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Occurrence of ^{137}Cs in Soil and Agricultural and Forest Products of the Contaminated Northeastern Part of the Czech Republic †

Petra Liszoková [1,*], Jan Škrkal [2], Barbara Stalmachová [1], Věra Záhorová [2] and Helena Pilátová [2]

[1] Faculty of Mining and Geology, VSB—Technical University of Ostrava, 17. Listopadu 2172/15, 708 00 Ostrava, Czech Republic; barbara.stalmachova@vsb.cz
[2] National Radiation Protection Institute (SÚRO, v. v. i.), Bartoškova 28, 140 00 Prague, Czech Republic; jan.skrkal@suro.cz (J.Š.); vera.zahorova@suro.cz (V.Z.); helena.pilatova@suro.cz (H.P.)
* Correspondence: petra.liszokova.staneckova@gmail.com
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Abstract: In the more contaminated northeastern region of the Czech Republic (Moravian-Silesian Region) in 2018 and 2019, soil samples and selected agricultural and forestry products were collected. The contamination of the environment was caused by radioactive cesium ^{137}Cs from the nuclear Chernobyl disaster, and the activity concentration of ^{137}Cs was determined in all soil samples taken, ranging from 0.177 Bq kg^{-1} dry matter (dm) to up to 299 Bq kg^{-1} dm, with an arithmetic mean of 38.4 Bq kg^{-1} dm. The activity concentrations of ^{137}Cs of agricultural and forestry products ranged from <0.02 Bq kg^{-1} to 1390 Bq kg^{-1} dm, and the transfer factors calculated based on these varied from 0.011 to 31 with an arithmetic mean of 3.4, with the highest values found in forest ecosystem products. The transfer factors and annual committed effective dose was calculated. It was statistically proven that the level of soil contamination with ^{137}Cs is related to the altitude and intensity of the precipitation in April and May of 1986, after the Chernobyl NPP accident.

Keywords: ^{137}Cs; radioactive contamination; soil samples; agricultural products; forestry products; transfer factor

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1. Introduction

Although 37 years have passed since the accident at the Chernobyl nuclear power plant (NPP), its legacy is still relevant, both for the continuous monitoring of the consequences and the preparedness for a possible similar situation, the risk of which has now significantly increased with the ongoing war and threat to the Ukrainian NPPs.

The Czech Republic (CR) belongs to the countries that were more significantly affected by the accident in Chernobyl. The Czech Republic (CR) is one of the countries that were more substantially affected by the Chernobyl accident. The mean value of the Chernobyl ground deposition of ^{137}Cs in the Czech Republic was 5 kBq m^{-2}, but, individually, the area exceeded 100 kBq m^{-2} [1–3]. The content of ^{137}Cs in soil immediately after an accident depends on local fallout conditions, e.g., the amount of precipitation, the prevailing direction of atmospheric flow, and the properties and ways of land use [4,5]. The impact of precipitation caused the passage of contaminated air masses across the Czech Republic from the northeast to southwest to release a large amount of ^{137}Cs in the areas of South Bohemia (southwest of the Czech Republic) and northern Moravia (northwest of the Czech Republic). This results in locally higher ground deposition levels, especially in some components of natural ecosystems.

As a result of the higher content of ^{137}Cs, there is a problematic situation with wild boars in the affected areas, where in the past period, animals significantly exceeding the limit values for meat consumption were captured; for example, a sample of wild boar with an activity of 4.9 kBq kg^{-1} was measured in the Olomouc region [6].

The transfer of Chernobyl cesium into plants depends not only on the content of ^{137}Cs in the root layer of the soil [7], but also on the nature of the radionuclide interaction with the soil and with the plants into which cesium passes through the soil solution. Also important are the chemical form of ^{137}Cs, the type of soil [8,9], the physico-chemical and biological properties of the soil [10], and the location of the soil (hill, hillside, slope, valley) [11]. In addition, ^{137}Cs is significantly better bound in organic soil matter with a higher mineral content, as when it is in soils with a high organic content, Cs is mobilized in soil solution [12,13]. It was calculated that there was an approximately tenfold increase in the transfer of Cs to plants when the organic matter content increased from 5 to 50% [14]. Clay minerals effectively reduce the migration of Cs in the soil [15], resulting in a lower uptake by plants [10]. It was stated that Cs is highly absorbed by clay particles and organic matter and is virtually non-exchangeable for other ions [16–18].

