Radioprotection, vol. 46, n° 6 (2011) S25–S28

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DOI: 10.1051/radiopro/20116476s

Accumulation of ⁶⁰Co, ⁹⁰Sr, ¹³⁷Cs, ²³⁸U and transuranic elements by berry shrubs in forest ecosystems of the Yenisei river basin

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Abstract. The study investigates accumulation of radionuclides in 5 species of berry shrubs in forests of the Yenisei River basin (the Krasnoyarskii Krai, Russia). Activity concentrations of 60 Co, 90 Sr, 137 Cs, 238 U and transuranic elements in aboveground organs of the shrubs have been determined and their transfer factors calculated. Analysis of the calculated TFs shows that shrubs concentrate 90 Sr more effectively than other study elements (TF = 0.25–1.9). The highest concentration of 137 Cs is recorded in the berries and leaves of *Ribes nigrum* and *Rubus idaeus*. For these species, the range of 137 Cs TFs (0.006–0.027) overlaps with the range of 238 U TFs (0.004–0.018). For the aboveground parts of *Ribes nigrum*, TFs for transuranic radionuclides (0.01–0.056) are similar to or higher than the TFs for 238 U and 137 Cs.

1. INTRODUCTION

Among terrestrial ecosystems contaminated with artificial radionuclides is the portion of the Yenisei River basin affected by the operation of the Mining-and-Chemical Combine (MCC), which produced weapons-grade plutonium. There are very few data on radionuclide accumulation in forest ecosystems of these areas, and they mainly concern contamination of trees, as the main object of forest management. Other components of forest ecosystems, such as shrubs, which are also involved in accumulation and redistribution of radionuclides, have not been investigated until now [1, 2].

The purpose of this study is to estimate the transfer of artificial radionuclides, including transuranic ones, into berry shrubs via roots.

2. METHODS

We studied radionuclide accumulation by the berry shrubs *Ribes hispidulum* (Jancz.) Pojark, *Ribes nigrum* L., *Rosa majalis* Herrm., *Rubus idaeus* L., and *Viburnum opulus* L. growing in the forests. Plant and soil samples were collected in the areas contaminated by radionuclides contained either in MCC aerosol releases or in its liquid effluents. Samples were collected from the following sites (Fig. 1): the background sampling site "Krasnoyarsk", with radioactivity due to global fallouts only; "Zheleznogorsk" (66 km downstream of Krasnoyarsk), "Atamanovo (village)" (88 km), and "Balchug (village)" (98 km) – the sites contaminated due to global fallouts and MCC aerosol discharges; and "Atamanovo (island)" and "Balchug (island)" – the Yenisei floodplain sites contaminated by water-transported artificial radionuclides. Investigations were performed from 2004 to 2010.

Samples of berry shrubs collected during the fruit-bearing season were split into branches, leaves, and berries, which were dried at 65°C. The dry samples were weighed and ashed in a muffle furnace at 450°C. The activity concentrations of the γ -emitting nuclides in the samples were measured on a Canberra γ -spectrometer (USA) coupled to a GX2320 23% hyper-pure germanium detector. The γ -spectra were processed using the CANBERRA GENIE-2000 software (USA). Concentration of 238 U



Figure 1. Diagrammatic map of the Krasnoyarsk Territory (Russia), showing villages and towns near which samples were collected.

in the samples was determined using the instrumental neutron-activation analysis at Tomsk Polytechnic University (Tomsk, Russia).

Radiochemical investigations of the samples of shrub species and the soil collected from a floodplain site ("Atamanovo (island)") to determine α -emitting isotopes (238 Pu, 239,240 Pu, 241 Am, and 243,244 Cm) and a β -emitting isotope, 90 Sr, were performed at the Centre for Information Processes and Technologies of the "RADON" Scientific & Industrial Association (Moscow). Methods and instrumentation for the analysis of samples for radiostrontium and transuranium elements were described elsewhere [3]. All activity concentrations of radionuclides were calculated for air dry mass of the samples.

3. RESULTS AND DISCUSSION

The contamination of the soils in the study area is not uniform owing to the complex topography of the area and the various routes of radionuclide entry. To date, the following radionuclides are recorded in the floodplain soils downstream of the MCC: 60 Co, 137 Cs, 152,154 Eu, 90 Sr, uranium, and isotopes of transuranic elements: 238 Pu, 239,240 Pu, 241 Am, 243,244 Cm [3]. 137 Cs concentration in the samples of *Rosa majalis*, *Rubus idaeus*, and *Viburnum opulus* collected at the study sites ranged from 0.2 Bq/kg to 64 Bq/kg. Samples of aboveground phytomass from the floodplain soils contained 60 Co – up to 8.4 Bq/kg; no other artificial γ -emitting radionuclides were found to accumulate in the phytomass. 137 Cs alone was detected in the study berry shrubs collected from the sites contaminated by aerosol-delivered radionuclides, and its activity was near the limit of detection. In *Ribes nigrum*, *Rosa majalis*, and *Viburnum opulus*, the maximum activity concentration of 137 Cs was recorded in the berries – up to 64 Bq/kg. In *Ribes hispidulum* and *Rubus idaeus* leaves, the highest 137 Cs activity concentration was 51 Bq/kg.

