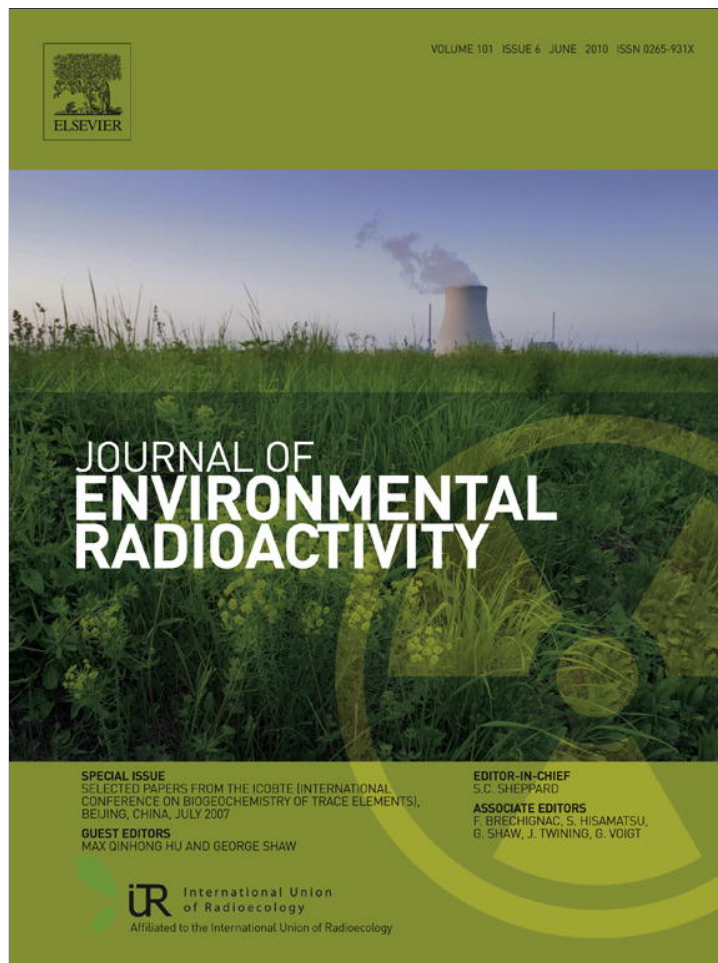


Provided for non-commercial research and education use.
Not for reproduction, distribution or commercial use.



This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

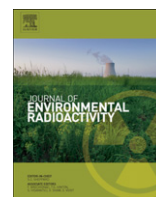
In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

<http://www.elsevier.com/copyright>



Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Radionuclides in terrestrial ecosystems of the zone of Kyshtym accident in the Urals

V.N. Pozolotina*, I.V. Molchanova, E.N. Karavaeva, L.N. Mikhaylovskaya, E.V. Antonova

Institute of Plant and Animal Ecology, Ural Division of Russian Academy of Sciences, 8 Marta Street 202, 620144 Yekaterinburg, Russia

ARTICLE INFO

Article history:

Received 1 November 2007

Received in revised form 26 May 2008

Accepted 8 June 2008

Keywords:

Radionuclides

Transport

Accumulation

Soil

Plant

Radiation

Doses

Kyshtym accident

ABSTRACT

It was shown that along the Eastern Ural Radioactive Trace central axis, about 100 km in length, decrease of the ^{90}Sr and ^{137}Cs deposition densities in soil samples may be described as an exponential function. At the western and eastern periphery of the trace, ^{90}Sr contents in soils approached to the background level due to global fallout. ^{90}Sr and ^{137}Cs concentrations in seeds of some herbaceous plants have been determined. The radionuclide concentrations and the resulting dose loads upon plant seeds showed an excess over the background level of about two or three orders of magnitude.

Published by Elsevier Ltd.

