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Title: Turnover of 137 Cs in 'soil-tree' system: an experience of measuring the isotope flows in a Siberian conifer forest

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Abstract: Little attention has been paid to the uptake of 137Cs in natural forests under low levels of the isotope fallout when no immediate ecological danger presents. Here we present the extended assessments of the soil-to-plant and canopy-to-litter flows of 137Cs recently evaluated in a native Siberian forested area. The area undergoes a typical after-fire long term succession, with light-conifer upper story being followed by undergrowth of Siberian fir and other dark-conifer species. The one-year-old needles of Siberian fir were found to accumulate the largest share of the isotope, 4.10 Bq/kg oven-dry weight during the first growth season, as compared with older needles that accumulated 4.67 Bq/kg oven-dry weight in 2-3 years of growth. Based on these data a new approach for estimation of the specific 137Cs activity in soil solutions was developed. The isotope activity in soil solutions was estimated to be 0.0061-0.0105 Bq/L. Based on the original data from the litter fall the annual flow of the isotope from the upper canopy to on-ground litter was found to be 0.42-0.84 Bq/m2. The stock of 137Cs that returns yearly back from canopy with falling litter was estimated to be 0.012 - 0.015% of the total soil isotope stock. A combination of the estimations obtained in our study with the values of global 137Cs fallouts allowed us to assess the ages (the time of formation) of horizons of the soils in the area.

Turnover of 137 Cs in 'soil–tree' system: an experience of measuring the isotope flows in a Siberian conifer forest

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***Highlights (for review)**

- Uptake of ^{137}Cs in forests under low fallout levels has received little attention.
- Most of ^{137}Cs is accumulated in one-year-old needles of Siberian fir.
- An approach was suggested to estimate ^{137}Cs activity concentrations in soil solutions.
- Flow of isotope from canopy to ground was found to be smaller than global fallout.
- Based on ^{137}Cs activity a method is suggested to estimate ages of soil horizons.

Turnover of ^{137}Cs in 'soil–tree' system: an experience of measuring the isotope flows in a Siberian conifer forest

Abstract

Little attention has been paid to the uptake of ^{137}Cs in natural forests under low levels of the isotope fallout when no immediate ecological danger presents. Here we present the extended assessments of the soil-to-plant and canopy-to-litter flows of ^{137}Cs recently evaluated in a native Siberian forested area. The area undergoes a typical after-fire long term succession, with light-conifer upper story being followed by undergrowth of Siberian fir and other dark-conifer species. The one-year-old needles of Siberian fir were found to accumulate the largest concentration of the isotope, 4.10 Bq/kg oven-dry weight during the first growth season, as compared with older needles that accumulated 4.67 Bq/kg oven-dry weight in 2–3 years of growth. Based on these data an approach was developed that, hypothetically, can allow one to estimate the ^{137}Cs activity concentration in soil solutions. Direct activity measurements in the soil solutions were not possible. The isotope activity in soil solutions was estimated to be 0.0061-0.0105 Bq/L. Based on the original data from the litter fall the annual flow of the isotope from the upper canopy to on-ground litter was found to be 0.42–0.84 Bq/m^2. The amount of ^{137}Cs that returns yearly back from canopy with falling litter was estimated to be 0.012 – 0.015% of the total soil isotope content. A combination of the estimations obtained in our study with the values of global ^{137}Cs fallout allowed us to assess the ages (the time of formation) of horizons of the soils in the area.

Introduction

As a result of nuclear weapon testing a total of 948·10^{15} Bq ^{137}Cs was released to the Earth atmosphere (UNSCEAR,2008). This amount of the isotope is unevenly dispersed all over the globe and provides the global atmospheric fallout. The uneven dispersion was a result of two factors, locations of testing areas and stratospheric convection. It has been also understood that the intensity of the depositions decrease from southern to northern regions in the temperate zone of the northern hemisphere (Baskaran et al., 1991; Baskaran et al., 1996). The depositions lead to that ^{137}Cs has been found in practically all the biosphere components – waters, bottom sediments, soil, living organisms etc.

Due to obvious importance of a possible effect on humans the studies of ^{137}Cs have mostly focused on transfer of the isotope from trees to edible plants and fungi. Many studies focus on biogeochemical fluxes of radioactive isotopes in terms of either dose estimation or radionuclide migration rates in various food chains (Oolbekkink and Kuyper, 1989; Fogh and Andersson, 2001; Fesenko et al., 2001; Soukhova et al., 2003; Goor and Thiry, 2004; Mosquera et al., 2006; Lehto et al., 2013; Vinichuk et al., 2010; Bulko et al., 2014).

Little attention has been paid to the uptake of ^{137}Cs in natural forests under low levels of the isotope fallout when no immediate ecological danger presents. It is relevant for various forested areas remote from places of nuclear power station accidents or old nuclear testing areas. Virgin dark-coniferous taiga forests may be of particular interest because they present naturally evolving ecosystems with minor human intervention. In the literature, sufficient attention has been paid to calculate the ^{137}Cs soil-to-plant transfers in forest ecosystems while in natural forests there is a constant flow of plant parts from canopies back to the on-ground litter and soil layers. Contribution of ^{137}Cs from needles back into the soil is of interest when investigating the duration of the isotope in a forest ecosystem (e.g. calculating of ecological half-life).This kind of information may be of importance for research purposes such as studies of forest ecosystem functioning and carbon balances in forests. Even if the ^{137}Cs fallout are of low or ‘background’ values there are measurable amounts of the isotope in soils accumulated in the past years. Furthermore, forest-soil ecosystem cycling may result in concentration of the isotope to sufficient levels in particular compartments of the ecosystem. In such low-fallout forested areas, valuable results may be obtained that elucidate biogeochemical cycle functioning and serve as a basis for erosion monitoring. Also, recordings of levels of ^{137}Cs activity in landscapes may be integrated into databases of international programs such as EMRAS II (Stocki et al., 2011), MODARIA (IAEA, 2012-2015), BORIS (Tamponnet et al., 2008).

The aims of the study was to answer a number of questions: I) what is the level of current ^{137}Cs activity in the forested area, II) what are the estimated rising and descending isotope flows in the plant-soil system and III) are the current contents of the isotope sufficient in the soils to use the isotope as a soil erosion tracer.

Methods

Area and objects of study

The study presented was focused on the levels of ^{137}Cs accumulation in various compartments of forest ecosystems in the State Nature Reserve 'Stolby' (fig. 1). This pioneer research was one of the first attempts to study the isotope turnover in undisturbed forested areas of Middle Siberia. Distribution of the isotope in the undisturbed forests should be of special interest because they turned out to be first Siberian forested areas that were subjected to the fallout and that included the isotope into the biological turnover. The relatively earlier exposure of the forests to ^{137}Cs fallout is partly explained by their geographical position.

The Semipalatinsk nuclear test site was located to the south-west from the Nature Reserve and the prevailing winds have been from the same direction. On the test site, a series of USSR nuclear testing started 1949 and by 1962 a total of 124 nuclear explosions have been there performed, both on-ground and atmospheric tests. This source has thus made the main contribution into radioactive contamination of our study area. The intensity of the fallout when the radioactive clouds from first tests crossed the area was registered by weather stations in the southern part of Krasnoyarsk territory. These fallout amounted up to 23700-74700 Bq/m^2 in one day (Dubasov et al., 1994).

The radioactive fallout from other USSR tests and accidents as well as of other states might give a contribution to the amounts of ^{137}Cs in our study area but they played a minor role. For example, the deposition of Chernobyl-derived ^{137}Cs in soils of Siberia is no more than 5% (Sukhorukov et al., 2000). A time span of over 45 years has passed since the period of intensive fallout (Walling and Quine, 1993) and according to the Treaty Banning Nuclear Weapon Tests in the Atmosphere, in Outer Space and Under Water no more on-ground tests were performed. In the subsequent years, only underground explosions were done.

Nowadays the protected undisturbed forested areas allow researchers to study the ^{137}Cs flows from the standpoints of remote consequences of radioactive fallout as well as ecosystem functioning of forests with complex structure.

The measurements were performed in a forested area within the State Nature Reserve ‘Stolby’ located south-west from Krasnoyarsk city on the right bank of Yenisei River (fig. 1). Center of the area is given by the global coordinates (WGS84) as E 92º42,587’ N 55º54,897’. The study was based on four permanent plots having sizes of 0.25 ha each.

