RESEARCH ARTICLE

Radionuclide uptake and dose assessment of 14 herbaceous species from the east-Ural radioactive trace area using the ERICA Tool

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Abstract

The evaluation of radiation exposure in 14 species of herbaceous plants from the East-Ural Radioactive Trace (EURT) zone was performed, using the ERICATool, v. 1.2. Recent (up to 2015) levels of radionuclide activity concentration were measured in soil and vegetative plant mass. ^{239,240}Pu content was used for the first time to estimate external dose rates for herbaceous plant species along the pollution gradient. In addition, a new approach to assessing the geometry of objects was adopted, including not only aboveground but also underground plant organs. This improved approach to the evaluation of radiation exposure confirms previous findings that herbaceous plant populations currently exist under low-level chronic exposure in the EURT area. This reassessment based on new data suggests a 48–977-fold increase in the total dose rate per plant organism at the most polluted site compared to background areas. The highest capacity for the transfer of ⁹⁰Sr and ¹³⁷Cs was observed in *Taraxacum officinale* and Plantago major. In these species, the total dose rate per plant exceeded 150 µGy h⁻¹ due to ⁹⁰Sr + ¹³⁷Cs + ^{239,240}Pu radionuclide anthropogenic pollution in the EURT zone. All estimated total dose rates per plant were below the dose rate screening value of 400 μGy h^{-1} .

Keywords Kyshtym accident \cdot ⁹⁰Sr \cdot ¹³⁷Cs \cdot ^{239,240}Pu \cdot Dose assessment \cdot CR

Introduction

The Kyshtym accident occurred at the Mayak Production Association (Mayak PA) in 1957 in the Southern Urals, USSR. It resulted in the release of 7.4 PBq (10% of the radioactive waste total volume) of man-made radionuclides to the environment in a northeast direction, dispersing

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across a territory of $23,000 \text{ km}^2$ (Romanov et al. [1990](#page-12-0)). At present, the East-Ural Radioactive Trace (EURT) zone is a convenient test site to evaluate the influence of radioactivity on natural ecosystems since living organisms here have been exposed to technogenic ionising radiation for over half a century (Antonova et al. [2015;](#page-11-0) Pozolotina et al. [2010\)](#page-12-0). The Population Radiobiology laboratory (Institute of Plant and Animal Ecology, Russia) investigates the long-term effects of radiation on plants at the population, organism, and cell levels. Within the last decade, a wide range of biological effects were observed in wild plant populations in the EURT area: rare and unique enzyme alleles or morphs (Pozolotina and Antonova [2009;](#page-12-0) Pozolotina et al. [2007](#page-12-0)), expanding the range of variability of germination rates of seed progeny (Karimullina et al. [2015](#page-12-0); Pozolotina and Antonova [2017](#page-12-0)), and a large number of different morphoses revealed at the early stages of ontogenesis in populations of Stellaria graminea (Pozolotina et al. [2010\)](#page-12-0), Melandrium album (Antonova et al. [2013](#page-11-0)), and Bromus inermis (Antonova et al. [2014\)](#page-11-0). The assessment of radiation doses is necessary to explain these radioecological and radiobiological effects due to elevated levels of technogenic radionuclides in the wild.

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The potential of the ERICA Tool in assessing radiation exposure on non-human biota has been demonstrated in numerous studies (Karimullina et al. [2013](#page-12-0); Kubota et al. [2015](#page-12-0); Mazeika et al. [2016](#page-12-0); Sotiropoulou et al. [2016;](#page-12-0) Wood et al. [2008](#page-12-0)). Because the accumulation of radionuclides in the phytomass depends on the chemical properties of the emitters, the chemical characteristics of the soil, and the biological features of individual species and their growing conditions, a unique combination of factors is formed in each area of operating atomic energy industrial enterprises. A considerable area of the Ural region, experiencing radioactive contamination stress, as a result of radiation accidents and routine operation of nuclear facilities, is of particular interest (Karavaeva et al. [2010](#page-11-0)). It was previously shown that the dose received by herbs exceeded the background level by several orders of magnitude in the most contaminated sites of the EURT area (Pozolotina et al. [2007](#page-12-0); Pozolotina et al. [2012b](#page-12-0)). Later, we conducted a new assessment of doses comparing novel approaches such as the ERICA Tool 1.0 (Beresford et al. [2007](#page-11-0); Brown et al. [2008](#page-11-0); Larsson [2008](#page-12-0)) and RandD128/SP1a (Copplestone et al. [2001](#page-11-0); Copplestone et al. [2003\)](#page-11-0) for four herbaceous plant species growing in the EURT zone (Karimullina et al. [2013\)](#page-12-0). In herbs, the interspecific accumulation capability of radionuclides may vary by four orders of magnitude (Kabata-Pendias [2010](#page-11-0); Pozolotina et al. [2012b](#page-12-0); Solecki et al. [2003\)](#page-12-0). Therefore, the spectrum of herb species, common in temperate environments, was expanded by ten more species in our new study. Recent (up to 2015) field data of ${}^{90}Sr + {}^{137}Cs$ radionuclide concentration and additional radionuclide $239,240$ Pu content in soil and plants has become available due to continuous ecological monitoring of the EURT zone. Species-specific concentration ratios of the main contaminants of the EURT area, therefore, need further evaluations which are provided in the present study.

The ERICA Tool is a continually evolving program developed by an international group of scientists (Avila et al. [2014](#page-11-0); Brown et al. [2013](#page-11-0); Copplestone et al. [2013;](#page-11-0) Hosseini et al. [2013](#page-11-0)). Researchers have sought to improve and modify the ERICATool, including efforts at consistency with developments originating from the International Atomic Energy Agency (IAEA) and International Commission on Radiological Protection (ICRP) (Brown et al. [2016\)](#page-11-0).

The aim of this study was the evaluation of a radionuclide uptake and dose assessment of 14 species of herbaceous plants from the EURT zone based on (1) new data measured directly in the EURT zone, (2) using geometry of root and shoot for each plant species and (3) the analysis of the contribution of 239,240Pu in addition to the activity concentrations of 90 Sr + 137 Cs in soil and plants.