1. Introduction

One of the most serious nuclear accidents took place in the South Urals in 1957. About 74 PBq of radioactive substances were released into the atmosphere, resulting in the formation of the Eastern Ural Radioactive Trace (EURT). During the first years after the accident, in the frame of complex program "Mirage" (1958–1960), there received information on radionuclide concentrations in different soils, accumulation and transport of those into different plant and animal species. The dose loads upon biota and humans were estimated (Experimental Scientific Research, 2005). Nine years later, the Eastern Ural State Radioactive Reserve (EUSRR) was established in the frontal part of this trace and actually became the test ground for experiments in nature. With time, ^{90}Sr left as the main contaminant, short-living radionuclides already decayed. On the other hand, EURT area was also polluted from some other sources. Since the beginning 1951 the small, natural Lake Karachay has been used for a deposition of medium-level radioactive waste. In 1967 from a strip exposed by drought along the shore of this Lake silt and fine sand were raised by wind. In this case the territory of EURT was again subjected by contamination with radionuclides. The ^{137}Cs was predominant in the composition of this contamination (Aarkrog et al., 1997).

For reasons of secrecy only fragmentary data were included in accessible publications, usually without indicating the place and conditions of its contamination (Klement et al., 1968). In the aftermath of the Chernobyl accident in 1986 unclassified information on contamination events in Southern Urals was released (Nikipelov et al., 1990). From 1993, complex radioecological studies are processed in the EURT zone (Yachmenev and Isaeva, 1996; Eastern Urals Radioactive Trace, 2000; Molchanova and Karavaeva, 2001; Pozolotina, 2003). The present study is a continuation of the former research conducted in collaboration with RISO National Laboratory, in which reported on radionuclide inventories in the EURT (Aarkrog et al., 1997). The first investigation did not consider contamination levels in the head part of the trace that coincide with the reserve area, which is explained by missing the relevant data.

The aim of this work have been to evaluate recent deposition densities in soils, distribution of radionuclides between ecosystem components as impact as buffer parts of trace along the contamination gradient and to estimate the dose loads upon seed embryos in some plant species.

2. Material and methods

With concern to the principle of ranging of the contaminated areas, three transects were established: a central one coinciding with EURT central axis, and the western and eastern ones, through the periphery parts of the EURT (Fig. 1). Two zones were distinguished within the central transect area: an impact one corresponding to the EUSRR area (6–30 km long) and a buffer zone within 30–100 km

* Corresponding author. Fax: +7 343 260 65 00.

E-mail address: pozolotina@ipae.uran.ru (V.N. Pozolotina).