According to climatic classification climate Koppen-Geiger classification Dfc. Annual precipitation over the region amounts 679,5 mm. The mean annual temperature is –1.3 °C. The vegetation period spans 138 days. Snow cover occurs about 200 days, with the mean depth being 85 to 105 cm (Fokina et al., 2006).

The Nature Reserve ‘Stolby’ was established in 1925 but its contemporary boundaries were set up only 1946 and its area amounts now 47.2 thousand ha. Before 1946 the territory was subjected to tree felling and fires were regular in the northern part of the Nature Reserve. Many of forest stands over the area present typical for Southern Siberia forest communities covering low-mountain areas in West Sayan and East Sayan mountain systems (Smagin et al., 1980).The forests often undergo a succession from a light-coniferous stage to a dark-coniferous one. The upper canopy is presently dominated by Scots pine (*Pinus sylvestris* L.) and to a lesser extent by Siberian larch (*Larix sibirica* Ledeb.). Among these canopy species sporadic specimen of Siberian fir (*Abies sibirica* Ledeb.) and Siberian spruce (*Picea obovata* Ledeb.) may be found. At the same time, the youngest tree layer is practically all dark-coniferous, mostly Siberian fir with addition of Siberian spruce and Siberian pine (*Pinus sibirica* Du Tour).A more detailed picture of the forest stands is given in frequency height distributions provided in the Electronic Supplement (fig. S2–S5).

Fig. 1

Over the area of study, the forests grow on gravelly loamy haplic cambisols dystric, dominating soil types, and lithic leptosol soils that have a lesser distribution. The soil types are given after World reference base for soil resources (2014). All the soil profiles are characterized by the presence of a not significant layer of spruce litter (O, further subdivided into L, F, H) with average thickness of 3-5 cm. In general, the soils are shallow or medium deep, rarely exceeding the depth of 40 cm. The most frequent colorations of the soil horizons are brownish gray (7.5 YR 4/1 - 5/1) and grayish brown (7.5 YR 4/2 – 5/2) (Munsell Soil Colour Charts, 1974). In the cambic horizon, spots of reddish colors (5YR) randomly appear. The concentration of fine clay minerals (fraction < 0.01 mm) in the soil varies between 10% and 26%. The dry density of soils varies within the range of 1.01-1.35 g/cm^3. Particle size distribution of the soils was measured after Kaczynski method (Kaczynski, 1958). The content of humus (measured after the Tyurin method) (Arinushkina, 1970) varied in the range of 9–13%. Soil characterized by dominance of fulvic acids (FAs) on HAs, while the ratio of HA to FA is about 0.8-0.9. The pH_{KCL} value varied from 4.9 to 5.3 in the haplic cambisols dystric soils.

Young undergrowth trees of Siberian fir, 2 to 4 m height, were taken as the object to study the biological soil-to-plant uptake of ^{137}Cs. The reasons of the choice were that the species was the dominant in the undergrowth. Also, the upper canopy of large trees can serve as a shield that is first to absorb the fallout coming from the atmosphere (Wedding et al., 1975; Nimis, 1996). The relative activity of the isotope in the small fir trees may be therefore supposed to be largely due to flows from the soil.

Twigs with needles were gathered up to the height of 4 m uniformly around the crowns in July 2014 when the newly grown twigs almost reached their final size but were clearly different in color (light-green) from the older twigs and later in October 2014. The same method was applied in 2015 as well.

In the laboratory, the twigs of the current year (below referred to as 1-year-old twigs) were separated from twigs aged 2 and 3 years. The twigs and needles were then oven-dried at 60°C for 24 h and then dried again for 24 h at 105°C. They were further weighted and incinerated in oven at 400 °C according to recommendations by Ermakov (1972).

Samples of above-ground grass cover were gathered from 60 0.5m x 0.5 m plots regularly distributed. The grass plants were oven-dried, weighted and incinerated.

Under the canopy of the studied forest stand, the litter falling from the crowns was gathered as follows. Pieces of 2 mm mesh fiberglass cloth were anchored to ground by plastic studs. Six pieces of the cloth of 5 m^2 each were regularly distributed in the forested area on 25th of July 2014. After the end of the exposition, on 10th of October 2014, the pieces of cloth were rolled with all the content and transported in plastic bags to laboratory where the litter was manually separated from the cloth. After the separation, the litter was dried in drying oven, weighted and incinerated. The same method was applied in 2015 as well.

The soil samples were collected in July 2014 by means of a 120 mm diameter borer. Each reference sample was a composite bulk sample from 3 cores sampled within 1 m^2. In every sampling location, the litter was manually cut with a knife and put into a plastic box. The samples were collected from 1–6 cm to the depth of 50 cm. Outside the sample plots, the depth of sampling ranged from 40 to 70 cm. Each core was sectioned into soil horizons and their depths were recorded. Additionally, the litter material was divided into 2 cm slides. All samples were first dried until their mass was constant. Afterwards samples were carefully mixed.

Measurements of isotope activity

The isotope activity was measured through a spectrometry approach. The spectra were received with the help of scintillation spectrometer MKGB-01 ‘RADEK’ with the BDEG-63 NaI (Tl) detector. For soils and pre-incinerated plant material Marinelli containers of 1 dm^3 and 0.25 dm^3 volume, respectively, were used. The density of measured specimen was always in the optimal range of 1.25 to 1.6 kg/dm^3.To achieve a higher density of soil and ash specimen a 12 ton hydraulic shop press and steel dies for powder compaction were used. In all cases, the minimal sample of incinerated material was over 180 g in weight. An estimated lower boundary of the device sensitivity amounted ca. 2.5 Bq/kg.

Results and discussion

A time span of over 45 years has passed since the moment of abundant Chernobyl fallout in the study area. The processes of the fallout migrations led to that the amounts of ^{137}Cs were unevenly distributed over soil horizons, grass and tree vegetation of the forest ecosystem. At present, the most important source of the isotope uptake should be the root absorption from soil solutions containing the ^{137}Cs ions. Thus a sort of balance may exist between the uptake by vegetation and the return of the isotope with the dead biomass to on-ground litter and further to the soil.

Analysis of spectrograms showed that along with known natural radionuclides (^{226}Ra, ^{232}Th, ^{40}K) the anthropogenic ^{137}Cs was presented in all measured specimen. Its activity concentration in the forested areas of the Natural Reserve varies widely, from the lower boundary of the device sensitivity (2.5 Bq/kg) up to 128 Bq/kg depending on relief locations and soil horizons. In the soils of the studied forested area, the ^{137}Cs activity concentration was under the value of 57.2 Bq/kg dry weight. Practically all the isotope activity is concentrated in the A soil horizon in which the stock of the isotope amounts up to 5500 Bq/m^2 (Figure2).

Fig. 2

The Figure 2 shows that ^{137}Cs was not found deeper than 15 cm in our study. Summed up in all soil horizons, a total ^{137}Cs stock varies between 2450 and 5840 Bq/m^2. The isotope stocks on the measurement points SP2 and SP31 are very similar and also very close to an average isotope stock over the whole central part of the Nature Reserve, which was found through analysis of 163 soil samples and amounted 2400 Bq/m^2. The measurement points SP9 and SP10 are different from others showing higher stocks of the isotope. According to the data by Central Siberian Administration for hydrometeorology and environmental monitoring, during the last two decades the fallout over the area were not over 6 Bq/m^2 yearly, which could not explain even 3% of the observed stock of the isotope in the studied soils.

In haplic cambisols dystric soils, the largest proportions of the isotope content are found in horizon A, amounting 95 to 99% of the total, which evidences of a long time when the isotope was delivered to the soils. Lithic leptosol soil showed a different picture, an isotope proportion in its O horizon amounted about 40% of the total content. On the background of a sufficient spatial variation of the isotope distribution in the soils the stocks range from 2110.3 to 3810.3 Bq/m^2, with the mean being 2950 ± 850 Bq/m^2.

The found values of ^{137}Cs activity concentration in soils are usually considerably higher than the minimal threshold of detection, which allows one to use the isotope as a soil erosion tracer (Mabit et al., 2013; Andrello et al., 2004). This condition will be met for at least two ^{137}Cs half-life periods or about 60 years.