The shrubs *Rubus idaeus* and *Ribes nigrum*, which accumulated the greatest amounts of ¹³⁷Cs, were used in a detailed study of radionuclide accumulation, employing radiochemical analysis [2, 3]. Analysis of the aboveground parts of *Ribes nigrum* showed that activity concentrations of ⁶⁰Co and ¹³⁷Cs were 1.5–3 times higher in the leaves than in the branches, which can be accounted for by conductive function of branches during the period of active plant growth. The distribution of ¹³⁷Cs in the organs of *Rubus idaeus* is similar to its distribution in *Ribes nigrum*. Radiochemical analysis showed that in *Ribes nigrum* ⁹⁰Sr was mainly accumulated in the leaves (up to 60 Bq/kg), while the berries contained the lowest ⁹⁰Sr concentrations – not higher than 3.8 Bq/kg [2].

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Table 1. Transfer factors of radionuclides to different parts of the shrubs for the sites contaminated by water-delivered radionuclides.

TFs	⁴⁰ K	⁶⁰ Co	⁹⁰ Sr	¹³⁷ Cs	²³⁸ U		
Ribes nigrum							
Branches	0.48	0.01	0.9	0.009	0.018		
Leaves	0.94	0.02	1.9	0.029	0.011		
Berries	1.19	<dl< td=""><td>0.25</td><td>0.027</td><td>0.004</td></dl<>	0.25	0.027	0.004		
Rubus idaeus							
Branches	0.69	<dl< td=""><td>n.d.</td><td>0.006</td><td>0.008</td></dl<>	n.d.	0.006	0.008		
Leaves	1.34	<dl< td=""><td>n.d.</td><td>0.008</td><td><dl< td=""></dl<></td></dl<>	n.d.	0.008	<dl< td=""></dl<>		
Berries	1.20	<dl< td=""><td>n.d.</td><td>0.010</td><td><dl< td=""></dl<></td></dl<>	n.d.	0.010	<dl< td=""></dl<>		

Note: DL – detection limit; n.d. – not determined.

The Yenisei River floodplain is contaminated by artificial radionuclides (60 Co, 90 Sr, 137 Cs, etc.), which are either products of stable isotope activation or 238 U fission products. 238 U and transuranic elements (Pu, Am, and Cm isotopes) may enter the environment together with artificial radionuclides. Measurements of 238 U in *Rubus idaeus* and *Ribes nigrum* showed that in *Ribes nigrum*, this isotope was mainly accumulated in the branches and leaves. 238 U concentration in the aboveground organs ranged from 0.012 mg/kg to 0.14 mg/kg. Samples collected in the most heavily contaminated floodplain site, "Atamanovo (island)", were analyzed for transuranic elements [3]. In the aboveground phytomass of *Ribes nigrum*, the highest concentrations of isotopes of transuranic elements were recorded in the leaves (up to 0.4 Bq/kg of 239,240 Pu and up to 0.05 Bq/kg of 243,244 Cm) and berries (up to 0.88 Bq/kg of 239,240 Pu and up to 0.03 Bq/kg of 243,244 Cm).

The capacities of the shrubs for accumulation of radionuclides in their biomass were estimated based on the calculated transfer factors (TFs). The leaves and berries of *Rubus idaeus* and *Ribes nigrum* had the highest TFs for 137 Cs (up to 0.029) (Table 1). The TFs for 137 Cs obtained for the berries of these species corresponded to the previous data of field investigations (0.020–0.034) [2]. In the branches, 137 Cs TF was not higher than 0.009. Of the studied isotopes, 90 Sr is the element best accumulated by *Ribes nigrum*: its TFs reach 1.9 in the leaves, 0.9 in the branches, and 0.25 in the berries, and they are similar to 137 Cs TFs in the mushrooms collected in the same area [2, 4]. The comparatively low 90 Sr TFs in the berries of *Ribes nigrum* correspond to the data on Chernobyl fallouts (0.26-0.66) [5]. Based on the calculated TFs, we constructed the following sequences of 137 Cs and 90 Sr accumulation by different organs of the shrubs: "branches < leaves \leq berries" and "berries < branches < leaves", respectively [2]. The highest 238 U TFs in the shrubs were recorded in the branches of *Ribes nigrum* and *Rubus idaeus*. The range of 238 U TFs (0.004–0.018) overlaps the range of 137 Cs TFs (0.006–0.027) (Table 1).

In the aboveground phytomass of *Ribes nigrum*, transuranic elements are mostly accumulated in the leaves and berries. Based on the calculated TFs (Table 2), we obtained the following sequences of ^{243,244}Cm and ^{239,240}Pu accumulation by different organs of the blackcurrant shrubs: "branches < berries < leaves" and "branches < leaves < berries", respectively. For the aboveground parts of *Ribes nigrum*, TFs for transuranic radionuclides (0.01-0.056) are similar to or higher than the TFs for ²³⁸U and ¹³⁷Cs. The previously published data [6, 7] suggest that the TFs for Pu and Am are an order of magnitude lower than the TFs for ¹³⁷Cs. The higher TFs of transuranic elements obtained in our study may be caused by the different routes of entry of transuranic radionuclides to the soil in the study area.

Thus, the non-uniform distribution of radionuclides in the soil and the comparable TF values for ¹³⁷Cs and transuranic radionuclides suggest a conclusion that plants growing at certain sites can be expected to contain higher activity concentrations of transuranic elements. Also, the behavior of transuranic elements in ecosystems is not sufficiently understood so far. Therefore, the problems in their

Table 2. Transfer factors of transuranic radionuclides to different parts of *Ribes nigrum* for the site "Atamanova (island)."

TFs	^{239,240} Pu	²⁴¹ Am	^{243,244} Cm
Roots	0.027	0.051	0.073
Branches	0.010	0.025	0.018
Leaves	0.026	0.051	0.045
Berries	0.056	<dl< td=""><td>0.027</td></dl<>	0.027

determination should not impede the progress of the studies on the distribution of these elements in ecosystems.

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