The ERICA Tool version 1.2, released in November 2014 (Brown et al. [2016\)](#page-11-0), was used to perform dose assessment for natural herb populations at six sites along the EURT pollution gradient. The most common herb species within the area were involved in the analysis.

Materials and methods

Characterisation of sites

The EURT zone is located within the Trans-Ural forest-steppe, characterised by the alternation of steppe meadows, small groves of birch and birch aspen, and pine forests (Gorchakovsky [1968](#page-11-0)). The detailed biogeochemical and geobotanical description of the area was given previously (Karimullina et al. [2013](#page-12-0); Pozolotina et al. [2012b](#page-12-0)). We investigated areas with diverse levels of radionuclide contamination in the EURT area and beyond. The map (Fig. [1](#page-2-0)) and the characterisation of sites is summarised here but is discussed elsewhere (Karimullina et al. [2013;](#page-12-0) Pozolotina et al. [2012b](#page-12-0)).

Two background plots are located outside the EURT area. Background 1 site [56°41′N61°02′E] is located in the birchpine forest about 112 km from the centre of the accident. Live vegetative cover is multi-layer. The projective cover of the grass, i.e. the proportion of the ground covered by the vertical projection of vegetation (Lindenmayer and Burgman [2005](#page-12-0)), is 95%. The soil type is brown forest. Close proximity to a village area has led to a high abundance of ruderals, i.e. plant species with high survival rates on lands disturbed by natural and anthropogenic factors (Grime [1977\)](#page-11-0). Background 2 site [56°47′N 61°18′E] is located in a birch-pine forest mixed with some aspen 125 km away from the centre of the accident. The projective cover of the grass is 90–95%.

Buffer 1 site [55°52′N 60°54′E] is located 15–18 km from the centre of the accident at the western border of the EURT zone. The site has a secondary forb-grass meadow, i.e. regrown vegetation containing herbaceous flowering plants in addition to grasses, which includes 40–45 species. The projective cover of the grass is 100%. Buffer site 2 55°49′N 60°55′E] is located 15–18 km from the centre of the accident on the west bank of Lake Uruskul. The site has a secondary forb-grass meadow with birch trees. The total projective cover is 75–80%. The projective cover of the grass is 50–60%.

Impact 1 site $[55^{\circ}46'$ N 60°53′E] is located 9–12 km from the centre of the accident on the south-west bank of Lake Berdenish. Leached chernozem and subtypes of grey forest soils predominate at this site. The involvement of ruderal species in meadows and forest communities of the impact zone is reduced, while plants typical to the communities play significant roles. The total projective cover is 80–90%. Impact 2 site [55°45′N 60°50′E] is the closest to the centre of the accident, located at a distance of 6–8 km. Soils are brown forest. The most transformed plant community includes ruderals, lungwort and thistle. The total projective cover is 100%.

Field sampling and preparation

Field sampling of soil and vegetation was undertaken during the period from 2006 to 2015. Soil cores were collected

Fig. 1 Location of the sample sites within the main part of the East-Ural Radioactive Trace and background areas. From the closest location of Mayak PA: imp-2 Impact 2 site, imp-1 Impact 1 site, buf-2 Buffer

2 site, buf-1 Buffer 1 site, bg-2 background-2 site, bg-1 background-1 site (Karimullina et al. [2013\)](#page-12-0)

randomly in 4–25 replicates at each site. To determine the level of radionuclide contamination of soil, samples were taken from soil profiles, placing them at the corners of a triangle with sides 10 m in length. The area of the soil samples was approximately and not more than 0.1 m^2 . Our independent preliminary studies have shown that the sampling within this scheme representatively characterises soil contamination in sites of this size. Samples of plants were taken in the immediate vicinity of the soil profile. Vegetation samples (2–10 replicates) were obtained by cutting with garden shears above the soil surface. The level of surface contamination was controlled visually, so that there was no physical contamination on the surface of the plants (soil particles easily visible to the naked eye). In a natural environment, the surface of the soil is covered with a layer of dead vegetation. Therefore, large particles of soil do not normally reach the surface of the leaves. The pubescence or stickiness of the leaves does not allow the microparticles to be washed away in some plant species. In most cases, with enhanced flushing, not only radionuclides that have entered the plant surface from the atmosphere or as a result of wind transfer with dust particles but also a part of the radionuclides taken up by other plant tissues will be lost. It is of fundamental importance not to rinse, but to take into account finely dispersed radionuclides because, being on the surface of leaves for a long time (3–4 months), they also contribute to the total dose.

Radiochemical analysis was performed to determine the ⁹⁰Sr content in soil and vegetation samples. The method was based on the leaching of radiostrontium with 6 N HCl-solution after preliminary preparation of samples. Separated ⁹⁰Sr was kept in balance with its daughter product of decay, $\rm{^{90}Y}$, after precipitation of 90 Sr in the oxalate form. Measurements of 90 Y were

established using an α-β-radiometer with a siliceous detector (Russia). The detection limit was 0.2 Bq.

The radionuclide analysis of 137 Cs content was performed using γ -ray spectroscopy, for which the Canberra-1510 multichannel analyser and GENIE-PC software (Canberra-Packard, USA) germanium detectors were used. The detection limit was 0.1 Bq.

Radiochemical determination of 239,240Pu included several steps, such as leaching isotopes by mixed concentrated acids $(HNO₃ + HCl)$ from preliminary prepared samples, precipitating plutonium on ion-exchange resin, purification, stripping from the ion-exchange column by H_2O and 0.5 N HNO₃ solution in succession and electro-deposition on disks made of stainless steel (Chen et al. [1993\)](#page-11-0). The system used for the measurements was an α-spectrometer Alpha Analyst (Canberra-Packard, USA) with semiconductor detectors (PIPS) and GENIE-2000 software. The lower limit of detection was 0.001 Bq. The procedural error of the methods did not exceed 20%. A correction for radioactive decay was applied for samples collected in different years.