Having known the times of the isotope maximal release it may be hypothesized that the main amount of ^{137}Cs was deposited over the area over the time span of 1955–1967 and the observed non-uniformity of the isotope deposition was formed during that time (UNSCEAR, 2008). The spatial non-uniformity appeared due to differences in the deposition rates between various land and plant locations. The sedimentation depends on wind speed and the properties of the underlying surface which can be bare stone surface, forested area, grassy area etc. (Il'in et al., 2010). Le Roux et al. (2008) have shown that radionuclide depositions depend on altitude and precipitation. The sample plots 9 and 10 are situated in higher slope locations where wind speed is on average higher as well, which can explain larger amounts of the isotope in their soils. Within higher elevated locations, a higher wind speed is usually observed. Low locations occur then in a wind shadow. So, over the surface of a high hill a larger volume of air would flow that contains submicron aerosols. The particles from the flow stick to tree crowns (Il'in et al., 2010). Higher locations intercept therefore more aerosols. Also, it has been shown (Le Roux, 2008) that high hills or mountains get more aerosols through wet deposition and feeder-seeder mechanism.

Takenaka et al. (1998) showed that the distribution of ^{137}Cs in forest soil could be linked to organic carbon content in the soil. The authors found that the isotope content and the soil organic carbon content correlated fairly well with each other. In our study objects, the content of organic carbon was always higher in litter horizons. Still, a high activity concentration of ^{137}Cs, expressed in Bq/kg, was observed in litter only in single instances, as e.g. on the sample plot 31 (see Fig.2). In all other cases, a high ^{137}Cs activity concentration was found in samples from AY (A1) horizons. From our standpoint, the use of isotope soil stock values rather than the isotope soil content (concentration) reflects the pattern of the ^{137}Cs distribution better. The stock parameter may be also important at studying of the isotope uptake by plant species that vary in penetration to different horizons and subhorizons. With this approach, larger stocks of soil organic carbon always corresponded to larger ^{137}Cs stocks in our study.

Besides the above mentioned isotopes, a pronounced peak of 447 keV was present in the most of the spectra. The peak belongs to a short-living natural isotope ^{7}Be. As a rule, the isotope has a cosmic origin, as a result of collisions of cosmic protons and neutrons with oxygen and nitrogen nuclei (Yoshimori, 2005). Within the study period, the activity concentration of ^{7}Be in needles of the lower tree layer was sufficiently lower than that in the litter horizon. The observation may be due to an interception of the ^{7}Be fallout by the upper layer canopy; later the isotope may come to the ground with the falling litter.

Measurements of 94 soil specimen sampled in various locations showed that activity concentration of ^{7}Be deeper than 4 cm was less than 2.5 Bq/kg in all cases. This means that the main source of ^{7}Be input is atmospheric fallout; otherwise the isotope should be at least sometimes registered in deeper horizons. From the methodological viewpoint, the non-cosmic sources of ^{7}Be may be neglected. As a possible non-cosmic ^{7}Be source, a reaction of nitrogen nuclei with alpha-particles that are produced in radioactive decay of Rn isotopes was reported (Batrakov et al., 2013).

Table 1.

This looks especially logical when the soil and not atmosphere is the main isotope source for moss and grasses. The isotope activity concentration in moss was always higher than that in grasses, which may be explained by a longer on average life of moss that gives rise to larger isotope accumulation over more cycles of vegetation. Also, biochemical particularities of the moss may play a certain role in the differences. According to Mattsson and Liden (1975), a major part of the deposited ^{137}Cs is available for transport from the dying to growing parts of the moss, which brings about a higher isotope activity in moss comparing to other plant species.

In our study, the moss ^{137}Cs activity was rather low, about 1.1 Bq/kg, which was under expectations (Table 1). A possible cause of the low ^{137}Cs content is that the isotope is mostly concentrated deeper than 3–4 cm in the soil. Mosses have no roots, solely rhizoids that are fixed only in the litter horizon.

Since the total grass biomass is higher over the sample plots the ^{137}Cs stock in grass is correspondingly higher than that of the moss. The grasses accumulate up to 0.025 Bq/m^2 while the value for mosses is under 0.016 Bq/m^2. Overall, compared to other forest compartments, the role of grass/moss layer in ^{137}Cs accumulation should be seen as rather non-substantial.

Laboratory measurements showed that the 1-year-old fir needled twigs had 64% moisture, while the older twigs had some lower moisture of 51%. The water content in Siberian fir needles is not subjected to substantial fluctuations over the vegetation period as it was observed in July and September for several years, which may be a result of the ability of the fir to regulate the water balance in some limits of water deficit. The ash content of the 1-year-old and the 2-3-year old twigs amounted 5.1% and 5.2%, respectively. The separated 1-year-old needles had 5.8% ash content and 68% moisture. The correspondent parameters for the 2-3-year-old needles are 5.7% and 58%. The needleless twigs had thus much lower ash content, around 0.8%, while having higher amounts of water in their tissues. Compared to the needles, the twig tissues were found to have sufficiently lower ^{137}Cs activity. As a result, when the needled twigs are being measured the twig tissues 'dilute' the isotope activity and lead to some underestimation of the activity in needles (Figure 3).

Fig. 3

The ^{137}Cs activity concentration in 2-3-years-old needles showed only a minor increase compared to that of 1-year-old needles. From begin of June, when young needles first appear, and by October needles take up 87-93% of all the isotope observed later in 2-3-year old needles. It is natural to suppose that the isotope uptake into the needles is the result of the soil solutions flow from tree root system due to transpiration which eventually enables the flow. The 1-year-old needles transpire more intensively than the older needles and especially when the seasonal growth takes place as it has been observed in a number of studies (Konovalov, Konovalov, 1982; Godzik, Staszewski,1994). In the course of evaporation the dissolved macro- and micro-minerals remain at the very place of transpiration.

According to data by Sen'kina (2002) Siberian fir belongs to poorly transpiring species, under 200 mg/(g·h), taking a value between those of Scots pine and Siberian spruce. The author found Siberian fir to use about 58 g of water coming up with soil solutions per 1 g of dry matter as a result of biological growth. Using the figure of water consumption and the data from figure 2 one can calculate an average ^{137}Cs activity concentration (A_{sol}) in soil solutions per volume unit as

$$A_{sol} = \frac{\sum_{j=1}^{n} \frac{A_x}{V_{H_2O}}}{n} \quad (1)$$

where Ax stands for ^{137}Cs activity concentration in the 1-year-old fir needles by the first week of October (oven-dry weight, Bq/kg), V_{H_2O} is the weight of water per 1 g of grown dry organic matter and n is the number of measurements.

Performing computations through formula (1) we used an averaged value of the isotope activity concentration in the A horizon. It seems reasonable because almost all the isotope is concentrated in the horizon. The ^{137}Cs activity concentration of the soil solutions, supposedly from the root uptake zone, differs for the sample plots. The minimal value was found in the SP2 (0.0061 Bq/L) while the maximum was recorded in the SP10 (0.0105 Bq/L).

The measured values were compared to other results given in literature. For the comparison, solid:liquid partition coefficients (Kd) (Gil-García et al., 2009) have been used. The information on Kd coefficients is widely presented in various publications. To compute Kd coefficients (L/Kg), the following formula was applied:

$$K_d = \frac{C_s}{C_l}\left(\frac{l}{kg}\right) \tag{2}$$

where Cs (Bq/kg) stands for the radionuclide activity in the soil (dry weight) and C_l (Bq/L) is the radionuclide activity in the liquid phase computed through equation (1). For example, for SP9 the mean activity of ^{137}Cs was 54.3±5.5 Bq/kg while the mean activity in needled twigs was 1.98±0.17 Bq/kg. The value Kd computed with the help of formula (2) amounted then 5480±610 L/kg. The Kd values obtained for all the soil types in our study are given in Figure 4. The values correspond closely to those reported in the literature (Jagercikova et al., 2015; Nakao et al., 2014; USEPA, 1999).

Fig. 4.

It is also known that root of trees are able to get minerals by the so-called contact exchange, i.e. through an immediate physical contact of roots with the ground (Molchanova, Karavaeva, 2001). At the same time, the relative importance of water-based exchange vs. contact exchange remains unclear. Therefore it should be remembered that the above given estimation of the ^{137}Cs activity concentration in soil solutions takes into account transpiration (i.e. water-based exchange) only.

As it has been mentioned above the atmospheric fallout of the isotope is supposed to be insignificant for the understory trees. The living and actively growing needles of the trees should act as 'concentrators' of minerals from soil solutions, ^{137}Cs included. Such a role of needles, hypothetically, makes it possible to estimate the isotope activity in soil solutions. Independent activity measurements were not performed by us, which was rather difficult due to extremely low isotope concentrations in them.

