

Accumulation of artificial radionuclides by edible wild mushrooms and berries in the forests of the central part of the Krasnoyarskii Krai

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Abstract. The study addresses accumulation of radionuclides by mushrooms and berry shrubs from the forest soil in the 30-km area around the MCC (the Krasnoyarskii Krai, Russia). The *Suillus granulatus* and *S. luteus* mushrooms have been found to be the best bioindicators in this area. The level of ^{137}Cs concentration in these species can reach 10 kBq/kg dry mass. The ^{137}Cs transfer factor (TF) by the mushrooms varies 5-fold among the sites that receive radionuclides from different sources. The level of radionuclide activities accumulated by berry shrubs is 2–3 orders of magnitude lower than the activities accumulated by mushrooms. No relationship has been found between the TFs of radionuclides to the shrubs and the pathway via which the radionuclides enter the soil. The contribution of the dose due to intake of mushrooms and berries to the annual effective dose of internal exposure to residents does not exceed 150 $\mu\text{Sv}/\text{year}$ and 1.4 $\mu\text{Sv}/\text{year}$, respectively, estimated from the registered levels of accumulated ^{137}Cs .

1. INTRODUCTION

The main contributors to environmental radiation on the territory of the Krasnoyarskii Krai are natural background radiation, global fallouts of radionuclides, and operation of nuclear facilities. The Mining-and-Chemical Combine (MCC) of Rosatom, situated at Zheleznogorsk, on the bank of the Yenisei River, includes a reactor plant and a radiochemical plant, which discharge radionuclides as aerosols and liquid effluents. The Yenisei floodplain and the area around the MCC are contaminated with radionuclides released by the MCC, which has been in operation for many years [1]. There are just fragmentary data on radionuclide accumulation in forest ecosystems of these regions, which mostly concern woody plants, as the main object of forest management. The published data on other components of forest ecosystems, such as mushrooms and berry shrubs, which can accumulate high levels of radionuclides, are scarce [2, 3]. Residents consuming these mushrooms and berries can be subjected to additional exposure.

The purpose of the study was to assess accumulation of artificial radionuclides by mushrooms and shrubs in the forest ecosystems of the Krasnoyarskii Krai situated in the area affected by the MCC operation.

2. MATERIALS AND METHODS

2.1 Sample collection

We studied radionuclide accumulation by the soil, 12 mushroom species and 5 berry shrub species growing in the forests. 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): “Krasnoyarsk” – the background sampling site, with radioactivity due to global fallouts only; “Zheleznogorsk” (66 km downstream of Krasnoyarsk), “Atamanovo (village)” (88 km), and “Balchug” (98 km) – the sites contaminated due to global fallouts and MCC aerosol discharges; and “Atamanovo (island)” and “Balchug (riverside)” – the Yenisei floodplain sites contaminated by water-transported artificial radionuclides. Investigations were performed from 2002 to 2007.



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

2.2 Chemical fractionation of soil and mushroom samples

To determine speciation of radionuclides in fresh soil and mushroom samples, sequential chemical separation techniques were used. Soil separation was performed using the procedure described by Klemm et al. [4].

Chemical fractionation of mushrooms was performed using a modified procedure of sequential chemical extraction for plants described by Bolsunovsky et al. [5]. The procedure was modified by uniting the “exchangeable” and the “adsorption” fractions into one “surface-adsorption” fraction. A modified sequential extraction scheme for mushrooms is presented in Table 1.

Table 1. Sequential extraction scheme for mushroom samples.

| Fraction | Chemical reagents | Treatment |
|---------------------------|---|---|
| I + II Surface-adsorption | H ₂ SO ₄ (0,1 M) | 4 h stirring |
| III Organic | H ₂ O ₂ (35%) + HNO ₃ (1 M) up to 0.05 M | 1–1.5 h heating at 85 °C |
| IV Mineral residue | | Residue left after removal of the previous fraction |

2.3 Determination of radionuclide activities in samples

The activity concentrations of the γ -emitting nuclides in the samples were measured on a γ -spectrometer (Canberra, USA) coupled to a hyper-pure germanium detector. The γ -spectra were processed using the CANBERRA GENIE 2000 software (USA). In samples of blackcurrant and the soil collected from “Atamanovo (island)”, a floodplain site, ⁹⁰Sr was determined by measuring its daughter element – ⁹⁰Y. The radiochemical extraction of ⁹⁰Sr and ⁹⁰Y was performed using the technique described in HASL-300 [6]. The extracted ⁹⁰Y was measured on an α - β gas-flow proportional counter (IN-20 Eurisys, France). All activity concentrations of radionuclides were calculated for air dry mass of the samples.

