems. Sci Total Environ 269:87–103
38. Sokolik GA, Ivanova TG, Leinova SL, Ovsiannikova SV, Kimlenko IM (2001) Migration ability of radionuclides in soil-vegetation cover of Belarus after Chernobyl accident. Environ Int 26:183–187
39. Andersson I, Lonsjo H, Rosen K (2001) Long-term studies on transfer of ^{137}Cs from soil to vegetation and to grazing lambs in a mountain area in northern Sweden. J Environ Radioact 52:45–66
40. Radiation contamination of flora in Belarus (due to the accident in Chernobyl NPP) (1995) Parfenov VI, Yakushev BI (eds) (in Russian). Navuka i tekhnika, Minsk
41. Mironov V, Matusevitch Z, Kudrjashov V (2001) Destruction of fuel particles from the Chernobyl fall-out on the territory of Belarus. In: Frontasieva M (eds) Radionuclides and heavy metals in environment. Kluwer, Dordrecht, pp 55–60
42. Ivanov Yu, Kashparov V, Sandalls J (1996) Fuel component of ChNPP release fallout: properties and behavior in the environment. In: The radiological consequences of the Chernobyl accident: proceedings of the first international conference, Minsk, 18–22 March 1996:173–177
43. Rudge SA, Johnson MS, Leah RT (1993) Biological transport of radiocaesium in seminatural grassland ecosystem. J Environ Radioact 19:199–212
44. Adamovich VL, Merkushina OS (1997) The effect of small radiation doses on biological changes in population groupings of murine rodents. Radiats Biol Radioecol 37:303–311
45. Kashparov VA, Ivanov YuA, Zvarich SI, Protsak VP, Khomutinin YuV, Kurepin AD, Pazukhin EM (1996) Formation of hot particles during the Chernobyl nuclear power plant accident. Nucl Technol 114:246–253
46. Carbol P, Solatie D, Erdmann N, Nylen T, Betti M (2003) Deposition and distribution of Chernobyl fallout fusion products and actinides in the Russian soil profile. J Environ Radioact 68:27–46
47. Kashparov VA, Ahamdach N, Zvarich SI, Yoschenko VI, Maloshtan IM, Dewiere L (2004) Kinetics of dissolution of Chernobyl fuel particles in soil in natural conditions. J Environ Radioact 72:335–353

48. Oleksyk TK, Gashchak SP, Glenn TC, Jagoe CH, Peles JD, Purdue JR, Tsyusk OV, Zalissky OO, Smith MH (2002) Frequency distributions of ^{137}Cs in fish and mammal populations. J Environ Radioact 61(1):55–74

49. Assimakopoulos PA, Ioannides KG, Pakou AA, Lolis D, Zikopoulos K, Dusias B (1989) Radiocesium levels measured in breast milk one year after the reactor accident at Chernobyl. Health Phys 56:103–106