Cesium transfer can also be influenced by the type of plant and specific plant variety [19], as well as by the density of sowing [20]. The transfer of Cs is also influenced by other factors such as plant structure, metabolism, habitat and plant nutrition, climate, weather, and, last but not least, the character and form of agricultural activity [21], or even the activity of animals [22].

To evaluate the risk of the radionuclide transfer of radionuclide from soil to living organisms, a parameter called the transfer factor (TF) is used, which expresses the ratio of radionuclide activity concentration in the target matrix to activity concentration in the soil. Transfer factors (TF) expressing the degree of the radionuclide transfer of radionuclide from the soil to plant are normally at the level of 0.001 to 1 for agricultural crops, with values higher than 1 being achieved by plants on sandy and organic soils [22,23].

In order to evaluate the level of contamination in the North Moravian region, we carried out a survey of agricultural and intact soils, from which soil samples were taken and the content of ^{137}Cs was analyzed. Agricultural, wild plants, and mushrooms were also sampled at selected locations, where, in addition to the activity concentration of ^{137}Cs, the TF was also determined.

2. Materials and Methods

2.1. Sampling

The sampling of soils, agricultural and forest products, and permanent grass cover took place in the summer months of 2018 and 2019 under clear weather in the northeastern part of the Czech Republic. The samples were collected in the number of 1 to 3 samples on an area of about 100 m^2 at 73 sites, as shown in Figure 1, using a shovel and a spade from an area of 20 cm × 20 cm to a depth of about 20 cm. The total number of soil samples was 176. Sampling was carried out on an area of 4.328 km^2. The altitudes of the individual sites ranged from 199 m to 827 m above sea level, with an average of 372 m above sea level. The native weight of each sample was weighed immediately after collection. The average native weight of all samples was around 14.5 kg, depending on the water content in the sample. Subsequently, the sample was thoroughly mixed, dried at a room temperature range of 19 °C to 20 °C for 14 days, reweighed, mixed, and crushed. Then, each soil sample was sieved through a 3 mm mesh pedological sieve and reweighed. After sieving, the soil was weighed again. A representative portion with an average weight of approx. 380 g was taken from each sample prepared in this way to measure activity concentration. For all soil samples, cadastral classification was also determined. The altitude and geographical coordinates were determined using the map background of the altimetry analysis. The cadastral classification of soils was determined through the State Administration of Land Surveying and Cadastre, an online cadastral map [24].

