Radioprotection, vol. **44**, n° 5 (2009) 371–375
© EDP Sciences, 2009
DOI: 10.1051/radiopro/20095071

Peculiarities of the technogenical radionuclides transfer from soils into plants in the radioactive contaminated areas

Y.N. Karavaeva, I.V. Molchanova and L.N. Mikhailovskaya

Institute of Plant and Animal Ecology, Russian Academy of Sciences, Urals Division, 8 Marta, 202, 620144 Ekaterinburg, Russia
e-mail: molchanova_i_v@mail.ru

Abstract. The peculiarities of ^{90}Sr, ^{137}Cs, ^{239,240}Pu accumulation by some species of herbaceous plants in radioactive contaminated areas of nuclear enterprises in Russia have been investigated. Gas and aerosol discharge from Beloyarskaya NPP (which operates for more than 30 years) did not make considerable contamination of soil-vegetational cover in the 30-km zone. In close proximity to the epicenter of nuclear accident of 1957 in the Urals the concentration of the main contaminant (^{90}Sr) in the plants reached maximal value (tens and hundred of thousands of Bq kg_{dw}^{-1} depending on species belonging of plant). The concentration of ^{90}Sr in aboveground phytomass of plants growing within floodplain of the Techa river ecosystems is thousands of Bq kg_{dw}^{-1}; at the same time, the ^{137}Cs concentration is units of Bq kg_{dw}^{-1}. The absence of direct dependence between the radionuclides content in the soils and their concentration in aboveground phytomass is noticeable. The values of the radionuclides transfer coefficients in the plants were estimated as well as the content in the soils of their water-soluble and firmly fixed physic-chemical forms. The radionuclides have been ranked into a row according to their mobility in observed soils and biogeocenotical soil-plant link.

1. INTRODUCTION

Large-scale nuclear weapon tests, intensive development of nuclear technologies and nuclear industry were accompanied by massed entering of artificial radionuclides into the biosphere. Radioactive materials enter the environment through the regular operation of nuclear plants. Also nuclear accidents and incidents, that take place in nuclear-fuel cycle plants, make an important contribution into radioactive contamination of certain areas. Thus, emergency discharge of liquid highly radioactive waste from nuclear enterprise “MAYAK” (Chelyabinsk region) in the Techa river from 1949 till 1952 caused the contamination of its floodplain all over its stretch. An accident in the same enterprise in 1957 resulted in atmosphere contamination by 74 PBq of radioactive substances that formed the Eastern Urals Radioactive Trace (EURT), 23 thousand of square kilometers in area. One of the main objects of monitoring in the regions with nuclear-fuel cycle plants is the soil-vegetational cover. When radionuclides come from the atmosphere, the most important is airborne contamination of aboveground parts of plants. Then a considerable part of radionuclides, primarily detained by the vegetational cover, enter the soil and form the pool, available for plant roots.

The authors have been conducting the radioecological investigations of soil-vegetational cover in the 30-km zone of regularly operating Beloyarskaya nuclear power plant (NPP). It is situated in the Middle Urals, 60 km eastward of Ekaterinburg. Besides, the long-term processes of migration and redistribution of radionuclides on the Eastern Urals Radioactive Trace territory, as well as in the Techa river floodplain were examined. The data, characterizing peculiarities of ^{90}Sr, ^{137}Cs and ^{239,240}Pu accumulation in the aboveground phytomass of the herbs, growing at the examined impact territories, are presented.

2. MATERIALS AND METHODS

In the 30-km zone of surveillance of Beloyarskaya NPP the samples of plants and soils from plots, laid on various distances from the plant, taking into consideration the wind rose, were collected.