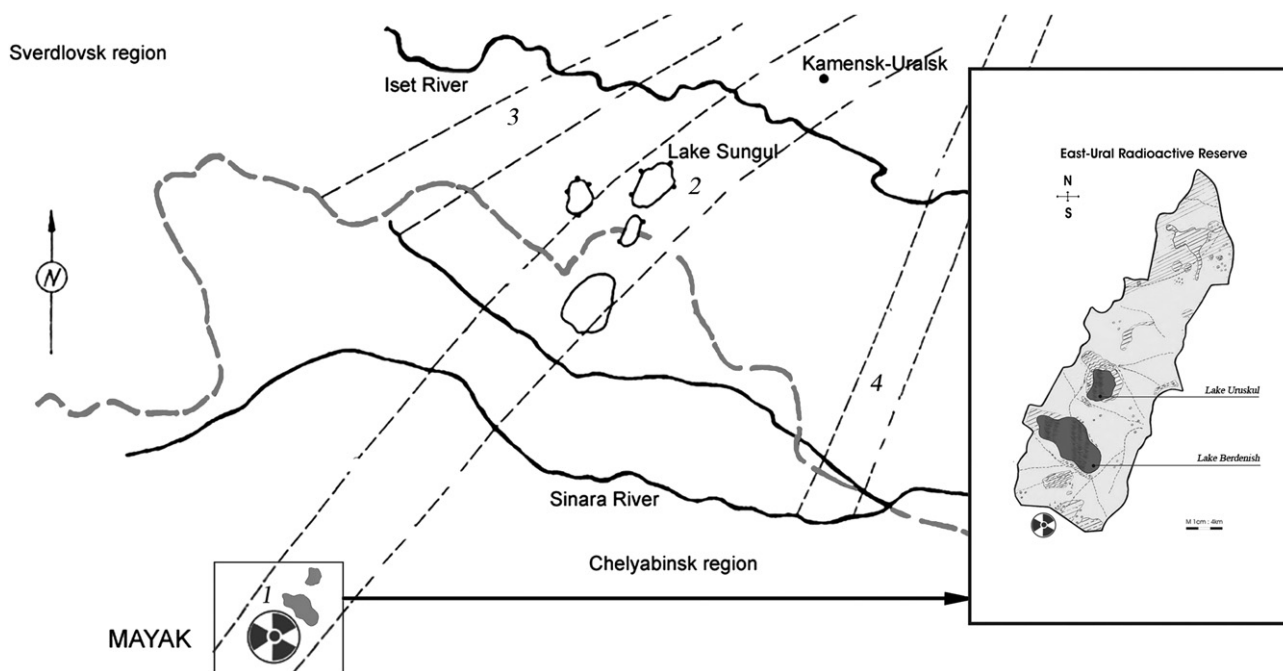


Fig. 1. The sketch map of the investigation region. Central transect: 1 – impact zone; 2 – buffer zone; 3 – western transect; 4 – eastern transect.

along the EURT central axis. On the territory of study there are forests, small birch groves, meadows with herbs and grasses, arable lands and old-ploughed fields became overgrown with weeds. Sedge-and-grass communities cover the lake banks. The background plots were selected beyond the polluted zone. They were similar by their soil and ecological features. Two or three soil profiles were examined for each of the selected plots. Plant vegetative mass and seeds of some herbaceous species were sampled near to the soil profiles.

Concentrations of ^{90}Sr in all samples were determined radiochemically, from the content of daughter ^{90}Y , and ^{137}Cs was determined using a Canberra multichannel gamma-analyzer with a semiconductor detector. The statistical error of measurements did not exceed 15%, and the radionuclide detection limit was 1 Bq/kg. Data on ^{90}Sr and ^{137}Cs in seeds were used to calculate dose loads upon the most sensitive ontogeny phases in wild herbs.

3. Results and discussion

3.1. The deposition densities and spatial distribution of radionuclides in EURT soils

Fig. 2 shows the contamination density of the radionuclides in the soils within the central transect, differently distant from the plant of “Mayak”. For the contamination density, it was agreed to consider the amount of radionuclides stored within a 0–40-cm soil layer. It was shown ^{90}Sr store in impact zone soils decreased with distance from the place of accident, from 25 000 to 4000 $^{\circ}$ kBq/m 2 ; in the buffer zone the values decreased from 3000 to 70 kBq/m 2 . It appeared that depositions decrease after the exponential function: $y = 13687e^{-0.0605x}$; with coefficient of determination $R^2 = 0.814$. One should notice that even 100 km away from the contamination source, ^{90}Sr stock in the soils was over 20 times higher than the level of control values varying from 1.5 to 1.8 kBq/m 2 , both according to the UNSCEAR data and our estimates (Aarkrog et al., 1997).

Just near to the accident epicenter (6 km), ^{137}Cs stock in soils was 670 kBq/m 2 . Farther away from the “Mayak” plant, its values decreased gradually. At the distances over 30 km the contamination density varied within 5–10 kBq/m 2 . The ^{137}Cs deposition density (within the central transect) also approximates exponential function: $y = 138.45e^{-0.0362x}$, at $R^2 = 0.538$. The total amount of ^{137}Cs decreased, but spatial distribution of this element showed an irregular pattern. For example, some samples collected in places,

close to each other revealed the values of ^{137}Cs stock differing at about 10 times. Heterogeneity of the contamination levels may be due to radionuclide input from some other sources, mainly because of wind-dispersion of dried sediments from shores of Karachay.

