

# RADIONUCLIDE ACCUMULATION BY OBJECTS OF ECOSYSTEM

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The isotope contents of family uranium and thorium in the ash of wood of trees and pine needles before and after leaching by water were measured. The element contents in the ash of wood of trees and pine needles were measured by  $\gamma$ -activation analysis and spectrophotometer. A significant excess of lead-210 in pine needles in the vicinity of thermal power plant relatively its average content in Ukraine was discovered. Differences of magnitude of leaching uranium series isotopes, <sup>137</sup>Cs and <sup>40</sup>K from the wood of trees and pine needles were defined, which is due to the different character of the formation of crystallites during ashing of these objects

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## 1. INTRODUCTION

The problem of radioactive environmental contamination is an important problem for an effective utilization of power resources of Ukraine. Many aspects of behavior of radioisotopes in biosphere and forecasting of their carrying over in environment are not solved till now. Therefore the intensive researches of behaviour of radioisotopes in biosphere is continuing. These are such radioisotopes, like <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>99</sup>Tc, <sup>226</sup>Ra, <sup>210</sup>Po, <sup>36</sup>Cl, <sup>129</sup>I, etc.

The radiation influence in each region has features. The adequate estimation of accumulation and migration of long-living radioisotopes will allow solving complex problems which are connected with a source of transfer of radioisotopes in biological environments.

Many problems of radioecology are already solved now. These results are received in field and laboratory conditions, and also by means of models. In our opinion the big perspective in a direction of a solution of a problem of radionuclide distribution in various ecosystems may be realized with using accelerating techniques and modern nuclear technologies. Using of  $\gamma$ -activation analysis for quantitative estimation of radioisotopes can be one of such directions.

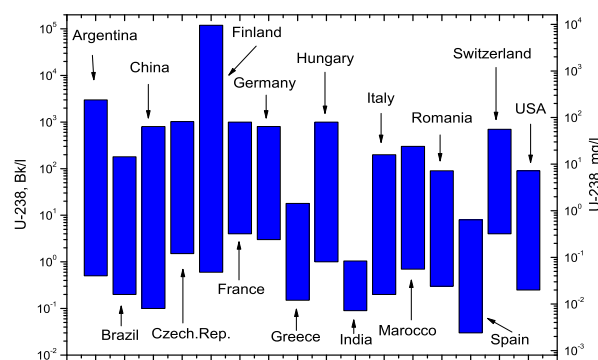
The radioactive waste which produced by the nuclear industry and atomic power stations contains radioisotopes which represent considerable danger to the population. Work of thermal power stations causes considerable release in environment of isotopes of family of uranium-235, 238 and thorium-232 too.

Food consumption is the main source of human exposure to radioisotopes which leads to dangerous

influence of radiation on the person [1]. Especially it concerns action of alpha-ray isotopes which are incorporated in a human body (Table 1). A significant amount of long-living alpha-ray isotopes can be incorporated in human body by means of water (Fig.1) [2].

**Table 1.** The influence of different radiation (1 mGy = 6.24 keV/ng). Cell doses are multiples of microdoses

	mGy	ROS/hit/ng
<sup>60</sup> Co $\gamma$ -rays	~0.3	~45
<sup>137</sup> Cs $\gamma$ -rays	~0.4	~60
250 kVp x-rays	~0.9	~135
100 kVp x-rays	~1.0	~150
10 MeV protons	~6.0	~900
4 MeV $\alpha$ -particles	~350.0	~52.5·10 <sup>3</sup>



**Fig.1.** The uranium content in potable water

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The purpose of the given article consisted in the analysis of radionuclide accumulation by wood and aboveground phytomass of pine forests and studying of leaching of isotopes of uranium family,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  from ashes of these objects.

## 2. MATERIALS AND METHODS

As an initial material the ashes of fruit-trees, oak ashes and ashes of pine needles from various areas of the Kharkov region have been used. According to the available literary data, pine needles differ from other kinds of plants by high radio sensitivity [3].