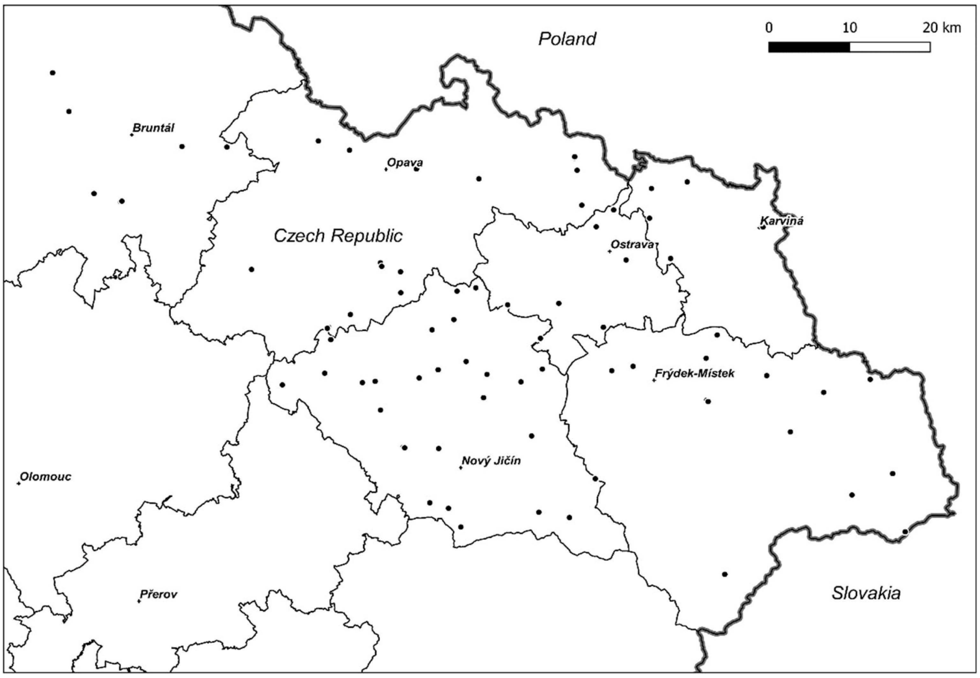

Figure 1. Map of soil sampling locations.

Forest and agricultural products were collected during the harvest period (September–October). Using a knife and a small shovel, the following items were sampled: cereals (*Triticum*), legumes (*Glycine max*), root crops (*Beta vulgaris*, *Solanum tuberosum*, *Apium graveolens*), annual fodder crops (*Zea mays*), oilseeds (*Brassica napus*, *Papaver somniferum*), grasses (*Festuca pratensis*), vegetables and root plants (*Brassica oleracea* var. *Capitata*, *Armoracia rusticana*), fruit (*Maleventum*), angiosperms (*Rosa canina*, *Sorbus aucuparia*), gymnosperms (*Pinus sylvestris*), fungi (*Lactarius lignyotus*, *Leccinum holopus*, *Neoboletus luridiformis*, *Amanita rubescens*) and spore-bearing plants (*Bryopsida*). In most cases, the sample consisted separately of the plant part intended for consumption and the rest of the plant body. In other cases, the plant part and the plant body were measured together within one sample. The samples of forest and agricultural products were first dried at room temperature and then in a biomass dryer at 52 °C with a drying time of 720 min. The collected forest and agricultural products were ground into pieces of max. 10 cm in length using a knife and cleaver. For the remaining biomass samples, only fruits, the plant parts of spore plants, and grasses were taken and measured.

2.2. Measurement of ^{137}Cs Activity

The ^{137}Cs activity in all dried samples (see Section 2.1) was measured at the National Radiation Protection Institute (SÚRO, v. v. i., Czech Republic) by semiconductor gamma spectrometry on HPGe detectors with efficiencies of 10–150%. The measurement time was chosen to achieve the uncertainty of ^{137}Cs determination lower than 10%, but no more than 1 week; if the activity was below the detection threshold, a minimum significant activity (MSA) was determined [25]. Both plant and soil samples were measured in 200 mL cylindrical plastic vessels and 500 mL Marinelli beakers on the detector. In the case of a larger sample, the cylindrical containers were around the detector. The energy and efficiency calibrations were completed using gel standards. Since the density of the vegetable and soil samples mostly varied from gel density, a correction for self-absorption had to be made. The measurement method is accredited. The residual moisture and dry matter were determined in a small portion of each sample at 105 °C. The ^{137}Cs activity in all measured samples are related to the sampling date and recalculated for dry matter.

2.3. Data Processing

The maps were created in the QGIS [26] and SAGA GIS [27] programs. The vector polygon layers of the administrative division of the Czech Republic come from the Register of Territorial Identification, Addresses and Real Estate (RÚIAN) (Czech Office for Surveying, Mapping and Cadastre—ČÚZK) [24]. The point data were transferred to the surface map by the Multilevel B-Spline interpolation method in the SAGA GIS program [27].

The transfer factors (TF) were calculated as the ratio of the activity concentration of ^{137}Cs in the dry matter of agricultural and forestry products to the activity concentration of ^{137}Cs in the dry matter of the soil. The IAEA (2010) [28] refers to TF as a concentration ratio.

Using the Excel office program, statistical characteristics were determined: simple arithmetic and geometric means (AM, GM), arithmetic and geometric standard deviations (SD, GSD), the minimum and maximum for soil activity concentration, and transfer factors, which were further divided into groups. Only data with a specified activity of ^{137}Cs higher than MSA were included in the calculation.