Test organisms

Fourteen common herbaceous plant species capacity to accumulate radionuclides was analysed. Dominant or codominant herbs, analysed in this study, include little starwort (Stellaria graminea L.), white campion (Melandrium album (Mill.) Garcke (= Silene latifolia)), smooth brome (Bromopsis $inermis$ Leyss. $(= Bromus$ inermis)), quinquelobate motherwort (Leonurus quinquelobatus Gilib.), creeping thistle (Cirsium setosum $(=$ Cirsium arvense s.l.), yellow sweet clover (Melilotus officinalis L.), common nettle (Urtica dioica L.), woolly burdock (Arctium tomentosum Mill.), common dandelion (Taraxacum officinale s.l.), broadleaf plantain (Plantago major L.), Russian dock (Rumex confertus Willd.), zigzag clover (*Trifolium medium L.*), meadow pea (Lathyrus pratensis L.) and common wormwood (Artemisia vulgaris L.). These plant species represent 14 genera and 8 families in our study.

Data analysis

To estimate the doses of external and internal radiation exposure using the ERICA Tool program, the average values of parameters (weight, height, width, length) were used for 14 species of plants commonly found in the surveyed territory (see supplementary material, Tables 1 and 2) The following assumptions characterise the model used in this study:

- 1. The dimensional parameters of the vegetation mass (see supplementary material, Table 1) of plants of different species were averaged for each species and are in good agreement with botanical literature for this region (Gorchakovsky et al. [1964\)](#page-11-0).
- 2. The average rates of activity concentrations of man-made radionuclides were used in each species of herbaceous plant for each investigated site, because the content of radionuclides in plants varies widely, depending on the type and age of the plants, the weather conditions during the vegetation period, the way radionuclides enter the soil, the properties of the soil and other factors (Molchanova et al. [2014a](#page-12-0); Shcheglov and Tsvetnova [2001;](#page-12-0) Willey and Tang [2006](#page-12-0)).
- 3. One of three ellipsoid dimensions, the height of the roots, was taken as 0.1 m for all herbaceous species (see supplementary material, Table 2), as the bulk of the roots is located in this the most contaminated layer of soil. The other two ellipsoid dimensions, width and length of the roots, forming a square area, were taken as equal to the projective vegetation mass; this parameter was conditional and unified for all herb species. This assumption was made according to previous findings that in the forest-steppe zone, the ratio of underground and aboveground phytomass in the out-of-place meadows, steppes and agricultural lands is close to 1 (Ravkin et al. [2011](#page-12-0)). We also observed that the mass of living and dead roots and the aboveground vegetation mass (living and dead) per one square meter were approximately the same (our unpublished data).

The ERICATool uses activity concentrations of vegetation on a fresh weight basis. The radionuclide content measurement procedure uses dried samples of vegetation. Therefore, we computed the fresh weight using the dry/wet ratio for Stellaria graminea = 1/2.5; Bromus inermis, Melilotus officinalis, Urtica dioica, and Leonurus quinquelobatus = $1/$ 2; Melandrium album, Arctium tomentosum, Artemisia vulgaris, Cirsium setosum, Lathyrus pratensis, and Rumex confertus = 1/3; Plantago major, Taraxacum officinale, and Trifolium medium = $1/4$. The difference was determined by weighing freshly harvested samples of vegetation before and after complete drying. This allowed us to calculate the concentration ratio for the wild organism $(CR_{wo-soil})$ for each of the 14 species of herbaceous plants. The standard equation (Beresford et al. [2007;](#page-11-0) Karimullina et al. [2013](#page-12-0)) was used for this purpose. Data are presented as the arithmetic mean (AM) \pm standard deviation (SD). Dose assessment was performed using the ERICA Tool 1.2 software (Brown et al. [2016\)](#page-11-0).

Results and discussion

Contamination gradient of soil

The spatial distribution of radionuclides in the EURT area was estimated in our previous works (Molchanova et al. [2009,](#page-12-0) [2011](#page-12-0), [2014b\)](#page-12-0). Within the central axis, the contamination densities of radionuclides change with increasing distance from the centre of the accident and satisfactorily approximate a power function (for example, stock of 90 Sr changes from 69 MBq m−² to 0.4 MBq m−²). On the eastern periphery of the trace zone, the value of 90 Sr and 137 Cs stocks are almost identical and with increasing distance reduce from 100 to 10 kBq m−² . On the western periphery of the trace, the content of 137 Cs in soil, regardless of the distance, is kept at a level (5– 10) kBq m−² . The stocks of 239,240Pu are much lower and change from 5 to 0.2 kBq m^{-2} with distance from the pollution source. On the whole territory of the EURT, the 20 cm soil layer keeps 80–92% of the total 90 Sr, 137 Cs and 239,240 Pu stocks. Thus, these radionuclides are characterised by low mobility in the soil profiles.

In our previous work (Karimullina et al. [2013\)](#page-12-0), two main EURT contaminant radionuclides in soil, 90 Sr and 137 Cs, were used to calculate the external dose rates. In the present study, we additionally included ^{239,240}Pu content in soil and vegetation to estimate its impact on the total exposure of plant populations to technogenic radionuclides. Our present data demonstrate an increase in 90Sr levels in soil at the Impact 2 site to 15,500 or even 92,700 times the pollution levels observed at the two background sites. Notably, the Impact 2 site is located closest to the site of the Kyshtym accident (Table [1\)](#page-4-0). 239,240Puand 137Cs-specific activity in Impact 2 soil samples was 2 and 3 orders of magnitude higher than those from background sites respectively. Soil samples from buffer sites had intermediate radionuclide activity concentrations rates.

Activity concentrations in 14 herbaceous species in the EURT area

Radionuclide activity concentrations in biota are required for Tier 2 and 3 assessments using the ERICA Tool (Beresford Table 1 Activity concentrations in soil samples from the EURT

zone and background area

et al. [2007](#page-11-0)). As for radionuclide concentrations in soil and plants, sufficient data are available in our case.