An uptake of ^{7}Be was not in the focus of the work but it is noteworthy to mention how ^{137}Cs and ^{7}Be uptakes differ depending on the source from which the isotopes are coming. July activity concentration of ^{7}Be in 1-year-old needles is substantially lower than that in older needles. For example, ^{7}Be activity concentration in 1-year-old needles amounted from 12 to 14 Bq/kg in July 2014on the sample plot 31. The older needles contained from 20 to 26 Bq/kg, that is, 40–47% more ^{7}Be than 1-year-old needles grown on the current season (Figure 5).

Fig. 5

As it has been said above, the most probable source of ^{7}Be is the atmospheric fallout that have two well expressed maxima, in spring and in autumn (Buraeva et al., 2007). Over the area of study, young needles appear from end of May to first days of June and they have small leaf area at the beginning. As a result, the young needles do not accumulate a notable amount of ^{7}Be that falls out in spring and the beginning of summer. By middle October however 1-year-old needles contain as much as 31 Bq/kg while the older needles contain 35 Bq/kg, i.e. there is only a small difference between them. It is explained by that the grown up young needles take up ever more ^{7}Be and the isotope content in older needles falls due to fast radioactive decay (half-life 53.22 days). By contrast to ^{7}Be, the uptake of ^{137}Cs goes through other sources (soil solutions) and after the phase of active growth its activity in plant tissues increases insignificantly.

Besides the soil-to-plant flow of ^{137}Cs there is a natural opposite flow. The isotope is fixed in living and dead biomass that sooner or later goes from the above-ground part of forest to the on-ground litter layer. The isotope flow can be therefore estimated; in particular, the amount of ^{137}Cs coming annually with falling litter to the ground was considered in the study.

As a rule, falling litter has a complex content, which is especially true for multispecies forests and may present sufficient technical difficulty for research. In this study, the litter was analyzed as it is, without separation into the fractions. The laboratory treatment of the collected litter showed that about 70% of it were needles of Scots pine, larch and fir, the balance consisted of larger components like cones, branch fragments and pieces of bark. In the locations of litter collection, the total oven-dry mass of litter coming to ground within four months of observation was in the range 112 to 130 g/m^2 and averaged 120-125 g/m^2. Specific ^{137}Cs activity in dry litter amounted from 1.18 to 2.17 Bq/kg.

According to studies by Ermolenko (2002), one of two annual peaks of falling off litter takes place in September, with the peak usually being three times more as the peak in May (see table S1 in Electronic Supplement). The author suggested some correction factors to estimate the total annual litter amount which were used in the study. The amounts of litter per annual basis and the activity of ^{137}Cs in it are given in Table 2.

Table 2.

It should be noted that not only falling off litter contributes to the accumulation of ^{137}Cs in soil horizons but primarily the global fallout coming on soil surface. In Figure 6, the data are given of the amounts of ^{137}Cs falling out annually in the vicinity of Krasnoyarsk. The measurements are performed by Central Siberian Administration for Hydrometeorology and Environmental Monitoring.

Fig. 6.

Comparing the data from Table 2 and fig.6 one can see that the amount of atmospheric ^{137}Cs fallout is larger than the flow of the isotope with falling off litter. It shows that not all the amount of the isotope fallout is included in the biological turnover of the studied forest area. Some factors may limit the amount of ^{137}Cs included in the turnover. First, it is an overall low concentration of the isotope, which makes the uptake into plants even weaker taking into account a certain isotope fixing capacity of soil. Second, for a longer period of the year, no vegetative activity takes place due to the low temperature, and therefore no isotope uptake occurs. However, the (global) fallout is deposited more independently of the season.

A summing up of the ^{137}Cs fallout values and amount of the isotope in the litter makes it possible to calculate the expected activities of ^{137}Cs in horizons and sub-horizons of soil. Taking into account the natural decay of ^{137}Cs the formula for calculations is as follows

$$A_n = \sum_{j=1}^{n} A_j \cdot e^{-\frac{ln2\cdot(j-1)\cdot\Delta t}{T}} + \sum_{j=1}^{n} C_j \cdot e^{-\frac{ln2\cdot(j-1)\cdot\Delta t}{T}} \tag{3}$$

where

A_n - stands for expected accumulated activity of ^{137}Cs in a horizon, Bq/m^2,

Aj - inflow of ^{137}Cs with falling litter, Bq/m^2,

n - is time span, years,

T - is the half-life of the isotope, years,

Cj - is the atmospheric fall-outs of the isotope in years j, Bq/m^2.

Calculations with the help of formula (3) show that if the upper litter soil sub-horizons on the sample plot 31, as an example, were formed over the time span 2009 to 2014 the stock of ^{137}Cs in the soil should be of the order 5.1 Bq/m^2. As it can be seen from Table 2 the isotope stock in the sub-horizons amounts more than 8.5 Bq/m^2. This means that the isotope accumulation in the sub-horizons took the time span of over 6 years, which gives an estimation of the age of the sub-horizons. It should be noted that the formula (3) is only correct when the atmospheric ^{137}Cs fallout can be considered as low and the soil solutions are the dominating source of the isotope uptake.

Such an approach may therefore provide estimations of soil horizon ages and rates of their formation. Having such estimations at hand should make it possible to reveal deviations from a natural ^{137}Cs concentration evolution due to fire events or slope erosion flows that cover soil horizons with deluvial deposits.

Compared to needles, wood of Siberian fir accumulates much less ^{137}Cs. An average measured activity of 30-years-old trees wood was 0.38±0.04 Bq/kg. This value is 11 times lower than the isotope activity in fir needles. Similar values in the range of 0.32–0.41 Bq/kg are characteristic of Scots pine wood. To calculate the isotope concentrations in all the forest stand we used previous measurements of wood stocks obtained earlier (Gavrikov et al., 2015). It follows from the calculations that a total of 11.7±3.4 Bq/m^2 ^{137}Cs is accumulated in wood of the plants, which comprises only 0.4% of the soil stocks of the isotope in the study area. Annual return of ^{137}Cs with litter amounts 0.61 Bq/m^2/year or 5.2% of the total isotope stock in the above-ground wood. By this time, it is hard to precisely estimate parameters of the isotope uptake across years because it is not known how the activity varies in annual rings of trees. It is possible however to point out a maximal value of the uptake that may reach 3.8 Bq/m^2/year, which corresponds to a ratio of the isotope concentration in needles to the isotope concentration in the soil. The value means that woody plants in the area can take up to 0.13% of the total isotope concentration in the soil.

Conclusion

A typical for Middle Siberia forested area has been studied, with the focus being done on soil-to-plant and canopy-to-soil flows of ^{137}Cs. Varying spatially, the stock of the isotope averaged 2950 ± 850 Bq/m^2. Above-ground woody mass contains 11,7±3,4 Bq/m^2 ^{137}Cs, which is only 0.4% of the soil stock of the isotope. Annual return of ^{137}Cs with litter is 0.61 Bq/m^2/year or 0.021% of the soil stock. This value is rather close to the estimations reported in the literature for dark-coniferous forests (Klyashtorin et al, 1999).

The forested area undergoes a typical succession from light-conifers to dark-conifers, a dominant representative of the latter being Siberian fir that currently presents as abundant undergrowth. Significant amounts of the isotope are accumulated in needles of the fir undergrowth. The most of the taken up isotope is accumulated in the youngest actively growing needles. The older, 2-3-year-old, needles continue to accumulate ^{137}Cs but the additional isotope amount is much smaller. The data obtained give evidence that the main process driving the accumulation of the isotope may be the transpiration-induced uptake of soil solutions from roots to needles, which provides an opportunity to estimate the ^{137}Cs activity in the soil solutions. In our view, measurements of ^{137}Cs accumulation in young needles in *Abies sibirica* relative to the isotope concentration in the soil may give a mean to measure variations of tree transpiration among different growing seasons. At least, the use of the ^{137}Cs marker may ease the getting of the information. On the other hand, no direct evidence has been as yet received of the isotope activity in the soil solutions so that estimations reported here are based on theoretical assumptions.

At a study of ^{137}Cs and ^{7}Be accumulation in needles and falling litter, it is important to take into account in what time of year the needles are sampled and what their age is because these sufficiently influence the variability of the isotope activity.

Combination of the canopy-to-ground flow with the data on fallout makes it is also possible to estimate ages of the forest soil horizons. The litter horizons in the studied area were shown to be formed in the time span of 11 and more years.