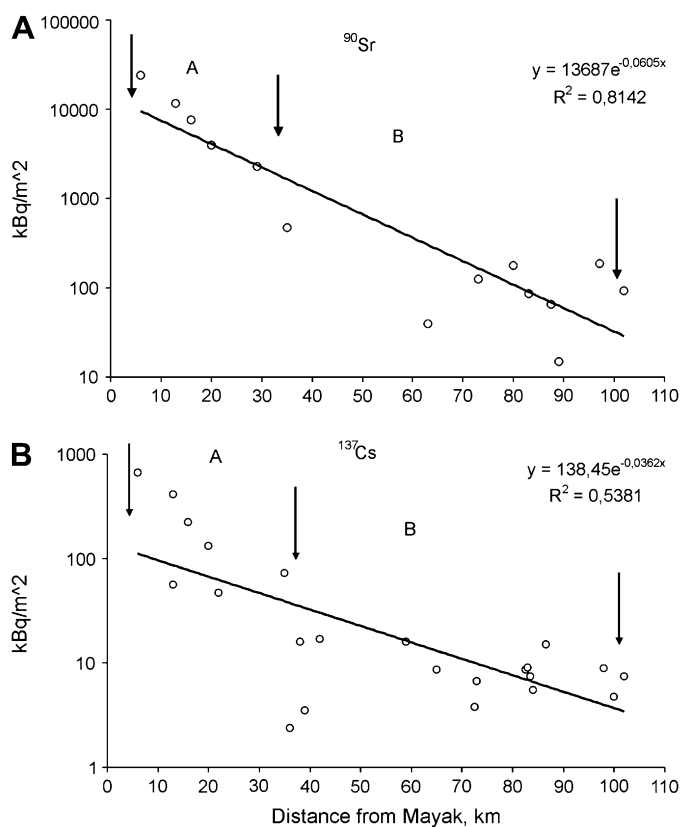


Fig. 2. Contamination density of EURT soils with the distance from accident place: (A) impact zone, (B) buffer zone.

To reveal features of the radionuclides' distribution pattern in the soil cover of impact and buffer areas, we examined the watershed and bank areas of lakes. Table 1 presents the data for one impact plot situated near the Berdenysh lake (55°48'N, 60°51'E) and those for two buffer plots, one in the vicinity of the Sungul lake (56°22'N, 61°45'E) and another at the Tygish lake (56°21'N, 61°33'E).

One can see that the impact plot demonstrated high heterogeneity of radionuclides' contamination. The lowest stocks have been registered in often-flooded nearest to bank zone. Farther from the water edge, radionuclides' contents increased, as well as the ⁹⁰Sr/¹³⁷Cs ratio in soils. In the watershed area, the highest ⁹⁰Sr stock was found in the native soil of a dry meadow.

In the buffer zone, gradient of the radionuclides' contamination was not evident enough. However, one can mark a significant increase of ⁹⁰Sr content in soils of the lake shores ($t_{St} > 3.9$ at $t_{0.05} = 2.6$). The found distinctions in distribution of radionuclides may be due to the following factors. At the territory of EUSRR no anthropogenic load exists. High density of the vegetation cover and deficient moisturing in summer limited run-off in the watershed area. Thus, the excessive moistening soils of the bank zone intensified the radionuclides' migration (Karavaeva and Molchanova, 1979), as a result, processes of self-decontamination predominating. In the buffer zone, intensive agricultural using of watershed areas increases water and wind erosion of soils, meanwhile resulted in redistribution of radionuclides and accumulation of those in by-lake depressions.