3. RESULTS AND DISCUSSION

3.1 Radionuclides in soil

Laboratory investigations of the soil samples collected at “Zheleznogorsk”, “Atamanovo (village)”, and “Balchug” revealed only ¹³⁷Cs. Its activity concentration in the area subjected to aerosol discharges of the MCC is 2–3 times higher than the background activity (at “Krasnoyarsk”), reaching 100 Bq/kg.

Gamma-spectrometric analysis showed that the floodplain soils of “Atamanovo (island)” contained ^{60}Co – up to 160 Bq/kg, ^{137}Cs – up to 1800 Bq/kg, ^{152}Eu – up to 400 Bq/kg, ^{154}Eu – up to 60 Bq/kg, ^{155}Eu – up to 12 Bq/kg, and ^{241}Am – up to 25 Bq/kg. These soils also contained ^{90}Sr – up to 26 Bq/kg. The radionuclide inventory of the Yenisei floodplain soils has become so large due to contaminated sediments carried there during very high floods. Our studies showed that radionuclides were distributed in the soil of the “Atamanovo (island)” site very non-uniformly: the average activity concentration of ^{137}Cs in the central part of the island was 2.5 times lower than in its riverside part. ^{137}Cs activity concentration varied over the range from 400 Bq/kg to 1800 Bq/kg. There was some, although not so strongly pronounced, non-uniformity in the distribution of ^{137}Cs over other sites, too. Similar non-uniformity was observed in the distribution of other artificial radionuclides.

Of all radionuclides present in the soils, mushrooms and plants can only take up radionuclides dissolved in soil solution or those capable of being dissolved. Results of sequential chemical separation show that over 90% ^{137}Cs is present in soil in a bound state (mineral residue); the bound and the mobile fractions (all fractions except the residue) of ^{60}Co and ^{152}Eu are equal.

3.2 Radionuclides in mushrooms

The 12 studied mushroom species (*Boletus edilis* Bull.: Fr., *Cantharellus cibarius* Fr., *Hydnum repandum* L.: Fr., *Lactarius deliciosus* (L.: Fr.) S.F. Gray, *Lactarius resimus* (Fr.: Fr.) Fr., *Leccinum scabrum* (Bull.:Fr.) S.F. Gray, *Lyophyllum gambosum* (Fr.) Sing, *Russula foetens* Pers.: Fr., *Russula vesca* Fr., *Suillus granulatus* (L.: Fr.) Roussel, *Suillus luteus* (L.: Fr.) Roussel, *Armillaria mellea* (Vahl : Fr.) Kumm)) accumulated only one γ -emitting artificial radionuclide – ^{137}Cs – and its accumulation was clearly species specific; accumulation levels ranged over two orders of magnitude. In the mushrooms collected from the sites contaminated with aerosol-delivered radionuclides (“Zheleznogorsk”, “Atamanovo (village)”, and “Balchug”), ^{137}Cs activity concentration varied from several Bq/kg to several hundred Bq/kg [3]. In the mushrooms collected from “Atamanovo (island)”, ^{137}Cs activity concentration varied from 100 to 10000 Bq/kg. Bioindicator properties were exhibited by *S. granulatus* and *Suillus luteus*; these species accumulated similar amounts of ^{137}Cs . The average activity concentration of ^{137}Cs in *S. granulatus* mushrooms collected at the sites that receive only aerosol radioactivity from the MCC is 2–4 times higher than in the mushrooms from “Krasnoyarsk” (the control site). In *S. granulatus* and *S. luteus* mushrooms from the floodplain sites ^{137}Cs activity concentration reached 10200 Bq/kg, exceeding the standard limit accepted in Russia; it was comparable with activity levels recorded at some sites of the 30-km zone around the Chernobyl NPP [7].

The investigation of possible concentration of ^{137}Cs in certain parts of mushroom fruiting bodies showed that ^{137}Cs activity concentration in caps was 1.7–2.3 times higher than in stems.

The determination of the strength of ^{137}Cs binding to biomass of *S. granulatus* and *S. luteus* showed that the distribution of ^{137}Cs was similar to that of ^{40}K . We have not found any differences in the distribution of ^{137}Cs in the mushrooms collected from the sites that received ^{137}Cs from different sources. The largest percentages of ^{137}Cs and ^{40}K are detected in the surface-adsorption fraction (56% to 71% of the total radionuclide content). In the organic fraction ^{137}Cs and ^{40}K vary from 23 to 37%. Less than 6% of the ^{137}Cs activity accumulated by the mushrooms was found to be strongly bound to the mushroom biomass (mineral residue). Hence, as the fruiting body decays, most of the accumulated ^{137}Cs will become available to other organisms.

To evaluate transfer of radionuclides from the soil to mushrooms, transfer factor (TF) is used. In the *S. granulatus* and *S. luteus* samples collected at the sites contaminated by aerosol-delivered ^{137}Cs , the absolute amount of ^{137}Cs grows linearly, as it increases in the soil, and the average transfer factors (TFs) for ^{137}Cs and ^{40}K are the same – 3.0 ± 0.3 (Table 2). However, in the mushroom samples collected from the soils contaminated by water-delivered ^{137}Cs , TFs for ^{137}Cs are more than 5 times higher.