The activity concentrations of 90Sr , 137Cs and $239,240\text{Pu}$ were used to calculate the transfer parameters in 14 species of herbaceous plants from the EURT zone and background area. These data were determined empirically and are present-ed in Table [2](#page-5-0). The lowest levels of 90 Sr and 137 Cs specific activity were observed in A. Tomentosum and L. quinquelobatus vegetative mass, especially at background sites. The minimum activity concentration of $239,240$ Pu was observed in L. pratensis. The highest activity concentrations were in *T. officinale* (90 Sr and 137 Cs) and two species from the Caryophyllaceae family (M. album and S. graminea) $(239,240)$ Pu) collected in impact plots. Previously, we observed a low capacity for biological transition to herbaceous plants for 137Cs at the EURT zone (Karimullina et al. [2013](#page-12-0); Pozolotina et al. [2012b\)](#page-12-0). The major contribution to internal dose levels is made by ⁹⁰Sr radionuclide in the technogenic EURT area. In the background sites, the ratio of activity concentrations of $\frac{90}{ST}$ $\frac{ST}{137}Cs$ and $\frac{90}{ST}$ $\frac{239,240}{SU}$ radionuclides was approximately 19:1 and 300:1 in the vegetative mass, respectively. In the EURT impact plots, that ratio reached 700:1 and 5000:1, respectively.

According to different studies, the ratio of concentrations of 90Sr and 137Cs in the roots of plants and plant organs above the ground varies greatly. This ratio may be either larger or smaller than one. Moreover, there are data indicating that when the conditions of mineral nutrition of plants change, the ratio of radionuclide concentrations in the aboveground vegetation mass and roots may change significantly even within the same species (Willey and Tang [2006](#page-12-0)). There are no such data for plutonium. However, there is evidence of an accumulation of its iron macro-analogue (Afanasyeva and Kashin [2015](#page-11-0); La Toya et al. [2010\)](#page-12-0), which allows the use of a root/shoot ratio equal to 1. Our spatial empirical data on the accumulation of radionuclides in the roots of herbaceous plants from the EURT zone allow us to proceed from the assumption that the concentrations in the aboveground mass and in the roots of the plants may be the same.

Input CRwo-soil values data

A standard equation (Beresford et al. [2007](#page-11-0); Karimullina et al. [2013\)](#page-12-0) was used to define the concentration ratios $(CR_{\text{wo-soil}})$ of 14 herbaceous species for the 90 Sr, 137 Cs and 239,240 Pu radionuclides (Table [3](#page-6-0)). The highest uptake of 90 Sr was observed in *T. officinale*. The next highest uptake was observed in P. major followed by M. album. U. dioica and T. medium, and C. arvense had intermediate $CR_{wo-soil}$ values. The lowest uptake was revealed in B. inermis, S. graminea and A. tomentosum species. The ^{137}Cs CR_{wo-soil} was low in most species. The weakest transfer of $137Cs$ was identified in L. quinquelobatus, B. inermis, A. tomentosum, M. officinalis and S. graminea species. M. album, T. medium, L. pratensis and U. dioica are grouped with ascending values of $137Cs$ $CR_{\text{wo-soil}}$. The highest transfer capacity for $137Cs$ was observed in T. officinale. Minimal CR values of $239,240$ Pu were determined in L. pratensis, which were two orders of

^a Single cases with no measured for a particular species; therefore, a mean value from the empirically derived values of activity concentrations for a species from the same taxonomical family (similar reference organism) growing in the same area was used

Table 3 Concentration ratios of ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu in 14 herbaceous species summarised by taxonomic family (concentrations were measured from shoots as described in the "[Material and methods](#page-1-0)" section)