Western and eastern transects of the EURT represented populated territories; samples were collected in the settlement outskirts. In the contiguous with the reserve areas, the soil cover revealed higher contamination density for ⁹⁰Sr, about 40–80 kBq/m² at the western boundary, and 30–50 kBq/m² at the eastern one. In both western and eastern transects, average ⁹⁰Sr stock varied within 1.5–4.5 kBq/m² (Fig. 3). The ¹³⁷Cs content in soil plots examined in the eastern sector varied within 3–9 kBq/m², and only in some cases reached value of 12–20 kBq/m². In EURT western part, ¹³⁷Cs deposition densities were higher than those in the eastern one. It indicated the additional input of ¹³⁷Cs from some other sources, probably due to wind transport from the lake Karachay (Aarkrog et al., 1997).

At earlier stages of the accident trace formation the nuclear fall-outs were delayed by the plant cover and thin top soil. Over 50-years period, due to the migration processes, both spatial and vertical redistributions of radionuclides in the soil occurred. Now in EURT

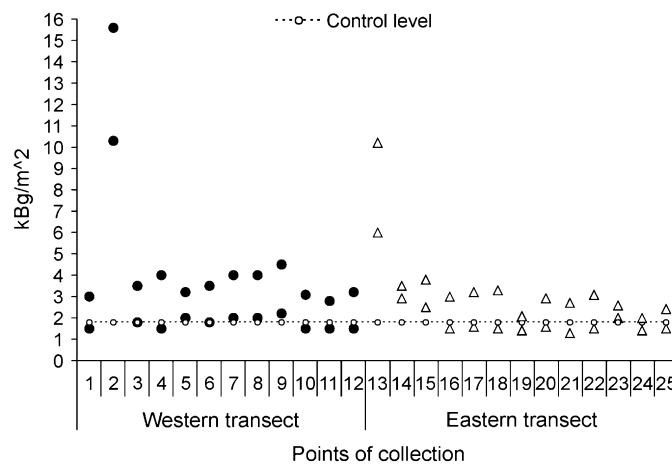


Fig. 3. ⁹⁰Sr contamination densities in soils of EURT transects. Sampling points situated in order of increasing distance from the accident point.

impact and buffer zones the 20-cm soil layer. It keeps 85–92% of the total ⁹⁰Sr and ¹³⁷Cs stocks. Thus, these radionuclides characterize low mobility in the soil profiles. Firmly absorbed in soils, radionuclides generated additional dose loads upon living organisms.

3.2. Transport of radionuclides into plants and evaluation of the absorbed doses upon seed embryos

Previously we have shown (Pozolotina et al., 2005) that in the impact zone, concentrations of the main contaminator (⁹⁰Sr) in aboveground parts of the herbaceous plant species were by 3–4 orders of magnitude higher than those in buffer and background zones, but the corresponding coefficients of accumulation were significantly lower. This may be due to either physical–chemical state of the radionuclides in soils, or due to biological peculiarities of plants adapted to life at the contaminated conditions. For example, Zheleznov et al. (2002) have shown that accumulation of ⁹⁰Sr by plants directly correlates with its contents in the soil, whereas the accumulation is controlled by the plant genotype within the same contamination density. In different populations of the Eastern Ural Radioactive Trace zone shifts of portion of plants with the high and low accumulation abilities were recorded.

The corresponding distinctions in ⁹⁰Sr contents have been also revealed in plant seeds collected in EURT different plots and background areas. The highest ⁹⁰Sr concentrations in seeds were found in plants growing at the impact plot. In seeds from the buffer plot the values were lower by 1–3 orders of magnitude, and they were hardly detectible in seeds of the control samples (Table 2). The highest concentrations of ¹³⁷Cs were also determined in seeds from impact zone.