The content of radioisotopes in ashes of initial objects is detected by means of Ge(Li)-detector with the energy resolution 3.25 keV. Activation of samples have been conducted with use brake radiation of the linear accelerator electrons NSC KIPT with electron energy 23 MeV and a current 500  $\mu\text{A}$ . Activation of samples was carried out on air, the temperature of samples in the course of activation did not exceed 40°C. Comparative research of the given samples in their initial and gamma activated conditions is conducted.

The determination of radionuclide content in ashes of fruit-trees, oak ashes and ashes of pine needles was carried out also by means of measurement their optical density in spectral area of regional absorption, and also in the field of a transparency of leachate and dithizone complexes. The dithizone (diphenylthiocarbazone) - one of most part of used organic photometric reagents. The dithizone has been used for extraction of micro quantities of elements. Solutions of dithizone metal in organic solvents are usually intensively painted. The steadiest dithizone complex allows realizing extraction at various pH (high sour, sub acidic, neutral or alkaline) depending on tasks in view. Formation of dithizone metals has various speed extractions. Therefore we carried out intensive hashing by means of a magnetic mixer with the subsequent measurement of absorption of light by dithizone of metal. The concentration of solutions of dithizone in  $\text{CCl}_4$  and  $\text{CHCl}_3$  0.001...0.002% have been used for spectrophotometer.

In the course of carrying out leaching the pH of leachate was measured on pH-meter-340. pH of solution of leaching was from 7 to 12.

## 3. RESULTS AND DISCUSSION

In Fig.2 the  $\gamma$ -spectrum of the initial sample of ashes of fruit trees is presented. In  $\gamma$ -spectrum of investigated sample the various elements are observed ( $^{226}\text{Ra}$ ,  $^7\text{Be}$ ,  $^{214}\text{Pb}$ ,  $^{208}\text{Tl}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$  and others). Special interest causes  $^7\text{Be}$  ( $T_{1/2}=53$  days).  $^7\text{Be}$  is generated by cosmic rays in the upper troposphere and stratosphere, and is a tracer of downward aerosol transport.

The  $\gamma$ -spectra of initial samples of oak ashes (Fig.2), phytomass of pine forest (Fig.3) and  $\gamma$ -spectrum of ashes of fruit trees are similar. Apparently, accumulation of radioisotopes in studied objects from various areas of the Kharkov region was

totally and uniformly. Specific activity of these elements do not exceed maximum concentration limit. The high content of  $^{137}\text{Cs}$  in ashes of fruit trees and an oak, apparently, is caused its moving by roots from soil and redistribution between bodies and tree tissues. Trunk wood, as a rule, is considered as the most conservative component of radioisotopes. It is connected with that wood has a small part of physiologically active fabrics participating in moving of radioisotopes [4, 5].