Family	Species	Number	AM	SD	Min	Max
90 Sr						
Asteraceae	Arctium tomentosum	6	$3.25E - 02$	$2.54E - 02$	$1.48E - 02$	$8.33E - 02$
	Artemisia vulgaris	3	$2.31E - 01$	$7.34E - 02$	$2.00E - 01$	$3.36E - 01$
	Cirsium arvense	5	$4.59E - 01$	$2.63E - 01$	8.45E-02	$6.54E - 01$
	Taraxacum officinale	3	8.25E-01	$8.84E - 01$	$2.86E - 01$	$1.85E + 00$
Caryophyllaceae	Stellaria graminea	2	4.81E-02	4.15E-02	$1.87E - 02$	$7.74E - 02$
	Melandrium album	4	$4.63E - 01$	$3.15E - 01$	$1.22E - 01$	8.18E-01
Fabaceae	Lathyrus pratensis	5	$1.42E - 01$	$1.26E - 01$	$1.35E - 02$	3.49E-01
	Melilotus officinalis	$\overline{2}$	$2.24E - 01$	$1.35E - 02$	$2.14E - 01$	$2.33E - 01$
	Trifolium medium	7	$4.59E - 01$	$2.38E - 01$	$1.10E - 01$	$6.67E - 01$
Lamiaceae	Leonurus quinquelobatus	5	$1.27E - 01$	$4.85E - 02$	5.29E-02	$1.82E - 01$
Poaceae	Bromus inermis	$\overline{7}$	8.22E-02	$1.14E - 01$	$9.34E - 03$	$1.27E - 01$
Polygonaceae	Rumex confertus	8	$1.83E - 01$	3.79E-01	$4.12E - 03$	3.59E-01
Plantaginaceae	Plantago major	3	$6.62E - 01$	$7.67E - 01$	$2.10E - 01$	5.00E-01
Urticaceae	Urtica dioica	8	4.59E-01	$2.13E - 01$	8.33E-02	5.23E-01
137 Cs						
Asteraceae	Arctium tomentosum	6	$1.29E - 03$	$9.34E - 04$	7.87E-04	$3.43E - 03$
	Artemisia vulgaris	3	$1.73E - 03$	$1.17E - 03$	$1.03E - 03$	$3.77E - 03$
	Cirsium arvense	5	$4.03E - 02$	$3.64E - 02$	$1.14E - 03$	$5.95E - 02$
	Taraxacum officinale	3	$1.84E - 01$	$2.58E - 01$	$2.00E - 02$	$4.81E - 01$
Caryophyllaceae	Stellaria graminea	2	$2.18E - 03$	2.28E-03	5.68E-04	$3.79E - 03$
	Melandrium album	4	$8.22E - 03$	$4.02E - 03$	5.53E-03	$1.42E - 02$
Fabaceae	Lathyrus pratensis	5	$1.22E - 02$	$1.21E - 02$	$2.15E - 03$	$3.65E - 02$
	Melilotus officinalis	\overline{c}	$2.95E - 03$	3.08E-04	$2.73E - 03$	3.16E-03
	Trifolium medium	7	$1.08E - 02$	5.46E-03	$5.62E - 03$	$3.72E - 02$
Lamiaceae	Leonurus quinquelobatus	5	$1.14E - 03$	$4.85E - 04$	$8.17E - 04$	$2.41E - 03$
Poaceae	Bromus inermis	$\overline{7}$	$1.22E - 03$	$1.21E - 03$	$4.29E - 04$	$4.53E - 03$
Polygonaceae	Rumex confertus	8	$4.68E - 02$	$1.18E - 01$	$1.40E - 05$	4.28E-02
Plantaginaceae	Plantago major	4	5.42E-02	5.38E-02	8.93E-03	$3.65E - 02$
Urticaceae	Urtica dioica	8	$1.65E - 02$	3.75E-02	$6.45E - 04$	8.98E-02
$^{239,240}Pu$						
Asteraceae	Arctium tomentosum	8	$1.07E - 02$	$1.68E - 02$	$1.84E - 04$	$4.32E - 02$
	Artemisia vulgaris	1	$7.21E - 03a$			
	Cirsium arvense	3	$3.67E - 03$	$1.01E - 03$	2.95E-03	4.38E-03
	Taraxacum officinale	1	$7.21E - 03a$			
Caryophyllaceae	Stellaria graminea	3	$3.32E - 02$	$6.09E - 03$	2.79E-02	3.99E-02
	Melandrium album	1	$3.32E - 02^a$			
Fabaceae	Lathyrus pratensis	4	$2.38E - 04$	$2.08E - 04$	$1.02E - 04$	5.45E-04
	Melilotus officinalis	$\overline{2}$	2.99E-02	2.98E-02	$2.73E - 03$	$3.16E - 03$
	Trifolium medium	4	3.21E-03	2.57E-03	$7.66E - 04$	$6.13E - 03$
Lamiaceae	Leonurus quinquelobatus	6	5.55E-03	$9.64E - 03$	3.19E-04	2.49E-02
Poaceae	Bromus inermis	7	$2.26E - 03$	$1.93E - 03$	5.74E-04	5.51E-03
Polygonaceae	Rumex confertus	8	$2.51E - 03$	3.24E-03	$1.84E - 04$	$9.97E - 03$
Plantaginaceae	Plantago major	2	$1.38E - 03$	$1.23E - 03$	5.08E-04	$2.25E - 03$
Urticaceae	Urtica dioica	4	$8.00E - 03$	7.74E-03	2.98E-04	2.29E-02

^a Single cases with no measured data for the particular species; therefore, a mean value from the empirically derived CR values of species from the same taxonomical family (similar reference organism) growing at the same area was used

magnitude lower than the maximal rates observed in S. graminea and M. officinalis. Overall, radionuclide 90 Sr showed higher soil-to-organism transfers than $137Cs$ and 239,240Pu in 14 herb species in the EURT area.

Patterns of transfer of ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu radionuclides from soil to plant were assessed by grouping herb taxa by their taxonomic families. The groups of families such as Asteraceae, Caryophyllaceae and Fabaceae presented by several species had high variability in interspecific CR rates and had no significant pattern that would indicate potent or weak uptake of radionuclides in the whole family. The absolute maximum CR rate of 137 Cs and the minimum CR rate of ⁹⁰Sr belong to members of Asteraceae. The maximum and minimum CR level of $239,240$ Pu was observed in the Fabaceae family (see Table [3](#page-6-0)).

Dose assessment using empirically derived data in soil and vegetation of 14 herbaceous species

The ERICA Tool has an input model of herbs as an ellipsoid with an on-soil occupancy factor. Therefore, previously, we used this model to analyse total dose rates. However, this approach misses the fact that the root system, representing in most cases half of the plant organism, is exposed to external doses at increased rates compared to that above the soil. Since the application of realistic occupancy factors will lead to less conservative dose estimates, we entered 28 new geometries as 28 new vegetation organisms: 14 geometries for shoots with occupancy factor on-soil equal to 1.0 and roots with occupancy factor in-soil equal to 1.0. This was performed for 14 herb species characterised by various root systems, mass, and ellipsoid dimensions (see supplementary material, Tables 1 and 2). In the wildlife group of plants, the specimen was chosen as "vegetation," and the "herb" subtype was picked. Internal and external dose conversion coefficient (DCC) values were calculated. Since the only way to calculate DCC for root and shoot separately in the ERICA Tool is to enter them into the software as two new organisms (Biermans et al. [2014\)](#page-11-0), these models were used to analyse doses at each site using the Tier 2 approach. Soil concentrations used for the analysis and CR values are presented in Tables [1](#page-4-0) and [3,](#page-6-0) respectively. The weighting factor for alpha radiation was set as 10 by default; the low-level β-radiation was set constant at 3, and for highenergy γ - and β -radiation, it was set at 1.

