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Estimation of ^{90}Sr content in Scots pine timber based on measurement of beta particle flux density from the surface of the trunk bark

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Abstract. The measurement of the ^{90}Sr content in the components of forest ecosystems using radiochemical methods requires considerable time and effort to obtain output results. A less precise, albeit quick, assessment of this radionuclide at the stage of field work can considerably accelerate the decision-making on the possibility of involving a certain forest plot in experimental work or using it for economic purposes. Ukrainian and foreign scientific groups often devote publications to similar express methods for determining the content of biologically mobile radionuclides (^{137}Cs and ^{90}Sr): specific activity, pollution density, etc., since they allow considerably reducing the amount of field and laboratory work. Proceeding from collected field materials from 13 experimental plots of pure pine stands, close (r=0.85-0.94) statistically significant relationships at p=0.05 were found between the specific activity of ^{90}Sr in stem wood and the results of measuring the density of beta flow with a radiometer from the surface of the bark of Scots pine tree trunks at breast height (1.3 m) within a 10 km zone around the Chornobyl NPP. Direct, linear relationships were established between the average content of ^{90}Sr in the anatomical parts of tree trunks of pine stands (sap, core, and all wood) and the surface density of the flow of beta particles from the bark of trees, which were used to create regression equations suitable for preliminary assessment of the specific activity of the radioisotope under study in timber in the field conditions (R^2=0.90-0.96). A close correlation (r=0.93) was found between the average stand diameter and the concentration ratio of ^{90}Sr in sapwood to the specific activity of radionuclide in the core wood of trunks. The dependence of the ^{90}Sr content on the density of the flow of beta particles from the surface of the bark of tree trunks was established, and the ratio of the specific activity of this radionuclide between the sapwood and the core can be recommended as a method for rapid measurement of ^{90}Sr in Scots pine wood. However, given the small sample size and the importance of the issue under study, it is necessary to continue working in this area with an increase in the number of empirical data for pine stands, as well as attracting observations of other major forest-forming tree species in the exclusion zone

Keywords: forest ecosystem, radionuclide, specific activity, sapwood, core, trunk wood, Chornobyl Exclusion Zone

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Introduction

The heterogeneity of radioactive contamination lies not only in considerable spatial variability, but also in the heterogeneity of the radionuclide composition and various physical and chemical features of its redistribution in ecosystems (Khomutinin et al., 2020). At the same time, the root availability of biologically mobile radionuclides (^{137}Cs and ^{90}Sr) for the biomass of forest communities can vary by orders of magnitude, which is most often expressed as corresponding transition coefficients (Bilous et al., 2020). With this in mind, there is always a possibility of obtaining tree stand elements with considerably higher levels of radiological contamination in a "relatively clean" stand area, or vice versa. The results of such a phenomenon can be the exceeding of the levels of hygienic standards for the content of ^{137}Cs and ^{90}Sr for wood and wood products in the places of wood harvesting (Bilous et al., 2020). Another reason may be the need to considerably increase the sampling volume at the experimental plots for studying the migration of radionuclides, even within the 10-kilometre zone around the Chornobyl Nuclear Power Plant. This is because the small values of the specific activities of these radionuclides in the components of forest ecosystems often do not allow establishing indicators of their distribution with sufficient statistical significance (Khomutinin et al., 2020). Therefore, a preliminary clarifying field assessment of the radionuclide content, with the possibility of obtaining initial results within a few hours, will help start forestry or scientific work on forest areas affected by radiation pollution, without fear of obtaining contaminated forest products or unsatisfactory empirical research data.