Article published by EDP Sciences and available at http://www.radioprotection.org
or http://dx.doi.org/10.1051/radiopro/20095071

The pre-dominant soil's type is brown forest soil (pH of water extract is 5.7; concentration of carbon, 7.0%; the content of exchangeable bases, 21.5 mg-eq/100g). Within the limits of the central axis of the Eastern Urals Radioactive Trace samples of soil-vegetational cover were taken at the different distances from the nuclear enterprise "MAYAK". The soil cover is represented by leached chernozem (pH of water extract is 6.2; concentration of carbon, 22.0%; the content of exchangeable bases, 20.5 mg-eq/100g). In the Techa river floodplain meadow-alluvial soils are predominantly being formed (pH of water extract is 6.8; concentration of carbon, 6.0%; the content of exchangeable bases, 39.0 mg-eq/100g). The samples of plants and soils were taken at a distance of 230 km from "MAYAK". The aboveground phytomass of the following species of herbaceous plants were examined: milfoil (*Achillea millefolium* L.), common tansy (*Tanacetum vulgare* L.), burdock (*Arctium tomentosum* L.), common nettle (*Urtica dioica* L.), and common wormwood (*Artemisia absinthium* L.). The same species were simultaneously collected at control plot on the territory of Biosphere State Reserve (Chelyabinsk region).

Sampling of soil-vegetational cover was being made in summer seasons during 2002–006 yr. On all plots under study the herbaceous plants and soil samples were collected from four fixed areas. In the places of plants collection after digging a pit, the layers of soil (5 cm thick) were cut horizontally to a depth of 30 cm taking into account the area of the soil layers. To exclude airborne contamination, the aboveground parts of herbs were washed by running water. The collected samples of plants and soils were dried up to the air-dry weight. Then the samples of plants were ashed at 400 °C, and the soils were pulverized and riddled through a sieve with 1 mm meshes. The water soluble physic-chemical form of radionuclides accumulated in the soils was educed with distilled water; the ratio between the solid and liquid phases was 1:5. The exchangeable form was educed with 1N CH_3COONH_4. The acid dissolved form – with 1N HCL (ratio 1:10). The firmly fixed form included radionuclides which remained in the soil after these procedures. The content of ^{90}Sr in the samples was estimated radiochemically by its daughter ^{90}Y. The isotopes of Pu were estimated as well radiochemically with using ion-exchange columns and electrochemical deposition on stainless steel discs. The content of ^{137}Cs was measured by Ge gamma-spectroscopy. The limit of ^{90}Sr, ^{137}Cs detection made 1 Bq kg_{dw}^{-1}, and ^{239,240}Pu – 0.1 Bq kg_{dw}^{-1}. The content of ^{238}Pu has lower limit of detection as a rule. Error of observation did not exceed 20%.

3. RESULTS

The results of the radionuclides detection in soils in the inspected territories have shown in the Table 1. The content of ^{90}Sr, ^{239,240}Pu on the control plot is equal, and of ^{137}Cs comes nearer to the global background level [1]. On the basis of the data given, it is possible to judge of the degree of contamination of the places where the plants grew. It appeared that on the territory of Eastern Urals Radioactive Trace the contamination density of the soil cover by long-living radionuclides decreased with the distance from the accident epicenter. Early it was noticed that this decrease is approximated by exponential function [2]. The content of ^{90}Sr and ^{137}Cs in the Techa river floodplain soils is higher than the same values on the control plot by a factor of 20–120, as the content of ^{239,240}Pu comes nearer to that on the control plot. The gas and aerosol discharge from the regular operation of Beloyarskaya NPP

Table 1. Radionuclides in the soils from investigated areas.

Sampling plot. Distance from the source of contamination, km	^{90}Sr	^{137}Cs	^{239,240}Pu
Control plot	1.6 ± 0.15	3.0 ± 0.5	0.096 ± 0.028
Beloyarskaya NPP	2.0 ± 0.4	6.0 ± 0.7	0.113 ± 0.058
EURT, 5	29300 ± 2500	860 ± 120	26.5 ± 3.52
90	60 ± 9.0	9.3 ± 1.0	0.168 ± 0.36
Floodplain of the Techa river, 230	190 ± 20.0	84 ± 10.0	0.145 ± 0.029

Inventories given in kBq $m^{-2} \pm 1\sigma$.