The absorbed dose rate was calculated as $M = q_1 L(^{90}\text{Sr} + ^{90}\text{Y}) + q_2 L(^{137}\text{Cs})$, where q_1 and q_2 are the specific activities of each radionuclide in plant seeds, and L is the absorbed dose (cGy/s) produced by this radionuclide within the homogeneously contaminated volume at $q_0 = 3.7 \times 10^4$ Bq/g (Gorshkov, 1967). Although the model is simple, it enables to evaluate, with a fair degree of confidence, absorbed doses for plant seeds and compares levels of radiation dose loads in various EURT locations, including background areas.

Table 2 demonstrates that in the impact zone dose loads upon embryos per month due to the radionuclides incorporated in seeds are by 3–4 orders of magnitude higher than those in the background samples (the significance of differences between populations, $\chi^2 = 18.33$, $df = 2$, $p = 0.0001$). High variety of absorbed doses upon seeds in the impact zone may be related to

Table 1
Stock of radionuclides in soils of different plots of landscape

| Plot | Distance from the water edge (m) | ⁹⁰ Sr (kBq/m ²) | ¹³⁷ Cs (kBq/m ²) | ⁹⁰ Sr/ ¹³⁷ Cs |
|------------------------------------|----------------------------------|--|---|-------------------------------------|
| Impact zone | | | | |
| Berdenysh lake | | | | |
| South-western bank | 1 | 270 ± 52 | 58 ± 10 | 4.6 |
| | 4 | 2333 ± 630 | 207 ± 48 | 11.3 |
| | 20 | 16690 ± 354 | 699 ± 180 | 23.9 |
| Watershed area: | | | | |
| Upland herb-grass meadow, native | 150 | 13888 ± 290 | 401 ± 95 | 34.6 |
| Anthropogenically disturbed | 150 | 9455 ± 270 | 249 ± 63 | 38.0 |
| Birch forest | 300 | 8152 ± 86 | 310 ± 86 | 26.3 |
| Buffer zone | | | | |
| North-western bank of Sungul' lake | | | | |
| | 10 | 22.7 ± 9.3 | 10.8 ± 3.5 | 2.1 |
| Northern bank of Tygish lake | | | | |
| | 10 | 34.3 ± 9.8 | 13.6 ± 3.0 | 2.5 |
| Watershed area: | | | | |
| Pasture | 200 | 13.0 ± 5.0 | 13.3 ± 3.8 | 1.0 |

Table 2
Concentrations of radionuclides in seeds of some plant species and resulting absorbed doses on the embryos