Table 2. ^{40}K and ^{137}Cs transfer factors for mushrooms (Bq/kg dry weight mushrooms to Bq/kg dry weight soil).

| Pathways of radionuclide delivery from the MCC | Sampling site | ^{40}K TTF | ^{137}Cs TF |
|--|-----------------------|---------------------|----------------------|
| Aerosol | “Atamanovo (village)” | 2.7 | 3.0 |
| | “Balchug” | 3.3 | 2.9 |
| | “Zheleznogorsk” | 3.2 | 3.4 |
| | “Krasnoyarsk” | 2.9 | 2.7 |
| Water | “Balchug (riverside)” | 2.1 | 4.7 |
| | “Atamanovo (island)” | 2.0 | 10–16 |

The study of radionuclide accumulation by mushrooms has confirmed that *S. granulatus* and *S. luteus* are concentrators of radiocesium. The calculated variations in ^{137}Cs TFs are similar to the data reported by other researchers [8].

3.3 Radionuclides in berry shrubs

Radionuclide accumulation in the shrub storey was investigated in components of the aboveground phytomass of *Ribes hispidulum* (Jancz.) Pojark, *Ribes nigrum* L., *Rosa majalis* Herrm., *Rubus idaeus* L., and *Viburnum opulus* L. All plant samples were found to contain ^{40}K and ^{137}Cs . Samples of aboveground phytomass from the floodplain soils also contained ^{60}Co and ^{90}Sr ; no other artificial radionuclides were found to accumulate in phytomass. The registered levels of ^{137}Cs in the plants were 2–3 orders of magnitude lower than in mushrooms. It was also found that differences in ^{137}Cs levels in berry shrubs of different species were not more than 2-fold, while in mushrooms from the same sites they reached 2 orders of magnitude.

Of the study species, the highest levels of accumulated radionuclides were registered in raspberry and blackcurrant and, thus, these species were chosen for a more detailed examination. The aboveground phytomass of the studied berry shrubs only accumulated ^{60}Co , ^{90}Sr , and ^{137}Cs (Table 3). ^{137}Cs and ^{90}Sr were accumulated in all shrub parts, but just leaves and branches accumulated ^{60}Co .

For all species there is a 2–3-fold difference in radionuclide accumulation between their leaves and branches, which can be accounted for by conductive function of branches during the period of active plant growth. In the studied berry shrubs collected from the sites contaminated by aerosol-delivered radionuclides only ^{137}Cs was detected and its activity was near the limit of detection.

To evaluate transfer of radionuclides from the soil to plants, transfer factor is used. The ^{40}K TFs for branches of raspberry and blackcurrant shrubs are 0.4–0.5 and for their leaves and berries – 1–1.4. The ^{137}Cs TFs calculated for the studied berry shrubs (Fig. 2) are within the range from 0.01 to 0.03, i.e. 1–2 orders of magnitude lower than the ^{40}K TFs. The ^{137}Cs TFs calculated for the sites that receive radionuclides from different sources fall within the same range of values. The highest transfer factors are recorded for ^{90}Sr . In blackcurrant shrubs, the ^{90}Sr TFs for leaves reach 1.9, for branches – 0.9, and for berries – 0.25, i.e. they are two orders of magnitude higher than the ^{137}Cs TFs. The ^{60}Co TFs are similar to those of ^{137}Cs (Fig. 2c), exceeding the values reported for the area around the Chernobyl NPP [9].

Based on the calculated TFs, the distribution of ^{137}Cs and ^{60}Co among the parts of these shrubs is as follows: branches < leaves ~ berries, and the distribution of ^{90}Sr – berries < branches < leaves.

The results we obtained for accumulation of artificial radionuclides by berry shrubs are in good agreement with the data reported by other authors. The ^{137}Cs TF to the green phytomass of raspberry shrubs in the 30-km zone around the Chernobyl NPP is 0.02–0.4 [9]. The field investigations of ^{137}Cs accumulation by raspberry fruits yielded similar results, too: 0.015–0.028. For blackcurrant berries collected in the vicinity of Sellafield the average ^{137}Cs TF was reported to be 0.015 [10], which is also similar to our values. However, the ^{60}Co TF to the green phytomass of raspberry shrubs in the 30-km zone around the Chernobyl NPP is 0.006–0.007 [9], which is lower than our values.