Acknowledgments

The authors are grateful to Dr. A.A. Knorre who greatly helped to collect plant material in the area of the Nature Reserve 'Stolby'. Dr. N.V. Pakharkova and Dr. O.M. Shabalina provided valuable information on botanical description of the study area. We are grateful to two anonymous reviewers whose comments helped to substantially improve our manuscript.

References

Andrello, A.C., C.R. Appoloni, and M.F. Guimaraes. 2004. Soil erosion determination in a watershed from Northern Parana (Brazil) using ^{137}Cs. Brazilian Archives of Biology and Technology 47(4):659-667.

Arinushkina, E.V. 1970. Manual on the chemical analysis of soil. Moscow: Moscow State University Press. 487 pp. (in Russian).

Batrakov, G.F., Kremenchutsky, D.A., Holoptsev, A.V. (2013). Annual variability of ^{7}Be concentration in ground air layer under tropic climate conditions. Ukrainskiigidrometeorologichniizhurnal, (13), 46-54. (in Russian).

Baskaran M, Kelley JJ, Naidu AS, Holleman DF. Environmental radiocesium in subarctic and Arctic Alaska following Chernobyl. Arctic 1991, 44: 346-350.

Baskaran M, Asbill S, Santschi P et al. Pu, Cs-137, and excess Pb-210 in the Russian Arctic sediments. Earth Planet SciLett 1996, 140: 243-257.

Bulko NI, Shabaleva MA, Kozlov AK, Tolkacheva NV, Mashkov LA (2014) The ^{137}Cs accumulation by forest-derived products in the Gomel Region. J Environ Radioact 127:150–154

Buraeva, E.A., Davydov M.G., Zorina L.V., Malyshevskii V.S., Stasov V.V. The content of cosmogenic ^{7}Be in the surface air of temperate latitudes. Atomic Energy, 102 (2007), p. 370-374. (in Russian).

Dubasov, Yu.V., et al., (1994): Chronological list of the atmospheric nuclear tests at the Semipalatinsk test site and their radiological characteristic. Bull Res Program "Semipalatinsk Test Site/Altai 4:78-86 (In Russian).

Ermakov, A.I. (1972) Methods of biochemical studies of plants. Leningrad: Kolos, 456 p. (in Russian).

Ermolenko, P.M. (2002). Seasonal variations of needle and leaf litter of trees in low-mountain belt of West Sajan. Entomologicheskieissledovaniya v Sibiri, Krasnoyarsk: KF SO REO, 2, 181–187. (in Russian).

Fesenko, S. V., Soukhova, N. V., Sanzharova, N. I., Avila, R., Spiridonov, S. I., Klein, D., ... &Badot, P. M. (2001). Identification of processes governing long-term accumulation of ^{137}Cs by forest trees following the Chernobyl accident. Radiation and environmental biophysics, 40(2), 105-113.

Fogh, C. L., &Andersson, K. G. (2001). Dynamic behaviour of ^{137} Cs contamination in trees of the Briansk region, Russia. Science of the total environment, 269(1), 105-115.

Fokina NV, Ligaeva NA, Andreeva EB, Dolzhkovaya NP (2006). Study the climatic characteristics of the reserve ‘Stolby’. Bulletin of the Krasnoyarsk State Pedagogical University. 2: 22-28

Gavrikov V.L., Sharafutdinov R.A., Knorre A.A., Pakharkova N.V., Shabalina O.M., Bezkorovaynaya I.N., Borisova, I.V., Erunova M.G., Khlebopros R.G. How much carbon can the Siberian boreal taiga store–a case study of partitioning among the above-ground and soil pools // Journal of Forestry Research, 2015, DOI 10.1007/s11676-015-0189-7.

Godzik, S., Staszewski T. (1994). The Effects of Pine (Pinussylvestris L.) Needle Surface Wax Layer Structure on Water Loss and Uptake of $14CO_2$ and $35SO_2$, in Air Pollutants and the Leaf Cuticle (eds K.E. Percy, J.N. Cape, R. Jagels and C.J. Simpson), Springer, Berlin, pp. 269-277.

Goor, F., &Thiry, Y. (2004). Processes, dynamics and modelling of radiocaesium cycling in a chronosequence of Chernobyl-contaminated Scots pine (Pinussylvestris L.) plantations. Science of the Total Environment, 325(1), 163-180.

IAEA, 2012-2015; see: http://www-ns.iaea.org/projects/modaria/

Il'in L.A., Kirillov B.F., Korenkov I.P. (2010). Radiation hygiene. Moscow: GEOTAR-Media, 384 p. (in Russian).

Kaczynski N.A. (1958). Mechanical and micro soil composition, methods of its study. Moscow, USSR Academy of Science, 193 p. (in Russian)

Klyashtorin A.L., Scheglov A.I., Tsvetnova O.B. (1999). Vertical migration of ^{137}Cs in coniferous forest ecosystems. Pochvovedenie, 12, pp. 1509–1514. (inRussian).

Konovalov V.N., Konovalov B.M. (1982). Water content and transpiration rate of differently aged spruce foliage on dried bog soils. Proceedings of All-Union conference "Problemyfiziologii I biohimiidrevesnyhrastenii", Krasnoyarsk: KF SB AS SSSR, v.2, p. 15. (in Russian).

Lehto J., Vaaramaa K., Leskinen A. (2013). ^{137}Cs, $^{239, 240}Pu$ and ^{241}Am in boreal forest soil and their transfer into wild mushrooms and berries. J Environ Radioact 116: 124-132

Mabit, L., Meusburger, K., Fulajtar, E., and Alewell, C.: The usefulness of ^{137}Cs as a tracer for soil erosion assessment: A critical reply to Parsons and Foster (2011), Earth-Sci. Rev 127:300 – 307.

Mattsson, S., Liden, K. ^{137}Cs in Carpets of the Forest Moss Pleuroziumschreberi, 1961-1973. Oikos, 3 (1975), pp. 323-327.).

Molchanova, I.V., Karavaeva, E.N. (2001). Ecological and geochemical aspects of radionuclide migration in soil-vegetation cover. Yekaterinburg: UroRAN, 161 pp. (in Russian).

Mosquera, B., Carvalho, C., Veiga, R., Mangia, L., &Anjos, R. M. (2006). ^{137}Cs distribution in tropical fruit trees after soil contamination. Environmental and experimental botany, 55(3), 273-281.

Munsell Soil Colour Charts. Baltimor. ML, 1974. 15 p.

Nimis, P. L. (1996). Radiocesium in plants of forest ecosystems. StudiaGeobotanica, 15(3-49), 3-38.

Oolbekkink, G. T. and T. W. Kuyper (1989). Radioactive caesium from Chernobyl in fungi. The Mycologist 3:3-6.

Le Roux, G., Pourcelot, L., Masson, O., Duffa, C., Vray, F., & Renaud, P. (2008). Aerosol deposition and origin in French mountains estimated with soil inventories of ^{210}Pb and artificial radionuclides. Atmospheric Environment, 42(7), 1517-1524.

Sen'kina, S.N. (2002). Moisture in plant production process. Vestnik In-ta biologii Komi nauch. Centra URO RAN, 11: 2–5. (in Russian).

Shuleikin, V.N. (2010). Radon of soil and atmospheric air and soil degassing. Georesursy. Geoenergetika. Geopolitika. M.: Institut problem neftiigasa RAN, 4-7. (in Russian).

Smagin V.N., Ilyinskaya S.A., Nazimova D.I., Novoseltseva I.F., CherednikovaYu.S. (1980). Forest types of Southern Siberia mountains. Novosibirsk: Nauka, 334 p. (in Russian).

Sukhorukov FV, Malikova IN, Gavshin VM et al. Man-made radionuclides in the environment of Western Siberia (the sources and levels of contamination) // Siberian Journal of Ecology.-2000- №1.- S. 31-38.

Soukhova, N. V., Fesenko, S. V., Klein, D., Spiridonov, S. I., Sanzharova, N. I., &Badot, P. M. (2003). ^{137}Cs distribution among annual rings of different tree species contaminated after the Chernobyl accident. Journal of environmental radioactivity, 65(1), 19-28.