3. ^{137}Cs Activity Concentration in Soils

A total of 176 soil samples were taken from 73 localities and 6 districts: Bruntál, Opava, Nový Jičín, Ostrava, Karviná and Frýdek-Místek. Table 1 shows the statistical characteristics of ^{137}Cs activity concentration in all soil samples and in the groups of soils divided by cadastral classification.

Table 1. Statistical characteristics of ^{137}Cs activities in soils divided by cadastral classification.

	All	**pg**	**Arable Land**	**Other Land**	**Forest**
Number of samples	176	29	52	42	53
AM [Bq kg^{-1} dm]	38.4	21.2	18.1	19.2	84.4
SD [Bq kg^{-1} dm]	57	21	12	19.41	84
AM_P [Bq m^{-2}]	11,213	6190	5285	5198	24,645
GM [Bq kg^{-1} dm]	16.8	14.5	13.3	11.0	37.8
GSD	3.97	2.44	2.52	3.18	5.61
GM/AM ratio	0.44	0.69	0.74	0.57	0.45
minimum [Bq kg^{-1} dm]	0.177	2.50	0.438	0.52	0.177
maximum [Bq kg^{-1} dm]	299	106	52.2	75	299
max/min ratio	1690	42	119	144	1690

Notes: pg is permanent grassland, AM_P [Bq m^{-2}]—the value was calculated using the mean soil density in the Czech Republic, which is 1460 kg/m^3, and a soil abstraction depth of 20 cm, according to the relationship $AM_P = AM \cdot 292$.

Overall, the activity concentration of ^{137}Cs lay in a wide three-order range, with the lowest activity concentration being 0.177 Bq kg^{-1} dry matter (dm) measured in the forest soil of the locality Studénka nad Odrou, and the highest ^{137}Cs activity being 299 Bq kg^{-1} dm, which was measured in the forest soil of the Spálov site. The overall arithmetic mean was equal to 38.4 Bq kg^{-1} dm. The results of the calculations for cadastral classification show that the highest mean activity concentration value of ^{137}Cs was, as expected, in the forest land system (AM 84.4 Bq kg^{-1} dm). The mean activity concentration of ^{137}Cs was about 4 times higher in forest soils than in permanent grasslands and arable and other soils. The arithmetic means for permanent grassland (21.2 Bq kg^{-1} dm), arable land (18.1 Bq kg^{-1} dm), and other land (19.2 Bq kg^{-1} dm) are very close, with the arable and other lands corresponding to systems disrupted by human activity.

Since no area activity was determined for the removed soils, to compare with the total soil deposition density of ^{137}Cs in CR, we calculated the mean area activity of AP_P from the arithmetic mean of the activity concentration of AP, the mean density of soils in the Czech Republic, and the depth of sampling. The results are presented in Table 1. The arithmetic mean of 5 kBq m^{-2} [1] in the post-Chernobyl soil survey was obtained in the CR mainly on unshaded and untouched soils, so this value corresponds most closely to permanent grasslands with AP_P 6.19 kBq m^{-2}. Given that 32 years have passed since 1986 to 2018, the decrease in ^{137}Cs should be, due to physical transformation, at least 50%. In terms of other environmental factors, the mean activity in the CR should now be 2.5 kBq m^{-2} or lower. The level of soil contamination in the region we monitored was therefore about 2.5 times higher than in relation to the whole Czech Republic.

The higher value of ^{137}Cs cannot be attributed to the nuclear accident in Fukushima, which affected the CR only minimally. Nor can it be attributed to the global fallout from nuclear weapons tests from 1945 to 1963 [29], which was approximately 5 kBq m^{-2} relatively homogeneously dispersed at our latitudes (40–50° N and 50–60° N) (calculated from UNSCEAR [30]). Figure 1 shows a map of sampling points covering six districts. Activity concentrations were put on a map [Figure 2]. In the largest part of the territory, the activities were less than 40 Bq kg^{-1} dm. The areas of higher contamination are in the Opava region and in the northwest of the Bruntál region, with activity concentrations exceeding 160 Bq kg^{-1} dm, which corresponds to a surface contamination of 46 kBq m^{-2} and higher. A place with higher activity was also found in the east of the Frýdek-Místek region.