The external dose rates for terrestrial herb species were mainly due to the $137Cs$ radionuclide (see supplementary material, Table 3). The external dose rate increased by 3–4 orders of magnitude compared to the background rate along the pollution gradient due to anthropogenic $^{90}Sr + ^{137}Cs + ^{239,240}Pu$ radionuclide pollution. The main technogenic radionuclide contributing to the estimated internal dose rate was ⁹⁰Sr at the Impact sites, while in background plots, all three radionuclides contributed mainly equally (see supplementary material, Table 4). Due to external and internal contributing radiostrontium and radiocesium, the values of the total dose rate per root or shoot model derived from these two radionuclides were higher by 1–3 orders of magnitude compared with the values of $239,240$ Pu at the polluted sites (see supplementary material, Table 5). The values of the total dose rate calculated using the ERICA Tool Tier 2 approach (Table [4\)](#page-8-0) varied slightly between root and shoot ellipsoids within each plant due to slightly different dimensions used for input (see supplementary material, Tables 1 and 2). At the most polluted Impact 2 site, total dose rates of T. officinale and P. major were the highest (see Table [4\)](#page-8-0).

Discussion

The gradual moving from the principle that the observance of radiation dose limits established for humans is a sufficient guarantee for the protection of biota is already declared in publication 103 of the International Commission on Radiological Protection (ICRP [2007](#page-11-0)). In the subsequent publication 108 (ICRP [2008](#page-11-0)), the ICRP expanded this concept and introduced the concept of "reference" organisms, which are hypothetical objects with biological characteristics generalised at the family level, derivatives of the "reference levels," as expressed by the dose rate of radiation and which are proposed for each of them based on the data on the radiation effects observed at different doses. For the purposes of protecting the environment under irradiation conditions, the ICRP proposes to consider the species directly exposed to radiation as representative organisms. The doses of irradiation of representative organisms are compared with the derived reference levels established for close reference organisms. There are limited possibilities of ensuring the radiation safety of biota in the situation of radioactive contamination compared a set of methods developed for human protection (ICRP [2009](#page-11-0), [2014](#page-11-0)). Doses of irradiation on animals and plants living in radioactively contaminated areas cannot be reduced by the resettlement, removal or removal of contaminated soil, etc. At the same time, the protection of non-human biota can be based on measures that while not directly affecting the source of radiation contribute to the conservation of biological diversity. In addition, when considering the possible approaches to ensuring the radiation safety of biota, it is necessary to study the mechanisms of the adaptation of animals and plants, relating them to the dose loads on specific populations, and also taking into account the interaction of the radiation factor with other environmental conditions.

In the first years after the Kyshtym disaster, environmental protection was not raised. The accident was classified as communal (agrarian), since agricultural and forest products containing radionuclides were the main source of human exposure. Fundamental studies of the migration and accumulation of radionuclides in living and stagnant components of ecosystems

Table 4 The total dose rate per plant organism for 14 herb species from the EURT zone and background sites Tier 2

Species	Total dose rate per:					
	Model of the upper part of plant	Model of root system	Whole plant			
Background 1						
Arctium tomentosum	$6.81E - 04$	$6.80E - 04$	1.36E-03			
Artemisia vulgaris	$8.74E - 04$	$8.69E - 04$	$1.74E - 03$			
Cirsium setosum	$1.14E - 03$	$1.12E - 03$	$2.26E - 03$			
Taraxacum officinale	$1.75E - 03$	$1.75E - 03$	$3.50E - 03$			
Stellaria graminea	$1.10E - 03$	$1.10E - 03$	$2.20E - 03$			
Melandrium album	$1.62E - 03$	$1.63E - 03$	$3.25E - 03$			
Lathyrus pratensis	$6.37E - 04$	$6.37E - 04$	$1.27E - 03$			
Melilotus officinalis	$1.26E - 03$	$1.27E - 03$	$2.53E - 03$			
Trifolium medium	$6.81E - 04$	$6.80E - 04$	$1.36E - 03$			
Leonurus quinquelobatus	$8.74E - 04$	$8.69E - 04$	$1.74E - 03$			
Bromus inermis	$1.14E - 03$	$1.12E - 03$	$2.26E - 03$			
Rumex confertus	$1.75E - 03$	$1.75E - 03$	$3.50E - 03$			
Plantago major	1.10E-03	$1.10E - 03$	$2.20E - 03$			
Urtica dioica	$1.62E - 03$	$1.63E - 03$	$3.25E - 03$			
Background 2						
Arctium tomentosum	$1.19E - 03$	$1.18E - 03$	2.37E-03			
Artemisia vulgaris	$2.63E - 03$	$2.60E - 03$	5.23E-03			
Cirsium setosum	$4.38E - 03$	$4.30E - 03$	$8.68E - 03$			
Taraxacum officinale	$7.34E - 03$	$7.34E - 03$	$1.47E - 02$			
Stellaria graminea	1.88E-03	1.88E-03	$3.76E - 03$			
Melandrium album	$5.01E - 03$	$5.05E - 03$	$1.01E - 02$			
Lathyrus pratensis	$1.74E - 03$	$1.74E - 03$	3.48E-03			
Melilotus officinalis	$3.12E - 03$	$3.14E - 03$	$6.26E - 03$			
Trifolium medium	$4.20E - 03$	$4.31E - 03$	$8.52E - 03$			
Leonurus quinquelobatus	1.78E-03	$1.75E - 03$	$3.53E - 03$			
Bromus inermis	$1.33E - 03$	$1.33E - 03$	$2.66E - 03$			
	$2.21E - 03$	$2.07E - 03$	$4.28E - 03$			
Rumex confertus			$1.15E - 02$			
Plantago major	$5.82E - 03$	$5.73E - 03$	$8.81E - 03$			
Urtica dioica Buffer 1	$4.38E - 03$	4.43E-03				
Arctium tomentosum	4.88E-02	4.86E-02	9.74E-02			
Artemisia vulgaris	$7.54E - 02$	7.49E-02	$1.50E - 01$			
Cirsium setosum	$1.09E - 01$	$1.08E - 01$	$2.17E - 01$			
Taraxacum officinale	$1.67E - 01$	$1.67E - 01$	3.34E-01			
Stellaria graminea	$5.33E - 02$	5.34E-02	$1.07E - 01$			
Melandrium album	$1.09E - 01$	$1.10E - 01$	2.19E-01			
Lathyrus pratensis	$6.25E - 02$	$6.25E - 02$	$1.25E - 01$			
Melilotus officinalis	$7.64E - 02$	$7.68E - 02$	1.53E-01			
Trifolium medium	$1.05E - 01$	$1.07E - 01$	2.12E-01			
Leonurus quinquelobatus	$6.09E - 02$	$6.05E - 02$	$1.21E - 01$			
Bromus inermis	5.42E-02	5.41E-02	1.08E-01			
Rumex confertus	7.18E-02	$6.91E - 02$	1.41E-01			
Plantago major	1.36E-01	$1.35E - 01$	2.71E-01			
Urtica dioica	$1.07E - 01$	$1.08E - 01$	$2.15E - 01$			