Non-destructive methods for measuring the content of radionuclides in components of forest ecosystems are crucial for lifetime radiodosimetric studies of woody plants. With their help, the values of internal exposure of reference organisms are determined. This value usually reaches over 50% of the total exposure in the near zone around the Chornobyl NPP, mainly due to the decay of ^{90}Sr and its daughter radionuclide ^{90}Y (Beresford et al., 2020). Important criteria for the possibility of spreading such express methods are the cost of the equipment and the necessary qualification of the personnel to operate it. The development of these approaches and confirmation of their reliability are often covered in publications by Ukrainian and foreign scientists (Aramrun et al., 2018; Keisuke et al., 2018; Yoschenko et al., 2011; Yoshihara et al., 2019). Improvement and testing of methods of radiation control of environmental components is an extremely urgent task for states that have a developed infrastructure of nuclear industry and nuclear power facilities or have experienced local radioactive contamination with technogenic radionuclides. In this aspect, special attention should be paid to forest ecosystems, since components of woody biomass can deposit up to 50% of the gross activity of ^{137}Cs and ^{90}Sr localized in forest areas (Holiaka et al., 2020a).

The relevance of this study is conditioned upon the possibility of using its results for an express estimation by indirect measurement of the content of ^{90}Sr in Scots pine wood, using one of the most common radiometers-dosimeters in Ukraine *STORA-TU RKS-01* (EKOTEST, Ukraine).

The purpose of the study **is** to characterize the dependence of the specific activity of ^{90}Sr of stem wood on the flow density of beta particles from the surface of the bark of Scots pine trees at breast height (1.3 m) in the exclusion zone.

To achieve the specified purpose, the main *research tasks* were set as follows: to identify and statistically evaluate the probable correlations and dependences of the specific activity of ^{90}Sr in the stem wood and its anatomical parts on the flow density of beta particles from the surface of the bark of Scots pine trees; to characterize the impact of forest inventory indicators on the ratio of ^{90}Sr content between the sapwood and the core of the trunks; propose an algorithm for rapid measurement of ^{90}Sr concentration in timber for forestry purposes and during radiological research.

This study was the first to confirm the statistical validity of using the surface density of the flow of beta particles for the separate assessment of the ^{90}Sr content in the anatomical parts of the trunk wood (sap, core) of Scots pine trees.

The practical significance of the results lies in the possibility of estimating the ^{90}Sr content in trunk wood in field conditions in forest plots of pine stands in a brief period of time (from 2 to 3 min). This will enable a more effective radiation monitoring of the forest ecosystems of the Chornobyl Exclusion Zone.

The development of methods for detecting and measuring radiation contamination levels is closely related to the technological advance of devices and

equipment used in this area. In fact, this is a separate area of research that is currently experiencing a "boom" thanks to modern advances in information technology and automation of most processes primarily for medical purposes, namely medical radiology and radiation medicine. However, improvements in this field are quickly finding practical application in other areas of radiology (Ewert, 2016; Volterrani et al., 2019). For instance, conventional personal dosimeters are used in coniferous forests in Japan affected by the Fukushima Nuclear Power Plant accident to assess the dynamics of ^{137}Cs content in forest floor throughout the year (Yoshihara et al., 2019). Approaches to mapping radionuclide pollution using robotic complexes and unmanned aerial vehicles are being developed and modified, where the content and composition of radioisotopes are estimated based on the measurement of their beta and gamma radiation (Briechle et al., 2018; Zabulonov et al., 2015). However, the mathematical apparatus used to implement the specified "ideas" was developed as early as the 1960s and 1970s (Khomutinin et al., 2018).

In radioecological monitoring of forest ecosystems, the rapid determination of radiation pollution of forest components without sampling allows solving two principal tasks simultaneously: the time spent by personnel in areas with increased levels of ionizing radiation is reduced; the barrier function of forests in contaminated areas is more fully ensured (samples are not transported to laboratories for measurements). With this in mind, Ukrainian and foreign scientists have developed several approaches for predicting the surface density of soil contamination by technogenic radionuclides and gamma emitters (IAEA, 2003), including aerial surveys (Pradeep Kumar et al., 2020), which later became the basis for creating pollution maps by radioisotopes characterized by alpha and beta decay (Kashparov et al., 2018; Kashparov et al., 2020). The establishment of restrictions on the removal of samples in case of exceeding the regulated levels of radiation pollution outside the Chornobyl Exclusion Zone contributed to the development of methods for assessing the content of radionuclides in the biomass of woody plant communities *in situ* and *in vivo* (Ministry of Health of Ukraine & State Emergency Service of Ukraine, 2008). This was successfully used to establish the lifetime internal exposure doses of pine and birch trees within temporary radioactive waste containment sites (TRWCS) (Yoschenko et al., 2011). The constant redistribution of biologically mobile radionuclides in forest biogeocenoses requires validation and verification of the listed methods every time in case of their application, especially for ^{90}Sr, which migrates 7-10 times faster in soil and biomass elements compared to ^{137}Cs.