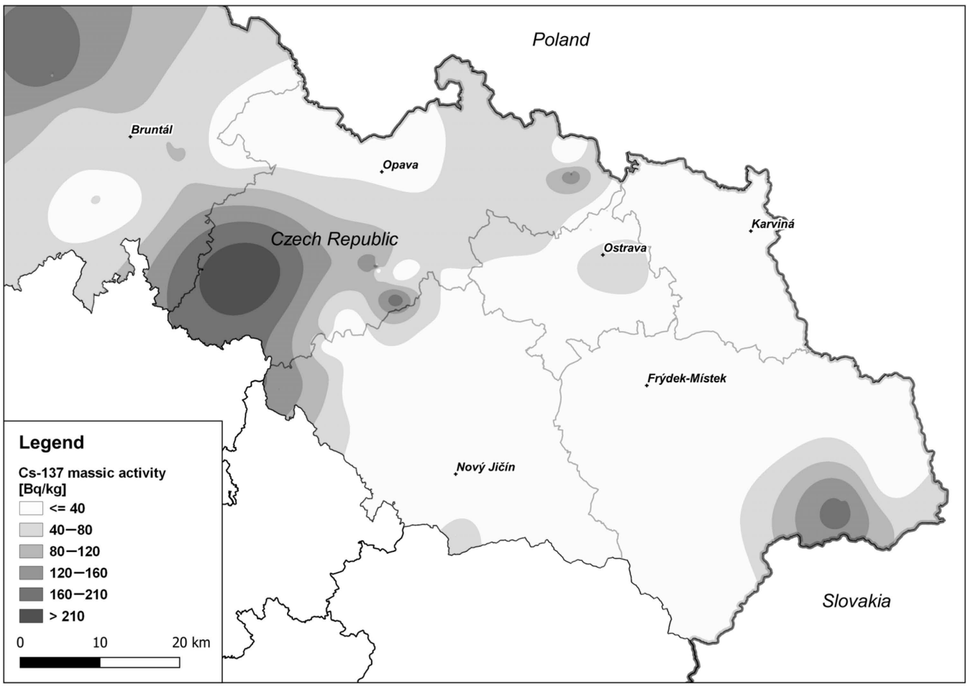

Figure 2. Map of ^{137}Cs activity concentration in soil.

The higher contamination of the Opava and Bruntál regions corresponds approximately to the map of Chernobyl contamination [2], on which the surface activity locally reached 20–40 kBq m^{-2} in these regions.

On the map [2], the region of Ostrava is also more contaminated, locally and with activity reaching the interval of 40–100 kBq m^{-2}. The differences must be attributed to the insufficient sampling density, not only ours but also the Chernobyl one (776 samples for the entire CR), as well as the inaccuracy associated with interpolation.

Due to the higher activity of ^{137}Cs in heavily sloping soils, all values were put into a graph depending on altitude. Figure 3 shows a statistically confirmed upward trend fitted with a linear slope ($p < 0.000$).

The fact that areas with higher altitudes were more contaminated is probably related to the higher frequency of precipitation in these areas in April and May 1986. The altitudes were divided into groups according to altitudes of 100–199 m above sea level (ASL), 200–299 m ASL, 300–399 m ASL, 400–499 m ASL, 500–599 m ASL, 600–699 m ASL, 700–799 m ASL, and 800–899 m ASL. For each interval, the arithmetic mean of the precipitation totals in April and May of 1986 was calculated, and related to half of the relevant altitude interval. The result was a statistically confirmed upward trend ($p < 0.000$).