Distance from soil, cm	Integral flux of beta particles, mGy/month. (D $(\varphi) \pm SE$)			
	With the grass stand	After cutting the grass stand off		
0	$1.60 \pm 0.14^{\circ}$	1.02 ± 0.14		
10	1.0 ± 0.12	0.70 ± 0.11		
20	1.0 ± 0.13	0.68 ± 0.12		
50	0.6 ± 0.09	0.59 ± 0.14		
100	0.24 ± 0.08	0.34 ± 0.08		
Ω	1.30 ± 0.16	1.40 ± 0.20		
10	0.80 ± 0.16^a	1.14 ± 0.19		
20	$0.40 \pm 0.13^{\circ}$	1.00 ± 0.22		
50	0.30 ± 0.09^4	0.80 ± 0.17		
100	0.25 ± 0.09	0.40 ± 0.11		

Table 5 The comparison of the doses of external β-radiation measured in the grass stand and after cutting these plants off at the EURT sites (Pozolotina et al. [2012a](#page-12-0))

^a Significant differences in the indices of measurements with and without undisturbed grass stand. The measurements were performed with a BDPB-01 radiometer

were associated with applied problems, with the development of recommendations for reducing radionuclide intake into plants and animals. These works were extremely important in the future in the aftermath of accidents at the nuclear power plants in Chernobyl (1986) and Fukushima (2011).

At present, the assessment of the consequences of a radiation accident in the Urals shows that the forests and meadows in the EURT area have recovered completely. Community biodiversity is assessed even higher than in the adjacent areas (Lagunov and Smagin [2007](#page-12-0); Sokolov and Krivolutskiy [1993\)](#page-12-0). This is due to several reasons: (1) there is always a seed stock in the soil that can germinate when dose loads have decreased; (2) the territory of the zone is narrow (6–12 km wide), and the introduction of seeds from "clean" areas, like the migration of

Fig. 2 The comparison of CR_{wo} . soil values for grasses and herbs from the measured data (current article), ICRP report (Strand et al. [2009\)](#page-12-0) and IAEATRS-479 (IAEA [2014\)](#page-11-0)

animals, is possible; (3) perennial rhizome hemicryptophytes, with good protection of the buds of renewal, predominate in the EURT zone, and annuals are almost never found. The only countermeasure that contributed to the restoration of natural communities was the lack of anthropogenic load on this area (Pozolotina et al. [2012b](#page-12-0)). However, this does not exclude the possibility of active measures contributing to the restoration of natural communities in radioactive contamination.

The mean values of the strontium soil-to-plant transfer factor for herbs in the temperate environment were reported in Table 5 of TRS-479 as 9.8E−01 for all soil types (IAEA [2014\)](#page-11-0). Our CR data are in the same order of magnitude for most species (see Table [3\)](#page-6-0) including Artemisia vulgaris, Cirsium arvense, Taraxacum officinale, Melandrium album, Lathyrus pratensis, Melilotus officinalis, Trifolium medium, Leonurus quinquelobatus, Rumex confertus, Plantago major, and Urtica dioica. The other three species, Arctium tomentosum, Stellaria graminea, and Bromus inermis, had lower rates of CR by one order of magnitude. In contrast, no trend was observed in species specificity for $137Cs$. Radiostrontium CR rates were two orders of magnitude lower for all studied species than the rate published in TRS-479 for grasses and herbs (1.2E+0) (IAEA [2014](#page-11-0)). It should be noted that the TRS-479 report does not specify CR values for each isotope giving instead averaged data for all isotopes of one element. Our mean CR values of 239,240 Pu were in good agreement for most of the investigated herbaceous species with the mean transfer factor value of plutonium (1.6E−02) for grasses and herbs given in TRS-479 (IAEA [2014](#page-11-0)). There was only Lathyrus pratensis which had a mean value (see Table [3\)](#page-6-0) two orders of magnitude lower than that calculated by IAEA.

At the same time, the mean CR values of $239,240$ Pu reported by Strand et al. [\(2009\)](#page-12-0) were in agreement with our CR data gathered from Melilotus officinalis, Stellaria graminea, Melandrium album, and Arctium tomentosum. We observed decreased CR rates of 90 Sr and 137 Cs

compared to ICRP data for herbs. With a rare exceptions (e.g., CR value of 90 Sr in *Taraxacum officinale*), the distribution of our new measured CR data was below the rates for soil-to-plant transfer factor values of radiostrontium and radiocesium for the temperate environment (Fig. [2\)](#page-9-0) given in TRS-479 (IAEA [2014](#page-11-0)) and the ICRP report for CR values of plutonium in wild grasses in terrestrial ecosystems (Strand et al. [2009\)](#page-12-0).