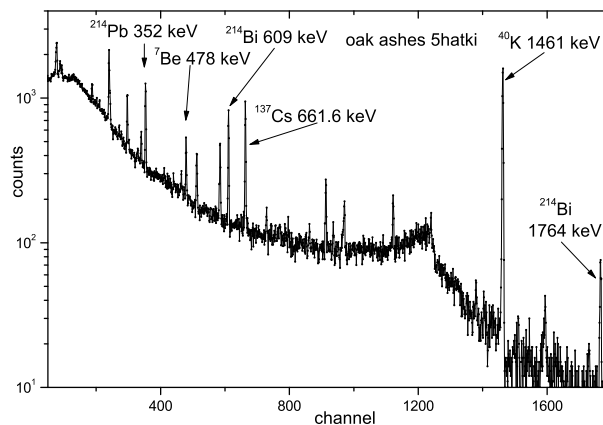


Fig.2. Energy spectrum of ashes of oak

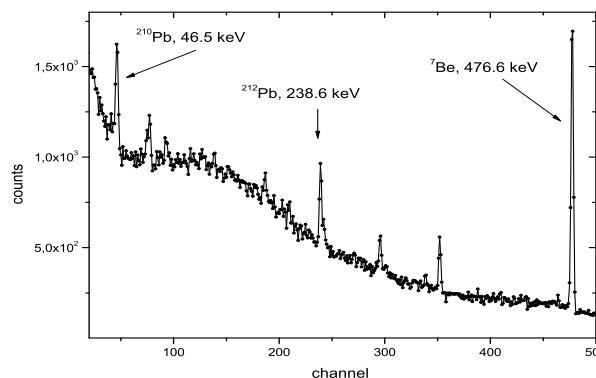


Fig.3. Energy spectrum of ashes of pine needles

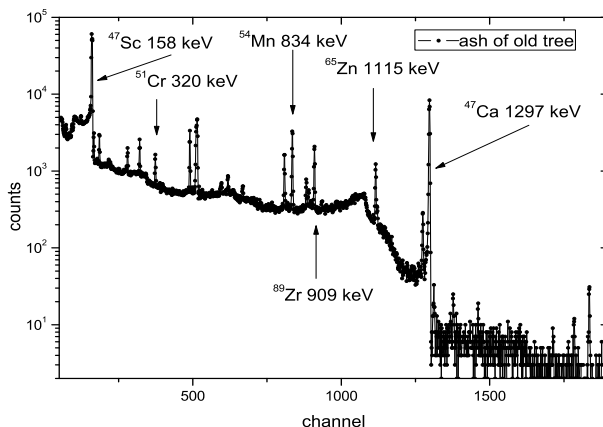


Fig.4. Energy spectrum of ashes of old tree after an irradiation on the electronic accelerator

The content of  $^{210}\text{Pb}$  in plants fluctuates from 100 to 1000 Bk/kg. The estimation of content of  $^{210}\text{Pb}$  in

aboveground phytomass of pine forest in area v. Gineevka is about 8000 Bk/kg (dry weight) (see Fig.3). Therefore the population of nearby areas of thermal station can be exposed to essential radiating influence. Especially, it concerns teenagers of pubertal period in the course of formation the bone system.

**Table 2.** The activity of ashes wood trees and pine needles after irradiation by bremsstrahlung, Bk/g

	ash old tree	ash young tree	ash pine needles	ash oak 5hatki
<sup>22</sup> Na	19.9	12.2	7.4	57.2
<sup>43</sup> K	3962	4377	318.8	2284
<sup>47</sup> Ca	2795	3105	236	1662
<sup>46</sup> Sc	10.8	5.4	6.2	21.9
<sup>48</sup> Sc	45.6	21	21.7	78
<sup>51</sup> Cr	21.8	4.9	1.9	4.36
<sup>54</sup> Mn	126.7	49.2	185.7	306.6
<sup>58</sup> Co	3.5	1.1	1.57	4.25
<sup>57</sup> Ni	52.7	61	18.6	64.3
<sup>67</sup> Cu	153.6	24.9	6.7	7.6
<sup>65</sup> Zn	116.8	20.9	5.5	5.5
<sup>74</sup> As	7.65	3.55	6.0	13.8
<sup>77</sup> Br		19.7	16.6	
<sup>84</sup> Rb	32.2	36.8	16.1	96
<sup>88</sup> Y	4.38	2.1	2.36	10.0
<sup>95</sup> Zr	2.55	1.8	2.71	5.46
<sup>89</sup> Zr	404.7	184.8	374.8	976.8
<sup>92m</sup> Nb	8.2	4.0	6.3	16.2
<sup>99</sup> Mo			0.95	
<sup>126</sup> I	8.5	4.3	10.4	28.5
<sup>135m</sup> Ba	296	320	20.5	195.5
<sup>141</sup> Ce	3.8	2.7	1.9	8.15
<sup>139</sup> Ce	10.3	5.9	5.8	24.4
<sup>203</sup> Pb	4.81	2.87	4.2	11.6
<sup>237</sup> U	4.81	2.87	4.2	11.6

The greatest accumulation of <sup>137</sup>Cs is noted in phytomass of pine forest which is indicator of entering by means of root of mineral elements and radioisotopes. The <sup>137</sup>Cs content in phytomass of pine forest is comparably with estimations of this indicator of other researchers [6]. The stream of <sup>137</sup>Cs in aboveground phytomass of pine needles for one vegetative period is 0.7...1.8% concerning its general the content