Stocki T.J., Telleria D.M., Bergman L., Proehl G., Amado V., Bonchuk I., Boyer P., Chyly P., Curti A., Heling R., Kliaus V., Krajewski P., Latouche G., Lauria D.C., Mourlon C., Newsome L., Sági L., Smith J. (2011). Reference methodologies for radioactive controlled discharges an activity within the IAEA's program environmental modelling for radiation safety II (EMRAS II). Radioprotection, 46: 687–693.

Takenaka, C., Y. Onda, Y. Hamajima. 1998. Distribution of cesium-137 in Japanese forest soils: Correlation with the contents of organic carbon. The Science of the Total Environment, vol. 222: pp. 193-199.

Tamponnet C, Martin-Garin A, Gonze MA, Parekh N, Vallejo R, Sauras-Yera T, Casadesus J, Plassard C, Staunton S, Norden M, Avila R, Shaw G (2008). An overview of BORIS: Bioavailability of Radionuclides in Soils. J Environ Radioact. 99(5): 820-30.

UNSCEAR (2008). Sources of ionizing radiation. UNSCEAR Report to the General Assembly. Volume I: Report to the General Assembly, Scientific Annexes A and B, Release Date: October, 2010, Vienna: United Nations, 472 p. http://www.unscear.org/unscear/en/publications/2008_1.html

Vinichuk M, Taylor AFS, Rosén K, Johanson KJ. (2010). Accumulation of potassium, rubidium and caesium (^{133}Cs and ^{137}Cs) in various fractions of soil and fungi in a Swedish forest. Science of the Total Environment 408 (12), 2543-2548

Walling, D.E., Quine, T.A., Use of Caesium-137 as a Tracer of Erosion and Sedimentation: Handbook for the Application of the Caesium-137 Technique, Report to the UK Overseas Development Administration (1993).

Wedding, J. B., Carlson, R. W., Stukel, J. J., &Bazzaz, F. A. (1975). Aerosol deposition on plant leaves. Environmental Science & Technology, 9(2), 151-153.

World reference base for soil resources 2014. International soil classification system for naming soils and creating legends for soil maps. IUSS Working Group. World Soil Resources Reports. № 106. FAO, Rome.

Yoshimori, M. (2005). Production and behavior of beryllium 7 radionuclide in the upper atmosphere. Advances in Space Research, 36(5), 922-926.

Figure1

Click here to download Figure: Fig. 1 A map showing location of the research area..docx

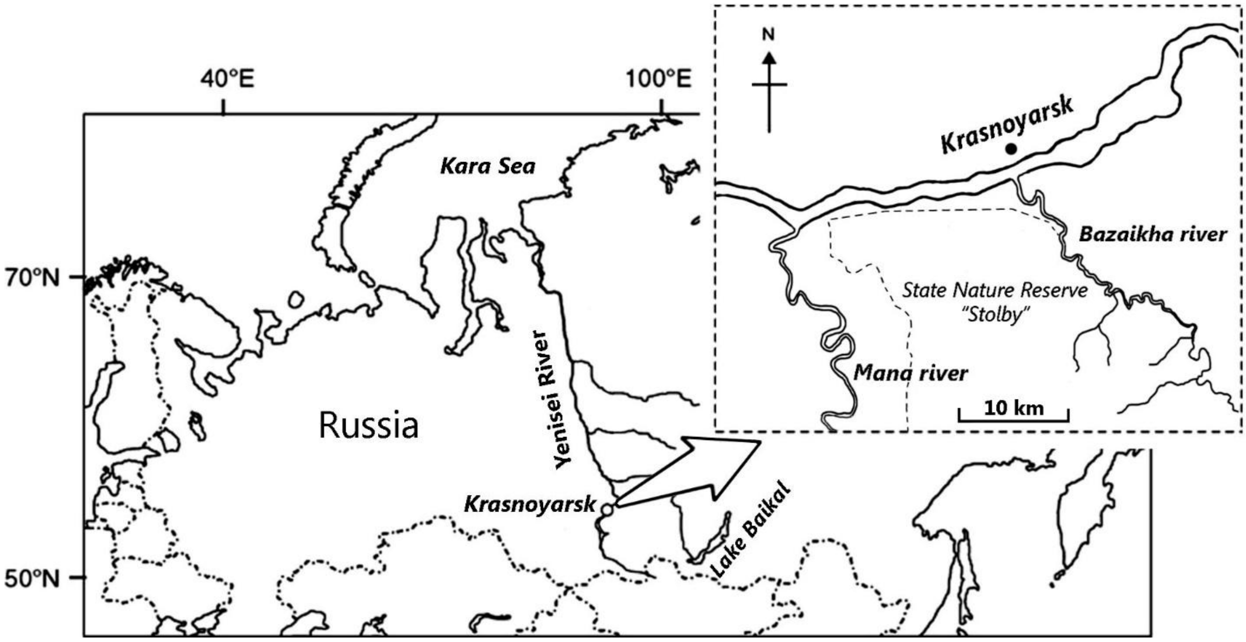

Fig. 1. A map showing location of the research area.

Figure2

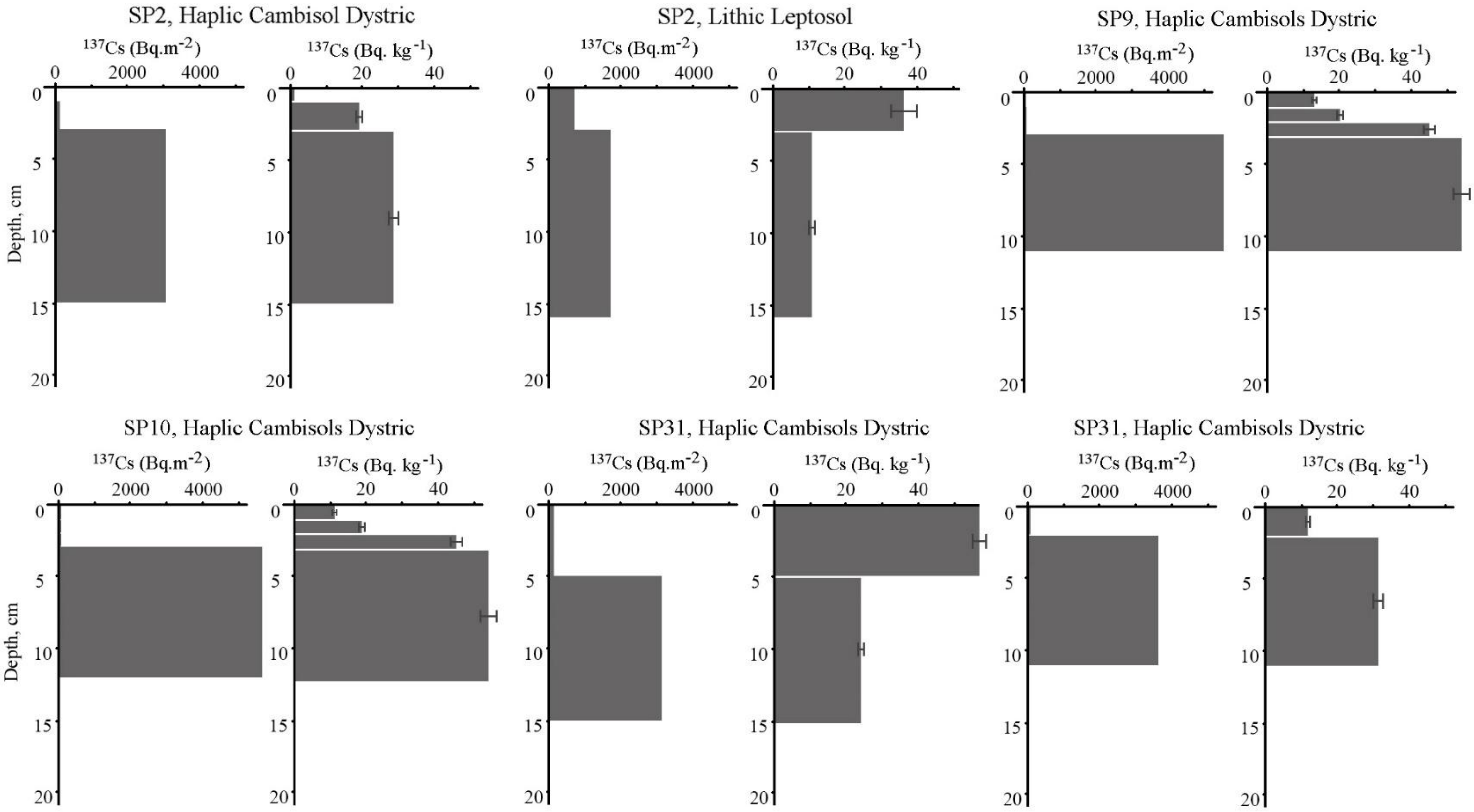

Fig. 2. Activity concentrations and stocks of ^{137}Cs in soils of the study area.