(which operates for more than 30 years) had not resulted in the considerable, in comparison with the background, increase of the radionuclides content in the soil of the 30-km monitored zone.

In four species (*Achillea millefolium, Arctium tomentosum, Tanacetum vulgare, Artemisia absinthium*) growing in the 30-km zone near Beloyarskaya NPP the content of the radionuclides under consideration a little bit differs from those of the control plot (Table 2). In the EURT areas in close proximity to epicenter of the accident the concentration of the main contaminant (^{90}Sr) in the plants reached thousands of Bq kg_{dw}^{-1}. It decreases with the distance from epicenter of the accident by a factor of 70–400. The maximum concentration of ^{137}Cs in these herbaceous plants is 960 Bq kg_{dw}^{-1} and minimum one is 1.4–2.7 Bq kg_{dw}^{-1} at the distance of 5 and 90 km accordingly. The high concentrations of ^{90}Sr are revealed in the plants collected in the Techa river floodplain. The low values of ^{137}Cs concentration in the plants growing there at relative high its content in floodplain soils attract attention. Depending on a specific belonging of plants, their ability to concentrate the investigated radionuclides may vary on the order of magnitudes. It is interested to mention, that maximum concentration of ^{90}Sr in *Urtica dioica* is noticed.

Table 2. ^{90}Sr and ^{137}Cs in herbs from areas of operating nuclear-fuel cycle facilities.

Plant species	30-km zone of Beloyarskaya NPP	EURT, distance from the source of contamination, km		Floodplain of the Techa river	Control plot
		5	90		
Achillea Millefolium	$\frac{10.0 \pm 1.0}{17.0 \pm 1.5}$[1]	$\frac{20190 \pm 2100}{190 \pm 56}$	$\frac{135 \pm 1.0}{1.4 \pm 0.1}$	$\frac{980 \pm 95}{1.0 \pm 0.1}$	$\frac{6.0 \pm 1.8}{10.0 \pm 2.8}$
Arctium tomentosum	$\frac{5.0 \pm 0.5}{8.0 \pm 0.2}$	$\frac{51230 \pm 4850}{560 \pm 96}$	$\frac{323 \pm 87.0}{\text{BDL}}$	$\frac{1030 \pm 120}{2.0 \pm 0.2}$	$\frac{6.0 \pm 0.2}{5.0 \pm 1.6}$
Urtica dioica	$\frac{46.0 \pm 23.0}{30.0 \pm 20.0}$	$\frac{134150 \pm 4270}{960 \pm 120}$	$\frac{347 \pm 75.0}{2.7 \pm 1.4}$	$\frac{3680 \pm 300}{4.4 \pm 0.4}$	$\frac{15.0 \pm 3.9}{12.0 \pm 1.5}$
Tanacetum vulgare	$\frac{4.0 \pm 0.4}{5.0 \pm 0.6}$	$\frac{32750 \pm 2387}{185 \pm 40}$	$\frac{80 \pm 11.0}{1.5 \pm 0.8}$	$\frac{1460 \pm 130}{1.7 \pm 0.2}$	$\frac{9.0 \pm 4.3}{11.0 \pm 5.1}$
Artemisia absinthium	$\frac{4.0 \pm 0.8}{1.4 \pm 0.9}$	$\frac{15900 \pm 4500}{320 \pm 55}$	$\frac{226 \pm 50.0}{1.6 \pm 0.5}$	$\frac{1040 \pm 90}{2.3 \pm 0.2}$	$\frac{3.8 \pm 0.9}{2.5 \pm 0.7}$

Concentration of activities given in Bq $kg_{dw}^{-1} \pm 1\sigma$.
BDL: below detection limit.