| Zone | Plant species | Concentration (Bq/kg) | | Absorbed dose per month (mGy) |
|----------------------------------|---|-------------------------|-------------------|-------------------------------|
| | | ⁹⁰ Sr | ¹³⁷ Cs | |
| Impact | <i>Cirsium vulgare</i> (Savi) Ten. | 15 933 ± 4500 | 400 ± 15 | 3.84 ± 0.61 |
| | <i>Centaurea scabiosa</i> L. | 45 233 ± 13 000 | 37 ± 12 | 10.61 ± 3.24 |
| | <i>Melandryum album</i> (Mill.) Garke | 8500 ± 2400 | 220 ± 54 | 2.05 ± 0.54 |
| | <i>Stellaria graminea</i> (L.) | 26 500 ± 8100 | 190 ± 60 | 6.26 ± 1.94 |
| | <i>Urtica dioica</i> L. | 28 406 ± 8500 | 267 ± 72 | 6.73 ± 1.91 |
| | <i>Sanguisorba officinalis</i> L. | 296 900 ± 89 700 | 80 ± 26 | 69.61 ± 21.82 |
| | <i>Plantago major</i> L. | 3536 ± 975 | 40 ± 15 | 0.84 ± 0.27 |
| | <i>Leonurus quinquelobatus</i> Gilib. (<i>L. vilosus</i> Desf.) | 27 100 ± 7925 | 10 ± 3 | 6.36 ± 1.88 |
| | <i>Rumex confertus</i> Willd. | 8430 ± 2430 | 252 ± 62 | 2.04 ± 0.55 |
| | Buffer | <i>Urtica dioica</i> L. | 135 ± 47 | 10 ± 4 |
| <i>Taraxacum officinale</i> s.l. | | 82 ± 21 | 15 ± 5 | 0.02 ± 0.006 |
| <i>Plantago major</i> L. | | 197 ± 45 | 61 ± 17 | 0.06 ± 0.01 |
| Back-ground | <i>Centaurea scabiosa</i> L. | 10 ± 4 | 1 ^a | 0.0026 ± 0.0005 |
| | <i>Melandryum album</i> (Mill.) Garke | 100 ± 25 | 21 ± 9 | 0.0288 ± 0.0098 |
| | <i>Stellaria graminea</i> (L.) | 10 ± 5 | 11 ± 4 | 0.0052 ± 0.0023 |
| | <i>Urtica dioica</i> L. | 11 ± 4 | 1 ^a | 0.0026 ± 0.00047 |
| | <i>Sanguisorba officinalis</i> L. | 10 ± 4 | 1 ^a | 0.0026 ± 0.00052 |
| | <i>Taraxacum officinale</i> s.l. | 12 ± 5 | 1 ^a | 0.0028 ± 0.00058 |
| | <i>Plantago major</i> L. | 10 ± 3 | 1 ^a | 0.0026 ± 0.00039 |
| | <i>Leonurus quinquelobatus</i> Gilib. (<i>L. vilosus</i> Desf.) | 9 ± 4 | 6 ± 2 | 0.0036 ± 0.0014 |
| | <i>Rumex confertus</i> Willd. | 5 ± 3 | 1 ^a | 0.0014 ± 0.00042 |

^a DL (detection limit).

different concentrations of ⁹⁰Sr and ¹³⁷Cs in soils of micro-localities. Besides, it may be caused by peculiarities of the radionuclides' accumulation in different plant species. One can see that maximum absorbed dose was found in *Sanguisorba officinalis* L., as ⁹⁰Sr concentration in the soil upper layer of this habitat averaged 300 kBq/kg. The lowest absorbed dose has been registered in *Plantago major* L., where ⁹⁰Sr concentration in the soil of micro-locality equals 89 kBq/kg. Among all studied species, *Urtica dioica* L. accumulated the greatest amounts of ⁹⁰Sr and ¹³⁷Cs (Pozolotina et al., 2005).

To calculate the dose loads, we took into account the absorbed doses provided by the radionuclides incorporated in seeds. Besides, dose rate of external γ -radiation was measured using a dose-meter DRG-01T. Accepting to coefficient of absorption for biological tissues (0.96), absorbed doses by the embryos per month were calculated. They were $6.9\text{--}7.2 \times 10^{-2}$ mGy on the background area, $10.4\text{--}13.8 \times 10^{-2}$ on the buffer plots, and $13.8\text{--}117.5 \times 10^{-2}$ on the impact territories. Absorbed doses within the EURT might be referred as the "low doses" category. Nevertheless, due to long-termed irradiation, they were shown to cause different biological effects described in a series of publications (Shevchenko et al., 2000; Pozolotina et al., 2005; Ul'yanova and Pozolotina, 2006).

4. Conclusion

Terrestrial ecosystems within EURT zone were examined regarding their radioecological conditions. Within the EURT central axis, about 100 km long, the radioactive contamination of soils decreased with the distance from the accident point and fairly described by an exponential function. Maximum stocks of ⁹⁰Sr (25 000 kBq/m²) and ¹³⁷Cs (670 kBq/m²) have been registered within the soil cover at a distance of about 6 km away from the accident epicenter. At the western and eastern periphery of the trace area within EUSR, ⁹⁰Sr contents in soils made 30–80 kBq/m², and beyond the reserve boundaries the values slightly exceeded the background level.