Materials and Methods

During 2017, at 13 experimental sites in the form of circular test plots, represented by pure pine stands, at a distance of 5-10 km from the Chornobyl NPP, the surface density of the flux of beta particles from the bark was measured, and wood samples were taken from 78 trees at breast height, according to the method of proportional-step representation. Average tax indicators of stands were in the following range: age – 27-97 years, diameter – 12-36 cm, height – 12-28 m, relative density – 0.62-1.19, stock of trunks – 129-409 $m^3 \cdot ha^{-1}$. As of January 2020, soil pollution densities (in conversion) were in the following ranges: for ^{137}Cs – 170-1700 $kBq \cdot m^{-2}$, for ^{90}Sr – 50-750 $kBq \cdot m^{-2}$. The ^{90}Sr content in the stem wood was 0.9-23.1 $kBq \cdot kg^{-1}$. Wood samples were taken using a *Haglof* increment borer (d=5.5 mm, l=500 mm) and mechanically divided into sapwood and heartwood anatomical parts.

The specific activity of ^{90}Sr in wood was measured using the radiochemical release of the radionuclide after drying the samples at 70°C, their mechanical homogenization, followed by ashing in muffle furnaces at temperatures up to 550°C (Central Research Institute of Agrochemical Service, 1985). The surface density of beta radiation particles of the trunk bark (φ_β) was determined using the *STORA-TU RKS-01* dosimeter radiometer in two stages: with the closed and open metal cover of the beta filter up to four Geiger–Müller counters, which corresponds to the values of estimates created only by gamma background (φ_γ) and gamma and beta pollution ($\varphi_{\beta+\gamma}$), respectively, after which the indicator was calculated as follows:

$$\varphi_\beta = \varphi_{\beta+\gamma} - \varphi_\gamma. \quad (1)$$

The analysis of the obtained initial field and laboratory data was performed by *Microsoft Excel 365* and *RStudio 1.2* software products (*R 3.6* programming language).

Results and Discussion

Correlation analysis revealed close relationships between the arithmetic mean values of the flux density of beta particles from the surface of the bark and the specific activity of ^{90}Sr in the trunk wood (r=0.94) of Scots pine trees at the experimental sites (critical value r=0.58 for p=0.05), as well as its anatomical parts: sapwood (r=0.85) and core (r=0.94). A strong relationship was identified between the average diameter of the stand and the ratio of the content of ^{90}Sr in the sapwood to the specific activity of this radionuclide in the core (r=0.93). The concentration of ^{90}Sr in the stem wood and its parts was characterized by significantly lower values of the correlation coefficient (r=0.02-0.49) with other inventory indicators.

Graphical interpretation of the dependences of the specific activity of ^{90}Sr ($A_{Sr\text{-}90}$) and elements of the wood of the trunk on the flux density of beta particles from the surface of the tree bark (φ_β) indicates the possibility of its description by a simple linear function with only one angular coefficient (Fig. 1). Predicted values of the angular coefficient of linear regressions were (± standard deviation) for sapwood – 23.1±8.5 Bq·cm^2·min·(kg·fr.)$^{-1}$, for heartwood – 42.3±10.8 Bq·cm^2·min·(kg·fr.)$^{-1}$, and the average value for the entire trunk wood (when combining anatomical parts) – 26.8±6.8 Bq·cm^2·min·(kg·fr.)$^{-1}$. The obtained results turned out to be comparable with the output data of a scientific study conducted 15 years ago in pine stands of 15-16 years of age within the boundaries of the TRWCS "Rudy lis" (Yoschenko et al., 2011), where the same coefficient for sapwood trunks was estimated at 26±8 Bq·cm^2·min·(kg·fr.)$^{-1}$. However, the mentioned studies were conducted at such an age of the trees, when the core part of the wood of the trunk was not yet formed, so the conditionally pointed indicator represents the entire wood of the trunk radially, which makes the compared results of the two experimental studies almost identical (Holiaka et al., 2020b). These studies, unlike the previous ones, allow using this method to estimate the content of the specific activity of ^{90}Sr in heartwood.