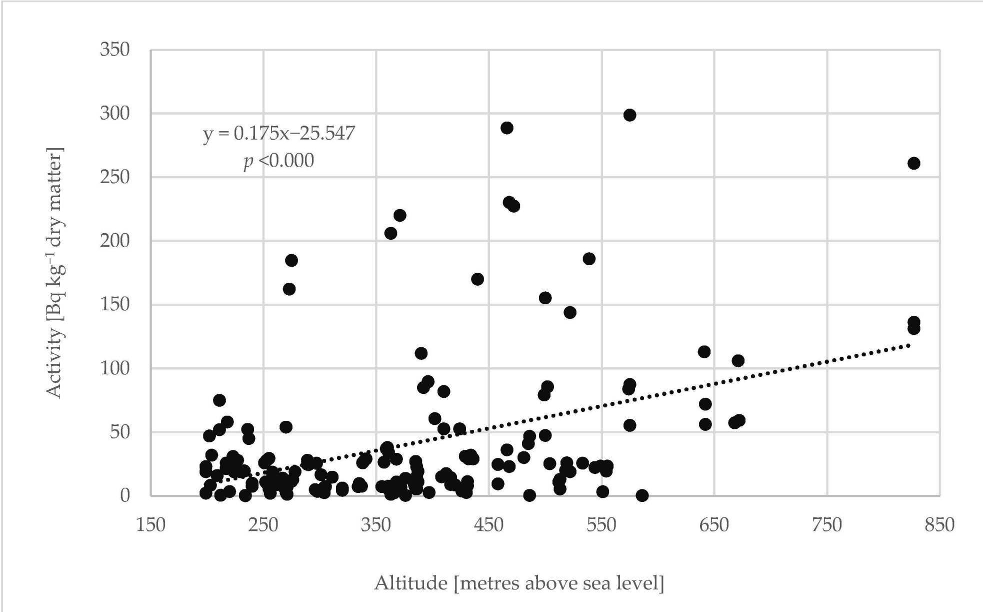

Figure 3. Dependence of ^{137}Cs activity concentration in all soils on the altitude.

The dependence of ^{137}Cs activity on the precipitation totals was also tested. In the vicinity of the soil sampling points, there are 39 weather stations, with available precipitation totals for April and May 1986 [31]. To calculate the values of the precipitation totals, the precipitation for the months of April and May in 1986 was averaged, and the nearest weather station to each sampling location was assigned to construct graphs of the dependence of ^{137}Cs activities in the soil at the altitude of the sampling point. The upward trend for areas with higher precipitation totals has been statistically confirmed ($p < 0.045$).

4. Results of Measurements of Agricultural and Forestry Products

A total of 28 biomass samples were measured. Three plant species were divided into two specimens: the part intended for consumption and the rest of the plant body. One sample contained two types of mushrooms mixed together. In 7 samples of agricultural products and plants of permanent grassland (sugar beet root, wheat grains, potato tubers from two sites, rose hip, soybean, and horseradish), it was not possible to measure the activity of ^{137}Cs, and therefore MSA (designation "<") was determined. The lowest activity concentration of ^{137}Cs < 0.02 Bq kg^{-1} dm was detected in a sample of sugar beet root; the highest ^{137}Cs activity 1390 Bq kg^{-1} dm was measured in a mixture of blacksmith's boletus and rosacea toadstool. The arithmetic mean of the activities of ^{137}Cs in plant samples was 89.4 Bq kg^{-1} dm. The arithmetic mean in the mushroom samples was 821.5 Bq kg^{-1} dm.

5. Transfer to Agricultural and Forestry Products

A total of 23 biomass-soil pairs were available for the calculation of transfer factors. In Tables 2 and 3, the TFs are sorted by cadastral division. Most biomass samples (14) were taken from arable lands, 2 from permanent grasslands, and 7 from forests.

On arable lands, the mean value of TF was equal to 0.12. The TF range agrees with the typical range of 0.001 to 1 for agricultural crops [23]. The determined values of TF [Table 2] are generally approximately at the level of the mean values presented in [30]. Higher values were found in wheat stalks (0.69), poppy seeds (0.11), and rapeseed stalks (0.1), while low values were found in sugar beet root (<0.0046) and fescue (0.012).

Table 2. Transfer factors (TF).