Radionuclide concentrations were examined in seeds of some herbs growing within the EURT zone. It was shown that in the seeds sampled from impact populations, ⁹⁰Sr values were significantly higher than writing as the background level (by 2–4 orders of magnitude). These plants revealed ⁹⁰Sr discrimination in the link: vegetative mass – seeds. Dose loads upon seed embryos in impact plants exceeded those of the background level by 3–4 orders of magnitude; they were about 10 times higher than the corresponding values in the buffer zone plants.

Acknowledgments

This work was supported by the Russian Foundation for Basic Research, projects nos. 07-05-00070 and 07-04-96098.

References

- Aarkrog, A., Dahlgaard, H., Nielsen, S.P., Trapeznikov, A.V., Molchanova, I.V., Pozolotina, V.N., Karavaeva, E.N., Yushkov, P.I., Polikarpov, G.G., 1997. Radioactive inventories from the Kyshtym and Karachay accident: estimates based on soil samples collected in the South Urals (1990–1995). *Science of the Total Environment* 201, 137–154.
- Eastern Urals Radioactive Trace, 2000. Rehabilitation Problems of Population and Territories of Sverdlovsk Region. Ural Division of RAS Publisher, Yekaterinburg (in Russian).
- Experimental Scientific Research Station of Production Association "Mayak", 2005. Radiation Publishing Center of VRB, Ozersk.
- Gorshkov, G.V., 1967. Penetrating Radiations from Radioactive Sources. Nauka, Leningrad (in Russian).
- Karavaeva, E.N., Molchanova, I.V., 1979. Role of some categories of soil moisture in the vertical migration of ⁹⁰Sr and ¹³⁷Cs in the soil. *The Soviet Journal of Ecology* 10, 35–48.
- Klement A.W., Lytle C.F., Schultz V., 1968. Russian radioecology: a bibliography of Soviet publications with citations of English translations and abstract. AEC Report TLD-3915. USA.
- Molchanova, I.V., Karavaeva, E.N., 2001. Ecogeochemical Aspects of Radionuclide Migration in the Soil and Plant Covers. Ural Branch RAS, Yekaterinburg (in Russian).
- Nikipelov, B.V., Nikiforov, A.S., Kedrovsky, O.L., Strakhov, M.V., Drozhko, E.G., 1990. Practical Rehabilitation of Territories Contaminated as a Result of Implementation of Nuclear Material Production Defence Programmes. Oak Ridge National Laboratory, USA.
- Pozolotina, V.N., 2003. Remote Consequences of Radiation Impact on Plants. Akademkniga, Yekaterinburg (in Russian).

- Pozolotina, V.N., Molchanova, I.V., Mihkaylovskaya, L.N., Ul'yanova, E.V., 2005. Recent levels of radionuclide contamination in the Eastern Ural radioactive trace and biological effects in local populations of *Plantago major* L. Russian Journal of Ecology 36, 320–328.
- Shevchenko, V.A., Kalchenko, V.A., Abramov, V.V., Rubanovich, A.V., Shevchenko, V.V., Grinikh, L.I., 2000. Genetic of populations in the zones of the Kyshtym and Chernobyl accidents. Low Doses of Radiation: Are they Dangerous? Publishers Inc., Huntington, New York.
- Ul'yanova, E.V., Pozolotina, V.N., 2006. Clonal diversity and rare phenes in *Taraxacum officinale* s.l. Coenopopulations from the East-Ural radioactive trace zone. Doklady Biological Sciences 406, 106–108.
- Yachmenev, V.A., Isaeva, L.N., 1996. Environmental monitoring in the vicinity of the Mayak atomic facility. Health Physics 71, 61–69.
- Zheleznov, A.V., Zheleznova, N.B., Smetanin, N.I., Sukhanovskaya, V.S., 2002. Intrapopulation variability of some meadow plant species in respect of their ability to accumulate ^{90}Sr . Russian Journal of Genetics 38, 521–525.