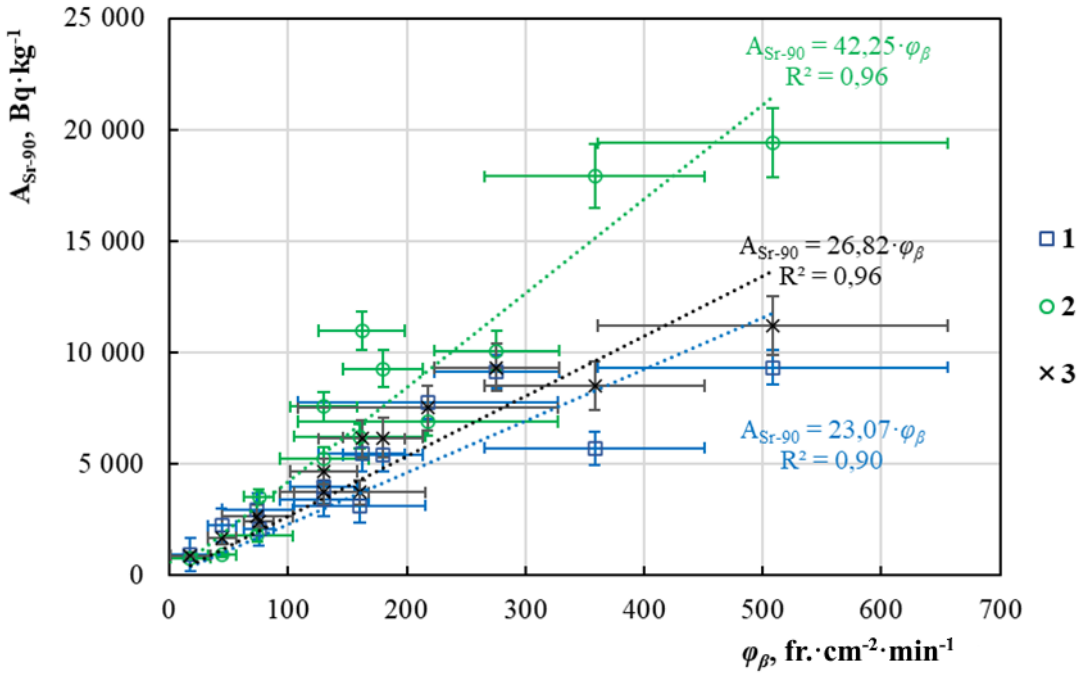

Figure 1. Dependence of the average specific activity of ^{90}Sr in the wood of the trunk (1 – sapwood, 2 – core, 3 – whole wood) on the surface density of the flux of beta particles at breast height