For the quantitative estimation of migration ability of radionuclides, a conventional in radioecology transfer coefficient (TC) was used. It was calculated as the ratio of nuclide concentration in the dry mass (Bq kg^{-1}) to its content in the soil (kBq m^{-2}) [3]. The transfer coefficients values of radionuclides in herbs growing on contaminated soils are shown in the Table 3. In the EURT areas in close proximity to epicenter of the accident the TC of ^{90}Sr in four species under consideration are characterized by minimal values. It may be proposed that the plants have acquired this feature in the course of selection under prolonged radiation loads [4]. In herbs from other contaminated areas as control plot the TC of ^{90}Sr vary from $1.3 \cdot 10^{-3}$ to $7.7 \cdot 10^{-3}$. In accordance with high concentration of ^{90}Sr in *Urtica dioica* the TC for this plant in all cases are higher than those in other species of the herbaceous plants. In herbs collected in the floodplain of the Techa river the TC for ^{137}Cs are lower by a factor 10–280 than those in other investigated areas. As a rule transfer coefficients of ^{90}Sr in herbaceous plants are higher than those of ^{137}Cs. Minimal differences between TC of ^{90}Sr and ^{137}Cs (2–10 times) were noticed both for control plot and for the 30-km zone of surveillance of Beloyarskaya NPP. Maximal differences (200–500 times) are in the floodplain soils of the Techa River. We may explain this fact with the differences in content of free (water soluble + exchangeable) and fixed forms of the radionuclides in the soils (Table 4).

[1] Here and in the Table 3 ^{90}Sr – above the line, ^{137}Cs – underline the line.

Table 3. The radionuclides transfer coefficients into herbs from soils in the investigated areas, $m^2 kg^{-1} 10^{-3}$.

Plant species	30-km zone of Beloyarskaya NPP	EURT, distance from the source of contamination, km		Floodplain of the Techa river	Control plot
		5	90		
Achillea millefolium	5.0 / 2.8	0.7 / 0.2	2.2 / 0.1	5.1 / 0.01	3.7 / 2.5
Arctium tomentosum	2.5 / 1.3	1.7 / 0.6	5.4 / BLD	5.4 / 0.02	3.7 / 1.2
Urtica dioica	23.0 / 5.0	4.5 / 1.1	5.8 / 0.3	19.4 / 0.05	9.4 / 3.0
Tanacetum vulgare	2.0 / 0.8	1.1 / 0.2	1.3 / 0.2	7.7 / 0.02	5.6 / 2.7
Artemisia absinthium	2.0 / 0.2	0.5 / 0.4	3.8 / 0.2	5.4 / 0.02	2.4 / 0.6

Table 4. Physic-chemical forms of ^{90}Sr and ^{137}Cs in the soil, %.

Sampling plot	Radionuclides	Physic-chemical forms	
		Free	Fixed
30-km zone of Beloyarskaya NPP	^{90}Sr	67.6 ± 5.9	BDL
	^{137}Cs	17.1 ± 2.5	64.9 ± 6.9
Floodplain of the Techa River	^{90}Sr	69.1 ± 4.0	4.2 ± 0.2
	^{137}Cs	2.5 ± 0.2	95.2 ± 0.2

BDL: below detection limit.

Table 5. The accumulation of $^{239,240}Pu$ in the aboveground phytomass of *Urtica dioica* from areas of operating nuclear-fuel cycle facilities.

30-km zone of Beloyarskaya NPP	EURT, distance from the source of contamination, km		Floodplain of the Techa river	Control plot
	5	90		
0.038 ± 0.006[2] / 0.330	0.048 ± 0.009 / 0.002	0.021 ± 0.002 / 0.120	0.031 ± 0.003 / 0.210	0.038 ± 0.007 / 0.390

Concentration of $^{239,240}Pu$ expressed in Bq $kg_{dw}^{-1} \pm 1\sigma$.
TC expressed in $m^2\,kg^{-1} 10^{-3}$.

The estimates of the $^{239,240}Pu$ accumulation in *Urtica dioica* from areas of operating nuclear-fuel cycle facilities and control plot are shown in the Table 5. There were not revealed any statistically significant differences of the $^{239,240}Pu$ concentration in *Urtica dioica* both for different contaminated plots and for the control plot. On an average, it is equal to $0.035\,kg_{dw}^{-1}$. The lowest values of the TC of $^{239,240}Pu$ reflected its minimal mobility in soil-plant link are noticed on the territory EURT, 5 km far from the source of contamination.