Figure3

Click here to download Figure: Fig. 3.Specific activity of 137Cs marked by Editor.docx

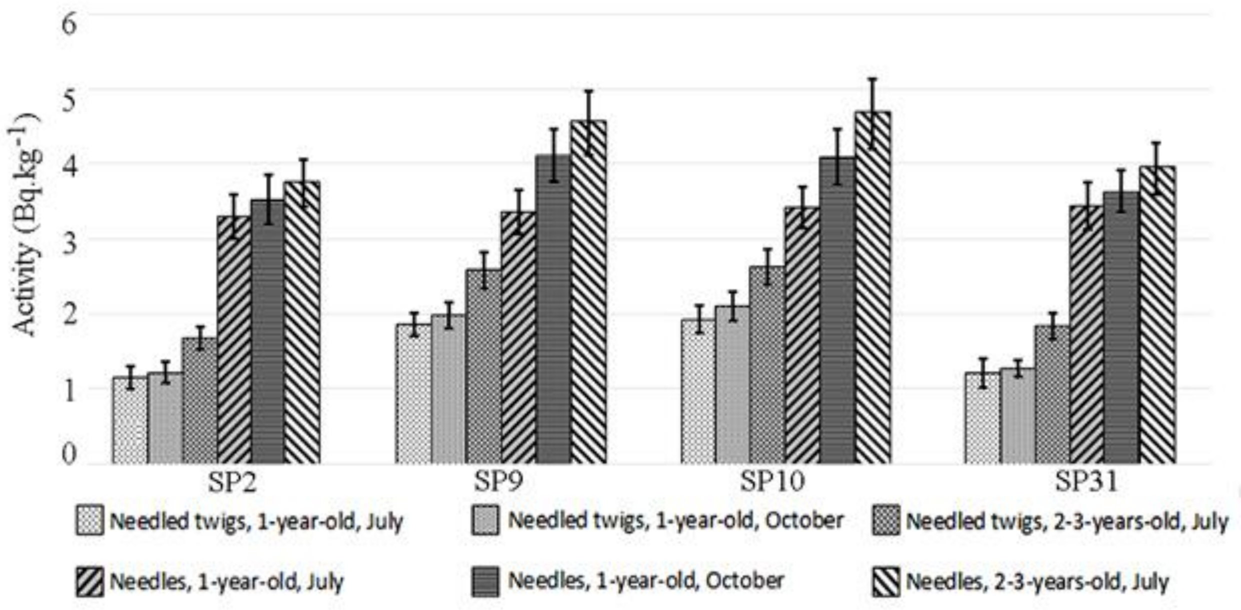

Fig. 3. Activity concentrations of ^{137}Cs in needled twigs and needles. Vertical lines represent standard errors.

Figure4

Click here to download Figure: Fig. 4.Computed values of partition coefficients (Kd) for the soils .docx

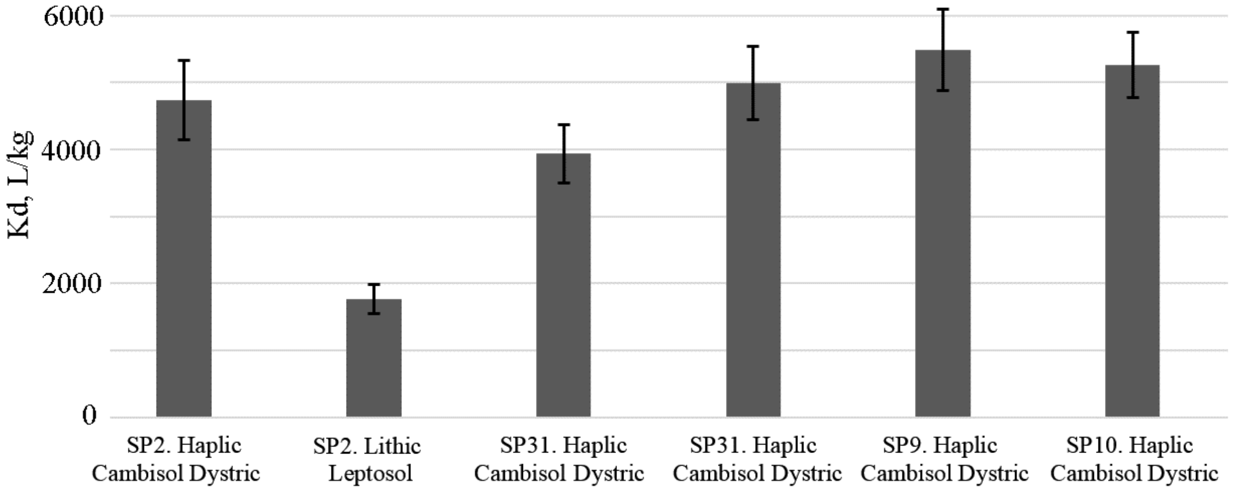

Fig. 4. Computed values of partition coefficients (Kd) for the soils of study area. Vertical lines represent standard errors.

Figure5
Click here to download Figure: Fig. 5.Specific activity of 7Be in needles marked by Editor.docx

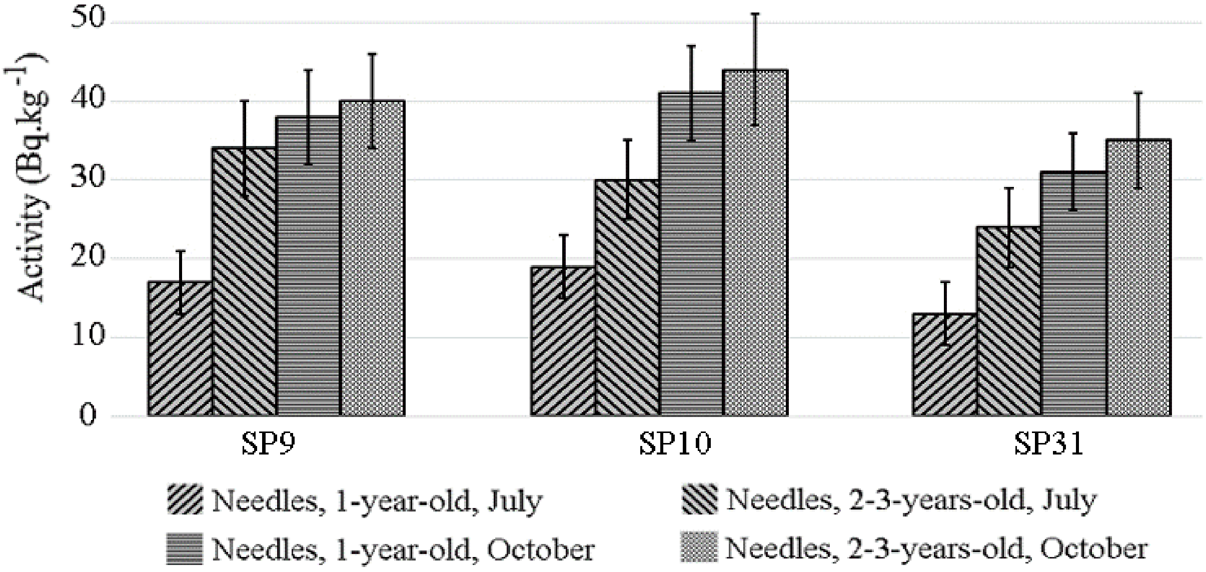

Fig. 5. Activity concentrations of ^{7}Be in needles. Vertical lines represent standard errors.