The increase in the ratio of the ^{90}Sr content in the sapwood to the specific activity of the radioisotope in the core ($A_{S/H}$) with an increase in the average diameter of the stand (D) in the pine plots (Fig. 2) is explained by the specific features of the deposition of this radionuclide in the stem wood. Presently, in the radial direction, ^{90}Sr accumulated most at the probable sapwood-core boundary of Scots pine trees in 1986-1992 (Holiaka et al., 2020b). From the indicated maximum of the specific activity of the radionuclide towards the bark of the trunk, the ^{90}Sr content in the wood gradually decreases several times. However, in trees older than 70 years (corresponding to an average stand diameter of more than 30 cm in this study), due to the low mobility of the radioisotope in heartwood after its formation, a site with a very low concentration of ^{90}Sr is formed in the centre of the trunk. This "dilutes" the average specific activity of the radionuclide in the core of the trunk, and accordingly, an increase in the values of the ratio of the ^{90}Sr content in sapwood to its specific activity in the core ($A_{S/H}$) is observed. This regularity contradicts the application of constructed linear regression equations (Fig. 1) and/or their predicted slope coefficients for estimating the content of ^{90}Sr in stem wood elements depending on the flux density of beta particles from the surface of the bark, since they do not reproduce the relationship between ^{90}Sr concentrations in the anatomical parts of the tree trunk, considering the average diameter of the stand. Therefore, it is necessary to apply different algorithms to estimate the total specific activity of all wood and for individual anatomical components of the trunk wood. The most appropriate way to conservatively estimate the ^{90}Sr content in the trunk wood without isolating anatomical parts is to use the linear dependence of the specific activity of the radionuclide on the flow density of beta particles from the surface of the tree bark at breast height (Fig. 1). However, if it is necessary to estimate the radionuclide content in the sapwood and the core separately (which is usually necessary during radiological research of forests), it is better to first calculate the concentration of ^{90}Sr in the sapwood using the appropriate linear regression (Fig. 1) and then calculate the probable specific activity of the radionuclide for the core based on the change in the ratio of ^{90}Sr concentrations in the sapwood to its content in the core depending on the average diameter of the stand (Fig. 2).

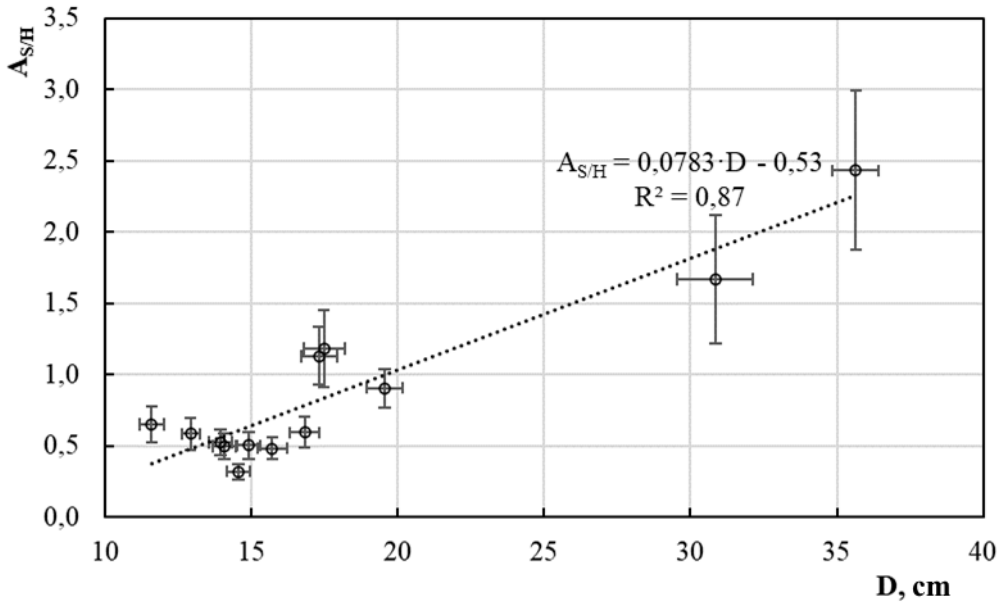

Figure 2. The dependence of the ratio of the specific activity of ^{90}Sr in the sapwood to the core on the average diameter of the stand

The obtained regularities for the express assessment of the ^{90}Sr content in the trunk wood of Scots pine stands by the developed method are not static. Since the release of the radionuclide into the environment in 1986, gradual transformation and redistribution of ^{90}Sr has been taking place in forest ecosystems, specifically in the trunks of woody plants. This requires periodic confirmation of the original quantitative indicators of this study in case of practical implementation of the method of indirect measurement of the specific activity of ^{90}Sr in stem wood based on the surface density of the flow of beta particles from the bark. It is also necessary to consider the "small" volume of observations obtained for reliable interpretation of the identified patterns to better understand the limiting factors for applying the presented approach.