All the estimated total dose rates per plant (see Table [4\)](#page-8-0) were below the dose rate screening value of 400 μ Gy h⁻¹ (USDoE [2002\)](#page-12-0) and higher than no-effect dose rate of 10 μGy h^{-1} predicted in the ERICA risk assessment (Garnier-Laplace et al. [2008\)](#page-11-0) for all herbaceous plants at the most polluted EURT zone. The ERICA Tool includes a tab summarising the radiobiological effects data as determined in conjunction with information from the FREDERICA radiation effects database (Copplestone et al. [2008](#page-11-0)). These data indicate the various effects in the Taraxacum species (dandelion) with rate ranges of 4–70 μ Gy h⁻¹. In our assessment, the total dose rate for the Taraxacum species was 93 and 199 μ Gy h⁻¹ at the Impact 1 and Impact 2 sites, respectively. In addition, another approach using Derived Consideration Reference Levels (DCRLs) indicated the possible radiation effects within the described exposure situation in the EURT zone. The ICRP described its approach to the protection of the environment (Copplestone et al. [2016](#page-11-0)) and derived DCRL for grass as $1-10 \text{ mGy/day}$ (i.e., 42 to 420 μ Gy/h) (ICRP [2008\)](#page-11-0). In fact, the total dose per 6 out of 14 species and 9 out of 14 species at the Impact 1 and Impact 2 sites, respectively, is within DCRL for grass.

At the buffer EURT sites, the average contribution to the radiation dose due to ⁹⁰Sr was 67%, ¹³⁷Cs − 31% and $239,240$ Pu – 1.7%. At the impact EURT sites, the domination of 90 Sr in the formation of the radiation dose increased (93%), while the contribution of ^{137}Cs and $^{239,240}Pu$ was minimal (6) and 1%, respectively). However, the 239,240 Pu decay period is considerably higher than that of $\rm{^{90}Sr}$ and $\rm{^{137}Cs}$. Therefore, the impact of radiation on non-human biota will persist for a long time in the EURT area.

The ERICA Integrated Approach should be used to assess the incremental doses from human activities only (Beresford et al. [2007\)](#page-11-0). However, natural background radiation acts on all ecosystems. Its contribution to the background sites was significant for herbs (0.1–0.2 μ Gy h⁻¹), while the ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu total dose rates ranged from 0.001 to 0.01 μ Gy h⁻¹ (see Table [4\)](#page-8-0). The opposite pattern was observed at the impact sites. Considering the summarised dose rates due to natural radiation and anthropogenic radionuclide pollution, our results point to a 48–977-fold increase in the dose rate per plant organism at the most polluted site compared to the background. These findings extend our understanding of the link between the anthropogenic impact and plant populations growing in the wild.

The biological effects of technogenic radiation were not the goal of this work; however, without assessing the dose rates on natural communities from the zones of radioactive contamination, it is difficult to understand the effects of radiation on non-human biota. One of the crucial challenges we undergo in our field studies to assess dose rates is that it is generally impossible to separate individual plants in natural populations of rhizome plants (cereals, for example) because they form a single cover, namely the sod. The roots of the individual parts are interconnected by rhizomes making the exchange of nutrients, and therefore radionuclides, possible.

Earlier, we compared the dose rates of β-radiation measured with a natural grass stand and after cutting these plants off at sites in the EURT zone (Pozolotina et al. [2012a\)](#page-12-0). In the herbage, two processes are superimposed. On the one hand, plant biomass causes the weakening of radiation from the soil surface (screening effect). On the other hand, radionuclides accumulated in plants contribute to radiation. Both morphological features of the species and their ability to accumulate radionuclides play an important role in the manifestation of the total doses. On sites where the Bromus inermis dominates, significant differences with undisturbed grass and without it are absent. At the same time, Urtica dioica leaves at an altitude of 10–50 cm create a significant screening effect (see Table [5\)](#page-9-0). The exposure dose of γ radiation with height has no change.

Based on data on radionuclide concentrations in different parts of plants, we calculated the contribution of incorporated radionuclides to the total irradiation dose of the herbage. Radionuclides accumulated in the rump biomass were added from 1.4 to 4.2% to the total dose, while the contribution of radionuclides in leaves and stems of nettle at an altitude of 40– 100 cm reached 22–27%. These results are in good agreement with the literature data (Bensen and Sparrow [1971](#page-11-0); Prister [2008\)](#page-12-0). The total dose of β-radiation from the loss of primary products of nuclear fission decreased by a factor of two regarding the height of cereal crops. In the lower tier, the contribution of irradiation from the soil surface predominated, and radiation from radionuclides accumulated by biomass prevailed in the upper tier (Prister [2008\)](#page-12-0). A survey of a meadow community polluted with 137Cs showed that the accumulation of radionuclides in the biomass leads to the formation of a volumetric radiation source (Bensen and Sparrow [1971](#page-11-0)).

Therefore, as a promising future direction, we would like to recommend ERICA developers to provide a user-friendly option for a dose calculation of the dose rates not only in individual plants, but in a whole grass stand.

Conclusions

This improved approach to the evaluation of radiation exposure confirms previous findings that herbaceous plant populations in the EURT area currently exist under low-level chronic exposure. A reassessment based on new expanded data suggests a 48–977-fold increase in the total dose rate per plant at the most polluted site as compared to the background. The highest capacity for the transfer of $137Cs$ and $90Sr$ was observed in T. officinale and P. major. The total dose rate per plant exceeded 150 μ Gy h⁻¹ in *T. officinale* and *P. major* due to ${}^{90}Sr + {}^{137}Cs + {}^{239,240}Pu$ radionuclide anthropogenic pollution in the EURT area.

At the buffer EURT sites, the average contribution to the radiation dose due to ⁹⁰Sr was 67%, $137Cs$ 31% and $239,240Pu$ 1.7%. At the impact EURT sites, the domination of 90 Sr in the formation of the radiation dose increased (93%), while the contribution of ^{137}Cs and $^{239,240}Pu$ was minimal (6 and 1% , respectively).

As a possible recommendation, we suggest the introduction into the program of an approach that takes into account the total doses not only for individual plants but for a grass stand. The calculation of the geometry of individual plants is very conditional, and grasses form in nature, as a rule, an organic whole, which is a grass stand of different specific density, and they can irradiate each other due to radionuclides accumulated in the aboveground mass.

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