Figure6

Click here to download Figure: Fig. 6.Mean annual fallouts of 137Cs over the area of.docx

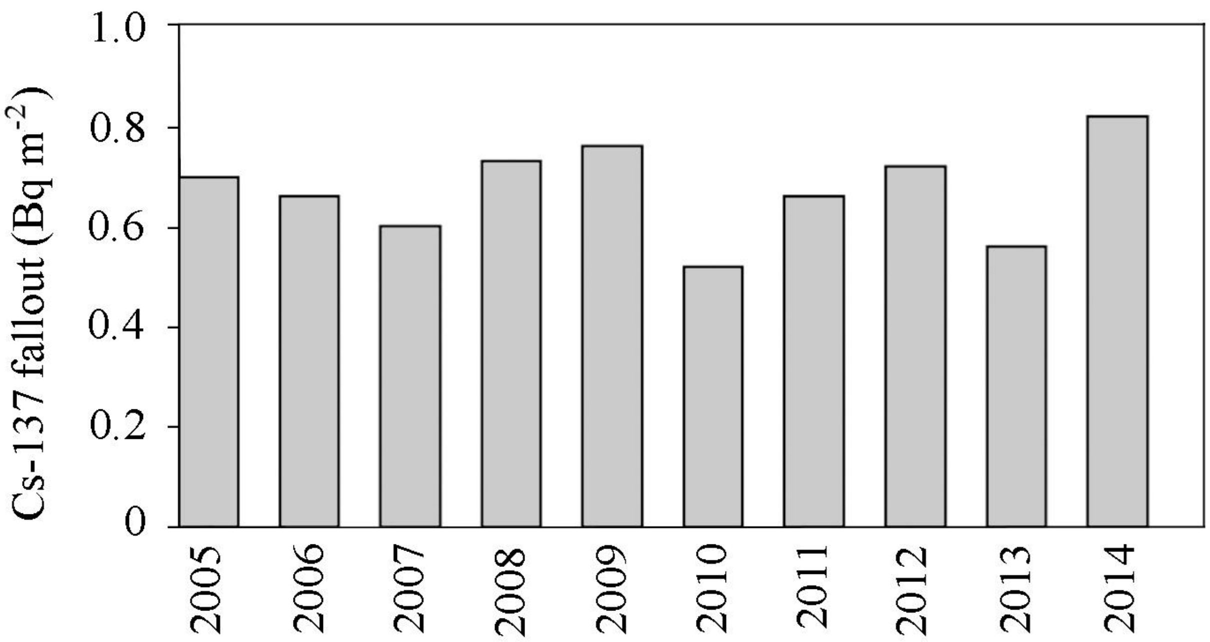

Fig. 6. Mean annual fallouts of ^{137}Cs over the area of meteorological station Experimental Field (in the vicinity of Krasnoyarsk).

Table 1. Moisture, ash content and ^{137}Cs activity concentrations for the plant fractions.

Point of measurement	Moisture, %	Ash content, %	^{137}Cs activity per weight of ash, Bq/kg	^{137}Cs activity per ODW[2], Bq/kg	MAE[3], Bq/kg	Phytomass, ODW, kg/m2	^{137}Cs stock, Bq/m2
Grass[+]							
SP2[1]	54.3	5.1	BD[4]	BD	BD	0.060	BD
SP9	52.6	9.2	5.3	0.49	0.07	0.035	0.017
SP10	55.8	8.9	5.9	0.53	0.08	0.047	0.025
SP31	51.7	6.9	BD	BD	BD	0.039	BD
Green moss[++]							
SP2	40.5	4.7	BD	BD	BD	0.0040	BD
SP31	37.8	4.6	12.4	0.57	0.097	0.0037	0.002
SP9	39.4	6.6	14.6	0.96	0.19	0.0036	0.003
SP10	42.1	6.5	16.9	1.10	0.20	0.0164	0.018

[1] SP = sample plot
[2] ODW = oven-dry weight
[3] MAE = the mean absolute error
[4] BD below detection limits
[+] mainly *Carex macroura Meinsh. subsp. kirilovii (Turcz.)*
[++] all species together

Table 2. Oven-dry weight of annual falling off litter and ^{137}Cs activity associated with it

Points of measurements	Weight of litter fallen off from July to October, g/m^2	Total annual weight of litter, kg/m^2	Flow of ^{137}Cs with litter, Bq/m^2 per year, ± SE**
Average data for 2014–2015			
SP2*	116.45	0.3456	0.428±0.054
SP9	126.4	0.3646	0.765±0.091
SP10	130.6	0.3868	0.839±0.116
SP31	112.8	0.3343	0.417±0.052

* SP = sample plot

** SE = standard error

Editor-in-Chief

Journal of Environmental Radioactivity

Re: Ms. Ref. No.: JENVRAD-D-16-00023R1

Title: Turnover of 137 Cs in 'soil-tree' system: an experience of measuring the isotope flows in a Siberian conifer forest

Dear Dr. Sheppard,

Thank you very much for your suggestion to revise our manuscript.

Please find below a table which contains all the reviewer's comments and our answers.

Sincerely,

Alexander Mitev

Reviewer's comments	Answers
Abstract: It should be absolutely clear that the new approach for estimation of 137Cs in soil water is not verified by any observations. This is a hypothesis that needs to be tested by measuring 137Cs in soil water.	Abstract was corrected
p. 3, line 5-24: I wonder whether some of this information should be in the site description instead. In any case, there should be a reference to the map as soon as the site is mentioned.	Text of the manuscript was reconfigured in this place
p.3, line 18: Unclear unit	Corrected
p.4, line 18: Not a full sentence	Corrected
p.4, line 22: "has been" should be "was"	Corrected
p. 8, line 13: Fig. 1 is a map. Do you mean Fig. 2	Corrected
p. 8, line 14-17: A problem throughout the manuscript is the inconsistent use of units. One example is line 14, where concentrations are given in Bq/m2, while kBq/m2 is used for concentrations in the same range only three lines further down	The units throughout the manuscript were consistently recalculated into Bq
p. 8, line 17: Here, for instance, reference is made to measurement points 5 and 6, but it has not been presented what type of points this is in the site description. It would be helpful with a map or a table, showing the location and the characteristics of the investigated sites.	Corrected
p. 9, line 1: Here the authors use kBq/ha, whereas kBq/m2 was used on the previous page	All units now are in Bq
p. 9, line 10: Sedimentation? Do you mean deposition?	The term is changed to 'deposition'
p. 11, line 7: Mosses have no roots. Therefore, they are sometimes used to monitor atmospheric deposition. As far as I understand, all 137Cs in	We don't mean mosses have roots. The sentence was corrected

moss should derive from atmospheric deposition, while grass can take up 137Cs from the soils with their roots.	
p. 14, lines 1-3: Since the activity of 137Cs in the soil water remains unknown, the statement has no direct support in the presented data. It should be clear that this is based only on estimations of the soil water activities so the method is not verified.	Corrections were introduced into the manuscript to clarify the issue according to reviewer's suggestions
p. 14, line 22: Is this hypothesis consistent with the assumptions used to explain the differences between needles of different age?	We believe that yes, the difference is due to differences in half-lives of 7Be and 137Cs
p. 14, line 25: Strange word order	The sentences were rephrased
p. 16, line 1: If the atmospheric deposition of 137Cs exceeds the 137Cs in litterfall, can we be sure that the 137Cs in the needles and other above-ground plant parts derive from biological uptake rather than atmospheric deposition?	We can be sure because the 137Cs activity in current year needles and the activity in older needles are comparable. If atmospheric depositions were the primary source then the activity in older needles would be multiply larger, proportional to the exposition time which for older needles is much longer than that for current year needles
p. 16, line 12: I understand what the authors are trying to do, but this equation still does not make sense to me. It is particularly the first term that bothers me. Why should you sum over the activities of previous years? Furthermore, the index j does not occur anywhere in the first term. It should also be clearly stated that no transport of 137Cs is assumed to occur between soil horizons and that 137Cs is lost only be decay.	The indices in formula were corrected, so that the formula should be now clearer for a reader
p.16, line 24: Does this refer to Eq. 3 rather than Eq. 2? What would be the problem with high fallout when this is accounted for in the formula?	Corrected
p. 18, line 8: It is quite possible that the authors are right, but there is no data on the transpiration or the soil water activities of 137Cs that provide this evidence. It is an interpretation of other data and estimated soil water activities of 137Cs.	The manuscript was corrected in this place in accordance with the reviewer's suggestions. A sentence has been added about the current absence of independent measurements in soil solutions
Figure 1: Caption missing	Corrected
Figure 2: Caption missing	Corrected
Figure 4: Since these Kd values only are not based on actual measurements of the 137Cs activities in soil water, I have not sure how relevant it is to plot them. As assumptions are made about the 137Cs concentrations in soil water, I think it would suffice just to make sure that the resulting Kd values are in reasonable agreement with observations made elsewhere	Correspondent corrections were introduced into manuscript as mentioned also above
Figure 6: Caption missing	Corrected

Supplementary Material

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