4. CONCLUSION

This research gives a set of data on the radioactive levels in the soils of the radioactive contaminated areas of nuclear enterprises in the Urals (Russia). Furthermore, it indicates the importance of investigation of the peculiarities of technogenical radionuclides accumulation for different species of herbaceous plants. Contaminated areas under investigation have been ranked into a row according

[2]Concentration of $^{239,240}Pu$ – above the line, TC – under the line.

to radionuclide load: Eastern Urals Radioactive Trace, 5 km far from the source of contamination > floodplain of the Techa River, 230 km far from the source of contamination > Eastern Urals Radioactive Trace, 90 km far from the source of contamination > the 30-km zone of Beloyarskaya NPP. It is known, that physic-chemical state of radionuclides in soils and their availability for plants are mainly determined by genesis of radioactive discharge, by own properties of the radionuclides and by the peculiarities of soils and climate of the region. Combination of these factors causes the difference in radionuclides' entering into plants, growing at the examined territories. The present investigation shows, that the concentration of the radionuclides in the investigated plants first of all depends on the level of their content in the soil. So the concentration of ^{90}Sr in the plants tested in the 30-km zone of Beloyarskaya NPP is low and varies within the limits from 4 to 46 Bq kg_{dw}^{-1}, and that of ^{137}Cs – from 1.4 to 30 Bq kg_{dw}^{-1}. The concentration of ^{90}Sr in aboveground phytomass of the plants growing within floodplain of the Techa river ecosystems is thousands of Bq kg_{dw}^{-1}, the ^{137}Cs concentration is units of Bq kg_{dw}^{-}. At the same time, in close proximity to the accident epicenter the concentration of the main contaminant (^{90}Sr) of EURT in the plants reached maximal value (tens and hundred of thousands of Bq kg_{dw}^{-1}). The ^{239,240}Pu concentration in aboveground phytomass in one of the investigated herbaceous plants (*Urtica dioica*) is hundredth parts of Bq kg_{dw}^{-1}. However, it was noticed that the transfer coefficients of ^{90}Sr and ^{239,240}Pu in different herbaceous plant species growing in the most contaminated areas of EURT are lower than those for other ones.

Transformation of the physic-chemical forms of radionuclides in soils is very complicated in its character and, as a rule, is accompanied by fluctuation of the depot of the mobile combinations. Low content of the mobile forms of ^{137}Cs, found in the soils of the Techa river floodplain, reflects a minimum relative availability of this radionuclide for the plants. In the floodplain soils the transfer coefficients of ^{137}Cs in herbs are less than those of ^{90}Sr by a factor of 200–500. In accordance with mobility in observed soils and biogeocenotical soil-plant link the radionuclides have been ranked into a row: ^{90}Sr > ^{137}Cs > ^{239,240}Pu. In the group of plants under study from the areas of operating nuclear-fuel cycle facilities in aboveground phytomass of *Urtica dioica* the most quantity of ^{90}Sr is noticed.

Acknowledgments

This study was supported by the Russian Foundation for Basic Research, project no. 07-05-00070.

References

[1] UNSCEAR. Ionizing radiation: sources and biological effects (United Nations, New York, 1993).
[2] A. Aarkrog, H. Dahlgaard, S. Nielsen, The Science of the Total Environment **201**, 137 (1997).
[3] R.M. Alexakhin, M.A. Naryshkin, in Migration of Radionuclides in the Forest Biogeocenoses, edited by A.A. Molchanov (Nauka, Moscow, 1977).
[4] V.N. Pozolotina, I.V. Molchanova, E.N. Karavaeva, in *Proceedings of the International Conference on Biogeochemistry of Trace Elements: Environmental Protection, Remediation and Human Health, Bei Jing, 2007,* edited by Y. Zhu, N. Lepp and R. Naidu (Tainghua University Press, Bei Jing, 2007), p. 437.