Conclusions

According to the results of this study, the ability to use the beta-particle flux density indicator obtained by measurements with the *STORA-TU RKS-01* radiometer-dosimeter from the surface of the bark of the tree trunk of Scots pine trees for the preliminary assessment of the ^{90}Sr content in the trunk wood in field conditions was confirmed. The relationship between the above-listed features is characterized by a direct linear dependence, which was used to construct regression equations and find their parameters. The express method of measuring the specific activity of a radionuclide described in this paper is suitable for use when the specific activity of the trunk wood exceeds 1 kBq·kg^{-1}. Therefore, this method can be practically implemented within the 30-kilometre exclusion zone around the Chornobyl NPP. This ^{90}Sr estimation approach can be recommended for preliminary assessment of the radionuclide content during radioecological and lifetime radiodosimetry studies of pine phytocenoses. However, it is desirable to verify the method on the "own" array of observations, since the sample obtained in this and previous studies and their representativeness provide an insignificant amount of empirical data.

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Оцінювання вмісту 90Sr у деревині сосни звичайної на основі вимірювання щільності потоку бета-частинок із поверхні кори стовбура

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Анотація. Процес вимірювання вмісту ^{90}Sr у компонентах лісових екосистем із застосуванням радіохімічних методів потребує значних затрат часу та праці для отримання вихідних результатів. Менш точна, однак швидке оцінювання цього радіонукліда на етапі польових робіт може значно пришвидшити процес прийняття рішення про можливість залучення певної лісової ділянки до експериментальних робіт чи використання її у господарських цілях. Вітчизняні й іноземні наукові колективи часто присвячують публікації схожим експрес-методам визначення вмісту біологічно мобільних радіонуклідів (^{137}Cs і ^{90}Sr): питомої активності, щільності забруднення і т. д., тому що вони дають змогу значно зменшити обсяги польових і лабораторних робіт. Спираючись на зібрані польові матеріали з 13 експериментальних майданчиків, чистих за складом соснових деревостанів, виявлено тісні (r=0,85–0,94), статистично значущі за p=0,05, зв'язки між питомою активністю 90Sr у стовбурній деревині та результатами вимірювання щільності бета-потоку радіометром із поверхні кори деревних стовбурів сосни звичайної на висоті 1,3 м у межах 10 км зони навколо Чорнобильської АЕС. Встановлено прямі, лінійні залежності між усередненим вмістом ^{90}Sr у анатомічних частинах стовбурів дерев соснових деревостанів (заболоні, ядрі й усій деревині) та поверхневою щільністю потоку бета-частинок з кори дерев, що використані для створення регресійних рівнянь, які придатні для попереднього оцінювання питомої активності досліджуваного радіоізотопу в деревині у польових умовах (R^2=0,90–0,96). Ідентифіковано тісну кореляцію (r=0,93) між середнім діаметром деревостану та відношенням концентрацій ^{90}Sr у заболоні до питомої активності радіонукліда в ядровій деревині стовбурів. Встановлено залежності вмісту ^{90}Sr від щільності потоку бета-частинок із поверхні кори стовбурів дерев та співвідношення питомої активності цього радіонукліда між заболонню і ядром цілком можна рекомендувати як спосіб для експрес-вимірювання ^{90}Sr у деревині сосни звичайної. Однак, враховуючи незначний обсяг вибірки та важливість розглянутого питання, необхідно продовжити роботи у вказаному напрямі зі збільшенням кількості емпіричних даних для соснових деревостанів, а також залученням спостережень за іншими основними лісотвірними деревними видами зони відчуження

Ключові слова: лісова екосистема, радіонуклід, питома активність, заболонь, ядро, стовбурна деревина, Чорнобильська зона відчуження