**Table 4.** The leaching of isotopes from ashes of old tree, oak and pine needles by means of water

samples	<sup>228</sup> Ac	<sup>212</sup> Pb	<sup>208</sup> Tl	<sup>226</sup> Ra	<sup>214</sup> Pb	<sup>214</sup> Bi	<sup>7</sup> Be	<sup>40</sup> K	<sup>137</sup> Cs
ashes of old tree, Bk/kg	0.76	0.6	0.64	1.67	0.92	3.5		240	1.44
leaching, %	12%	7.54%	6.7%	5.9%	7.6%	30.3%		47.1%	4.8%
ashes of oak 5hatki, Bk/kg	3.72	0.34	3.2		3.27	1.7	0.1	255	0.38
leaching, %	28.8%	2.3%	23%	0.1%	19%	9.1%		34.97%	2.6%
ashes pine needles, Bk/kg	3.49	1.63	2.1	6.6	1.55	2.94	65.5	36.2	5.25
leaching, %	68%	50.6%	70%	56.7%	36.7%	70%	36.7%	49.3%	24.9%

in an ecosystem. As a result of entering of aboveground phytomass of pine forest in soil the effect of returning <sup>137</sup>Cs in a plant is observed. This indicator is 0.1...0.5% <sup>137</sup>Cs from total quantity of coniferous landing.

In Fig.4 is presented  $\gamma$ -spectrum of ashes of the old trees activated by bremsstrahlung on electron accelerator. In  $\gamma$ -spectrum are present radioisotopes <sup>89</sup>Zr, <sup>51</sup>Cr, <sup>54</sup>Mn, <sup>47</sup>Sc and other elements which were not observed in an initial spectrum of ashes of these plants.

**Table 3.** The element content of wood ash [7]

elem.	ash oak, mg/kg	elem.	ash wood, mg/kg
Ca	314	Hg	<0.4
Fe	0.9	Cd	0.4-0.7
K	102.5	Co	0-7
Mg	75.7	As	0.3-3
Mn	1.4	Cr	>60
P	5.6	Ni	40-250
S	12.1	Pb	15-60
Si	1.3	Cu	15-300
		V	10-120
		Zn	15-103

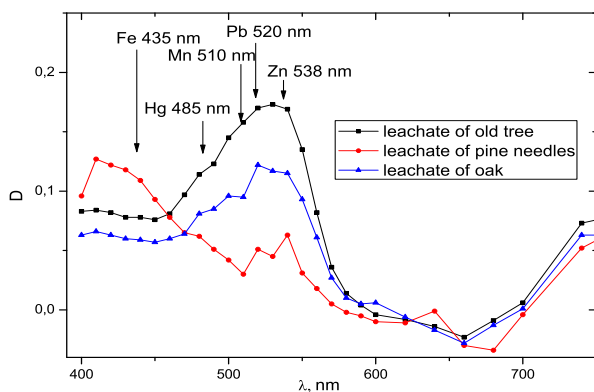
The content of radioisotopes in ashes of old and young trees and also in ashes of oak and phytomass of pine forest is presented in Tables 2,3. The analysis of the obtained results has shown that the more content of radioisotopes is in ashes of old trees that confirms the dynamic of accumulation by plants radioisotopes. It is important for forecasting of pollution of radioisotopes. In this case it is possible to carry out calculation of accumulation and distribution radioisotopes in wood plants. Content of radioisotopes in oak ashes considerably more, than in ashes of fruit trees. Higher moisture capacity of soils in forest, than in gardens can be one of possible factors of more considerable transition and accumulation radioisotopes in oak ashes.

Also it is necessary to notice that the calcium and potassium content in aboveground phytomass of pine forest in 15 times is less, than in wood of old trees. It causes formation considerably smaller quantity of steady compounds in ashes with content of calcium, potassium, silicon and phosphorus.