Table 3. Accumulation of radionuclides in shrubs growing at “Atamanovo (island)”, Bq/kg.

| | Radionuclide | <i>Ribes nigrum</i> | <i>Rubus idaeus</i> |
|----------|-------------------|---------------------|---------------------|
| Branches | ⁴⁰ K | 204 ± 18 | 160 ± 15 |
| | ⁹⁰ Sr | 11 ± 2 | n.d. |
| | ⁶⁰ Co | 0.8 ± 0.2 | <DL |
| | ¹³⁷ Cs | 13 ± 1 | 11 ± 2 |
| Leaves | ⁴⁰ K | 620 ± 50 | 480 ± 40 |
| | ⁹⁰ Sr | 45 ± 6 | n.d. |
| | ⁶⁰ Co | 2.8 ± 0.5 | 1.6 ± 0.4 |
| | ¹³⁷ Cs | 29 ± 2 | 17 ± 1 |
| Berries | ⁴⁰ K | 490 ± 40 | 420 ± 30 |
| | ⁹⁰ Sr | 3.8 ± 0.6 | n.d. |
| | ⁶⁰ Co | <DL | <DL |
| | ¹³⁷ Cs | 23 ± 3 | 17 ± 2 |

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

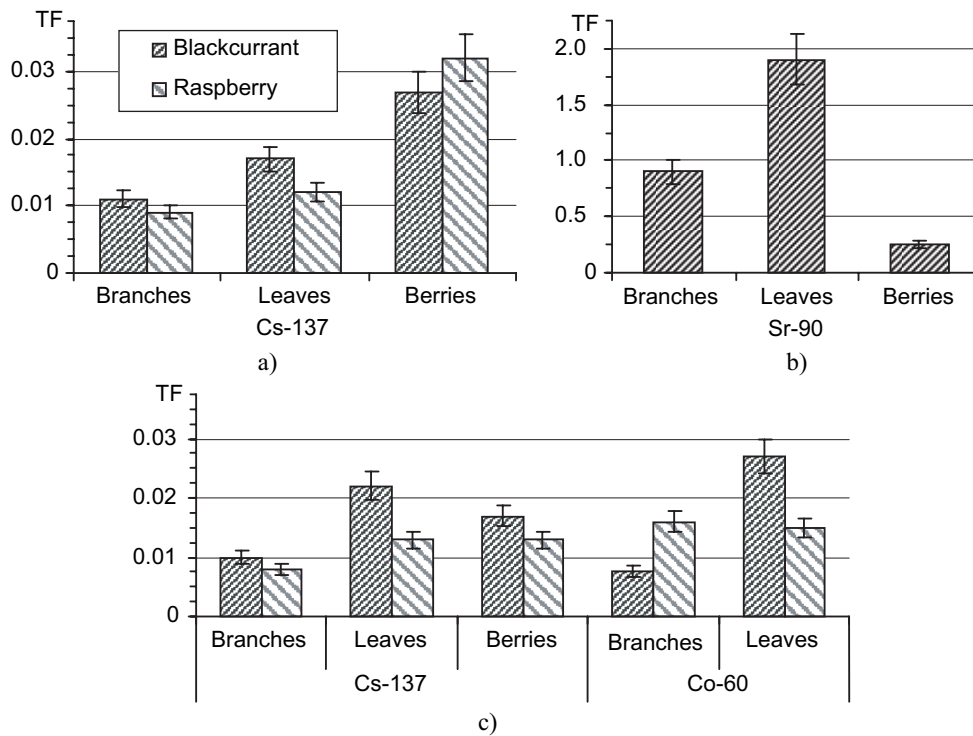


Figure 2. Transfer factors (Bq/kg dry weight of the plant part to Bq/kg dry weight soil) of artificial radionuclides to different parts of the shrubs for the sites contaminated by aerosol- (a) and water-delivered (b, c) radionuclides.

3.4 The contribution of the intake of mushrooms and berries to the radiation exposure of residents

To estimate the contribution of the consumed mushrooms and berries to the effective dose to residents of the studied area, we calculated the annual effective dose of internal exposure per person. The calculation

was based on the annual intake of mushrooms and berries by residents of Russia – 1 to 14 kg and 10 kg, respectively.

The highest possible dose due to intake of mushrooms collected in the area affected by MCC operation is $150 \mu\text{Sv}/\text{year}$ for the sites contaminated by water-delivered radionuclides; this is comparable with $200 \mu\text{Sv}/\text{year}$ received by residents consuming mushrooms and berries in the Bryansk Region, which was affected by the Chernobyl disaster [11]. The dose due to intake of mushrooms for the sites contaminated by MCC aerosol-delivered radionuclides is $5 \mu\text{Sv}/\text{year}$. The annual effective dose due to intake of berries does not exceed $1.4 \mu\text{Sv}/\text{year}$.

The contribution of the annual effective dose of internal exposure due to intake of mushrooms and berries collected in the area affected by the operation of the MCC is 15% of the standard limit accepted in Russia ($1000 \mu\text{Sv}/\text{year}$) for mushrooms and 0.1% for berries.

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