Title	Latin Name	Activity Concentrations Soil ^{137}Cs [Bq kg^{-1} dm]	TF	Cadastra Classification of Land
sugar beet—plant body	*Beta vulgaris*	25.6	0.031	acreage
sugar beet—root	*Beta vulgaris*	25.6	<0.0046	acreage
sugar beet—leaf	*Beta vulgaris*	8.7	0.077	acreage
wheat—stem	*Triticum*	16.0	0.036	acreage
wheat—grain	*Triticum*	16.0	<0.081	acreage
wheat—stem	*Triticum*	16.0	0.69	acreage
soybeans—beans	*Glycine max*	25.3	<0.024	acreage
soybean legume—stem	*Glycine max*	25.3	0.032	acreage
meadow fescue/hay—stem	*Festuca pratensis*	17.5	0.012	acreage
White cabbage—plant body	*Brassica oleracea* var. *Capitata*	29.5	0.041	acreage
rapeseed—stem	*Brassica napus*	45.0	0.1	acreage
poppy—poppy	*Papaver somniferum*	22.3	0.13	acreage
potato eggplant—plant body	*Solanum tuberosum*	7.50	<0.16	acreage
apple—whole fruit	Maleventum	52.2	0.011	acreage
wine rose—fruit	*Rosa canina*	14.7	<0.054	permanent grassland
horseradish—plant body	*Armoracia Rusticana*	9.4	<0.12	permanent grassland
moss	*Bryopsida*	170	0.11	forest land
moss	*Bryopsida*	30.1	0.17	forest land
moss	*Bryopsida*	60.0	2	forest land
Rowan—brood	*Sorbus aucuparia*	26.5	0.52	forest land
black grouse—whole fruit	*Lactarius lignyotus*	47.3	5.3	forest land
Boletus blacksmith + toadstool rosacea—whole fruit	*Neoboletus luridiformis + Amanita rubescens*	79.2	18	forest land
pine cones—whole fruit	*Pinus sylvestris*	2.7	31	forest land

Note: < sign is followed by the value of the transfer factor (TF) calculated from the minimum significant activity by weight of dry matter of forest and agricultural products.

Table 3. Statistical characteristics of transfer factors divided by cadastral focus.

	All	Arable Land	Forest Land
number of samples	17	10	7
AM	3.4	0.12	8.2
SD	8.4	0.20	12
GM	0.22	0.051	1.8
GSD	11.4	3.39	9.21
GM/AM ratio	0.064	0.44	0.22
minimum	0.011	0.011	0.11
maximum	31	0.69	31
max/min ratio	2990	65	296

The transfer factor for rapeseed of 0.10 came out approximately the same as the arithmetic mean of the transfer factors for the same plant of 0.13 [32]. The transfer to fruit trees is an order of magnitude lower, and the value (0.011) of the fruit of the apple tree is about twice as high as the mean value (0.0058) [31].

Tyson et al., 1999 [33], showed that ^{134}Cs is unevenly distributed in plants. Its higher concentration was found in young shoots and growth tissues. Radionuclides often transfer in higher concentrations to stems and leaves than to the generative parts of plants [31]. There are also differences in the values of TF found in our results.

The influence of the change in soil composition on the transfer is evident when comparing arable soils with forest soils, which are often characterized by a thick layer of overlying humus. While the total AM transfer factors for all samples were equal to 3.4, the TF for forest land was very high (AM 8.2), and the TF in arable land samples was an order of magnitude lower (AM 0.12). Also, the maximum-to-minimum ratio is higher for forest lands, 4.5 times higher compared to arable lands. The transfer to plants mainly reduces the content of clay minerals, and vice versa, it increases the amount of organic matter. In highly organic and peaty soils, cesium is weakly adsorbed on organic surfaces and can be easily desorbed into soil solution [12,13,34]. In addition, in forest soil, TF increases at a lower pH. A higher transfer to blueberries that prefer acidic soils is known. IAEA [35] described a trend in barley and cabbage towards a decrease in TF ^{137}Cs with an increasing pH. TF increases in the transfer of Cs from humus to spruce bark with a decrease in pH (H_2O) and pH (KCl) [36].