Let's notice that products of decay of  $^{235,238}\text{U}$  and  $^{232}\text{Th}$  in ashes of pine needles, as a rule, are more than in old trees and in oak (Table 4). This is due to the initial isotope content in pine needles, as well as by aerosol deposition of radionuclides from the thermal power working nearby. The content of  $^{40}\text{K}$  in ashes of pine needles is much less, than in ashes of old trees and an oak. It has caused a neutral of solution leachate for ashes of pine needles (pH=7). pH of leachates of ashes of old trees and an oak were 12. In process of ashing of pine needles (Table 5) it is formed less than insoluble compounds of type of  $\text{KAlSi}_3\text{O}_8$ ,  $\text{SiO}_2$ ,  $\text{Fe}_2\text{O}_3$  and others which are formed at ashing oak and old trees [8, 9]. In process of ashing oak and old trees also are formed nanometer sizes crystallites.

**Table 5.** The element content of pine needles [10]

element	C	H	N	S	O	Cl	ash
wt. %	50.4	6.5	0.8	0.01	37.9	0.22	4.5



**Fig. 5.** Transmission UV-VIS spectra of ash leachate of old tree, pine needles and oak

Study of mechanisms of release of radioisotopes from plants as a result of contact to water solutions is extremely important. UV-visible absorption of leachate by various complexes of dithizone in their dynamics leaching in a range of lengths of waves of 200...800 nanometers are shown in Fig.5. Apparently from Fig.5, the resulted spectra show almost identical profiles in area 400...700 nanometer, except for ashes of pine needles. All spectra are characterized by presence of an intensive strip in area 550 nanometers that corresponds to presence such elements, as Mn, Zn and Pb. Let's notice that for pH=7 the speed of leaching of Mn, Zn and Pb is minimum [11]. Therefore for aboveground phytomass of pine forest value of leaching speed (pH=7) is smaller than of leaching speed for wood of old trees and oak (pH=12).

#### 4. CONCLUSIONS

1. The isotope contents from family of  $^{235,238}\text{U}$  and  $^{232}\text{Th}$  in aboveground phytomass of pine forest, as a rule, are more than in wood of old trees. It is due to deposition of radioisotopes by means of aerosols from a thermal power station working nearby.

2. It is established significant excess of lead-210 in pine needles in the vicinity of thermal power plant relatively its average value in Ukraine.

3. Differences of magnitude of leaching uranium series isotopes,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  from the wood of trees

and pine needles were defined, which is due to the different character of the formation of crystallites during ashing of these objects.

4. The content of calcium and potassium in aboveground phytomass of pine forest in 15 times is less, than in wood of old trees. It causes formation considerably smaller quantity of steady compounds of ashes in which enter calcium, potassium, silicon and phosphorus.

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## НАКОПЛЕНИЕ РАДИОНУКЛИДОВ ОБЪЕКТАМИ ЭКОСИСТЕМЫ

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Проведены измерения содержания изотопов ряда урана и тория в золе деревьев и хвои сосны до и после выщелачивания водой. Измерено содержание элементов в золе деревьев и хвое сосны при помощи  $\gamma$ -активационного анализа и спектрофотометрии. Обнаружено значительное превышение содержания свинца-210 в хвое сосны в районе тепловой электростанции относительно его среднего содержания в Украине. Обнаружены различия величины выщелачивания изотопов ряда урана,  $^{137}\text{Cs}$  и  $^{40}\text{K}$  из деревьев и хвои сосны, которые обусловлены разным характером формирования кристаллитов при озолении в этих объектах.

## НАКОПИЧЕННЯ РАДІОНУКЛІДІВ ОБ'ЄКТАМИ ЕКОСИСТЕМИ

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Проведено вимірювання вмісту ізотопів ряду урану і торію в золі дерев і хвої сосни до і після вилугування водою. Виміряно вміст елементів в золі дерев і хвої сосни за допомогою  $\gamma$ -активаційного аналізу та спектрофотометрії. Виявлено значне перевищення вмісту свинцю-210 в хвої сосни в районі теплової електростанції щодо його середнього вмісту в Україні. Виявлені відмінності величини вилугування ізотопів ряду урану,  $^{137}\text{Cs}$  і  $^{40}\text{K}$  з дерев і хвої сосни, які обумовлені різним характером формування кристалітів при озоленні в цих об'єктах.