The lowest values of TF in forest soils were within the range of 0.11–2. Interestingly, the TF in pine cones was really high (31); it was even higher than the TF in mushrooms (5.3; 18), to which ^{137}Cs usually accumulate highly. This may be due to the longer reach of the pine roots and, accordingly, their ability to draw nutrients from a more distant place with higher soil activity.

6. Committed Effective Dose

As mentioned in the introduction, the monitored region was one of the most contaminated in the Czech Republic by the post-Chernobyl fallout. To evaluate the exposure risk, from the observed concentration activities, we can approximately estimate the committed effective dose of adults from plant agricultural products and compare it with the mean value of the dose for the whole Czech Republic. In 2015, the geometric mean of the total annual intake of ^{137}Cs of food was 110 Bq, which, when multiplied by a conversion coefficient of 0.013 μSv/Bq, corresponds to an effective dose of 1.4 μSv [36]. Plant agricultural products (vegetables, fruits, cereals, and potatoes) accounted for 4.6% and mushrooms for 76% of the total intake of ^{137}Cs. Thus, the corresponding mean effective dose in 2015 was 0.065 μSv for plant agricultural products and 1.1 μSv for fungi. The effective dose of agricultural crops was calculated from the activity concentrations above the MSA of white cabbage, sugar beet, apple, poppy, beetroot leaves, wheat stalks, leguminous soybean, and oilseed rape for our samples. The geometric mean of ^{137}Cs activity on a fresh basis is equal to 0.37 Bq/kg for our samples grown on agricultural soils. Using the annual consumption of fresh plant agricultural products in the Czech Republic of 181 kg, the annual mean effective dose of adults is 0.9 μSv. For our two mushroom samples, with a mean annual consumption of 2.8 kg and a mean fresh activity concentration of 62.3 Bq/kg, the effective dose is 2.3 μSv.

It can therefore be seen that the mean committed effective dose from ^{137}Cs from plant agricultural products for our area of interest is significantly higher, about 14 times the national mean. The effective dose from mushrooms is approximately twice as high. Of course, there is a time gap, and nationwide doses will be slightly lower today, but due to the currently very slow decrease in the activity of post-Chernobyl ^{137}Cs in agricultural ecosystems, the difference between the doses in 2015 and 2018–2019 is only slight. The observed higher values of the effective dose for agricultural products in our region are consistent with the higher soil contamination discussed above, although the ratio of the effective dose of our region to the rest of the Czech Republic is higher, which may be due to the relatively small number of samples and the associated lack of representativeness.

7. Conclusions

The research analyzed 176 soil samples from six districts in the northeast of the Czech Republic: Bruntál, Opava, Nový Jičín, Ostrava, Karviná, and Frýdek-Místek. For ^{137}Cs activity concentration, 28 biomass samples were also measured.

The average activity concentration of ^{137}Cs in the soil was 38.4 Bq kg^{-1}, while the highest value was found in the soils of forest ecosystems and the lowest in arable soils. The average activity in grass soils was 21.2 Bq kg^{-1}, which corresponds to a surface activity value of 6.19 kBq m^{-2}, which is at least 2.5 times higher than the surface activity of soil in the Czech Republic. The connection between the increase in the activity concentration of the soil with the increasing height above sea level and the intensity of precipitation totals confirms that mountain areas with lower agricultural production are the most contaminated.

The activity concentrations of ^{137}Cs in agricultural and forest products ranged from <0.021 Bq kg^{-1} to 1392 Bq kg^{-1} dry matter, which corresponded to the value of the transfer factor <0.0046 to 31, with the highest transfer to products of the forest ecosystem. Transfer factors were influenced by the plant type, part, and soil composition. The calculated annual effective dose from agricultural products was 0.9 µSv, i.e., approx. 14 times higher than the average for the Czech Republic, which agrees with the higher contamination of the northeast of the Czech Republic.

The results of our experiments therefore confirmed a higher occurrence of ^{137}Cs both in the soil and in plant products. Considering that the detected value is not representative due to the limited number of analyzed samples, it is necessary to further investigate similar areas not only in the Czech Republic, but also elsewhere in the world, and thus protect against the consumption of both contaminated agricultural products and products of forest ecosystems.

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