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RADIOCARBON IN ELEMENTS OF THE LANDSCAPE (BELARUS)

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Abstract: It was found that in areas distant from an operating NPP the ^{14}C concentration is characterized by values of 105-108 pMC. In the area of the Ignalina plant rather high ^{14}C contents of vegetation (up to 150 pMC), and water of the lake Drisvyaty (127-154 pMC) connected by canals with the Ignalina NPP were noted. Biocarbonates (shells of lake mollusks *Anadonta cygnaea* and *Dreissena,* eggs poultry fed) are also enriched in radiocarbon.
The concentrations of ^{14}C in plants sampled in 1994-1996 in Gomel region (Chernobyl zone) are similar to those from the Ignalina NPP region and show increased values (up to 130-140 pMC). On the other hand, an emission of the radiocarbon resulted from the Chernobyl accident was recorded in the biosphere, at its concentrations increased toward the plant. So, ^{14}C values were about 200 pMC near Minsk (May-June, 1986) and reached 600-700 pMC near Gomel. These data permit a realistic estimate of the concentrations of the radiocarbon emission at the instant of the accident. The radioisotope is well preserved and its redistribution occurs mainly due to biochemical processes. This is well shown by the radiocarbon distributions in a profile of the soil boggy medium.

1. INTRODUCTION

Nuclear power plants (NPP) operating under even standard conditions release annually about 92.5 10^{10} Bq of radiocarbon (^{14}C) to the environment for generating 1GW of electrical energy (Sobotovich, 1992). This isotope is formed in the active zone of nuclear power reactors of any types, where neutron stream is coming in contact with reactor construction materials, substance of heat carrier, moderator, fuel and admixtures (Kovalyukh *et al.*, 1993; 1996; 1998).

A contribution of all these sources of radiocarbon may vary considerably depending on the reactor types and structural features.

Water-boiler nuclear reactors with graphite moderators (Chernobyl, Ignalina, Smolensk NPPs) have no analogues abroad. Their major features are abundant nitrogen in the active zone, which mixed with helium is used to cool moderator and a great amount of carbon in the composition of moderator itself. This causes a high rate of ^{14}C generation, which is almost an order of magnitude greater than that obtained in water-cooled and water-moderated reactors used in European countries and the USA (Rublevsky *et al.*, 1979). Radiocarbon generated in the heat carrier and moderator is released partly or completely into the environment as gas aerosols and as a component of radioactive wastes – from reactor fuels. Its biological action is associated with not only radiation, but also transmutation effect that arises when ^{14}C atoms decay into ^{14}N atoms. These processes may be especially dangerous when radiocarbon is incorporated into desoxyribose nucleic and ribonucleic acid of gametal cells, as even single decays may stimulate unavoidable mutation in the organism (Vasilenko *et al.*, 1992).

Accidents in nuclear power plants cause emission of large amount of ^{14}C, which is an integral part of the general radioisotope contamination. Data of many investigations (Kovalyukh *et al.*, 1993; Kovalyukh *et al.*, 1996; Buzinny *et al.*, 1998; Kovalyukh *et al.*, 1998) report that the Chernobyl NPP accident caused a discharge of radiocarbon as fine-grained reactor graphite and carbon dioxide – a graphite combustion product. Calculations by

G. Gofman (1994) show that about 162,000 Ci of radiocarbon was released as a result of the Chernobyl NPP accident. This information was used to suggest that Chernobyl-born radiocarbon may appear 1.26 times more detrimental to heredity of man than radiocaesium, and its harmful effect may manifest itself for much longer period of time.

Investigation of the radiocarbon distribution in natural objects is a permanent constituent part of environmental monitoring in most of European countries over a long period of time (Levin *et al.*, 1985). The first consideration is given to regions adjacent to nuclear power plants or sites of land-filled radioactive wastes. In Hungary, Slovenia, Slovakia ^{14}C monitoring involves determination of the radiocarbon concentrations in air and vegetation. It was found that several times increased radiocarbon concentration in the mentioned landscape elements in the neighbourhood of operating nuclear power plants may be due to abundant nitrogen admixture in the primary cooler (Uchrin *et al.*, 1998). Weekly ^{14}C monitoring in the air nearby a nuclear power plant in Slovenia indicated the radiocarbon concentration averaging 15 Bq/m^3 in the near-surface air; however, some accidental releases as high as 1.900 Bq/m^3 in activity were noted (Vokal *et al.*, 1996). Rather high ^{14}C concentrations were determined in rings of trees and air samples around a Swedish nuclear power plant (Stenstrom *et al.*, 1998). It is interesting to note that Swedish (Olson, 1989) and Finnish (Salonen, 1987) scientists were first no detect the radiocarbon concentration increase to more than 300 pMC in the air in northern Sweden in June-July 1986 after the Chernobyl NPP accident.

Calculation of the total amount of ^{14}C released to the atmosphere of Great Britain as a result of operation of nuclear power plants before 1994 gave a value about 448 kBq (McNamara *et al.*, 1998). An impact of nuclear power generation on the radiocarbon concentration variations in the atmosphere, groundwater and vegetation was assessed in the vicinity of the operating Ignalina NPP in Lithuania (Banis, 1988; Mazheika and Pyatroshus, 1992). Investigations have shown that since the NPP putting in operation the ^{14}C content of the Drisvyaty (Drukshai) Lake water varied between 115 and 150 pMC; some ^{14}C concentration increases were also noted in vegetation and near-surface air.

2. METHODS AND OBJECTS OF INVESTIGATION

Though Belarus, has no it's own nuclear energetic, three nuclear power plants (NPP): the Chernobyl, Ignalina and Smolensk ones equipped with water-boiler reactors with graphite moderators are located at its frontiers.

In 1994 the Institute of Geological Sciences of the National Academy of Sciences of Belarus was engaged in works for the radiocarbon concentration determination in vegetation, near-surface air, biocarbonates (eggshell of poultry and wildfowl, mollusk shells), surface and groundwater and studies of their distribution pattern in the territory of Belarus. Investigation were carried out in several test ground (**Fig. 1**) and intended:

- to determine the background radiocarbon concentration in regions presumably not subject to the NPP impact;
- to assess the ^{14}C concentration in the territories adjacent to nuclear power plants under regular operation conditions;
- to assess the radiocarbon amount released into the atmosphere as a result of the Chernobyl NPP accident.

Fig. 1. Map showing sites of sample collection for radiocarbon analysis.
1– Volozhin district; 2 – Braslav district, Drisvyaty lake; 3 – Braslav district, Muysata Lake; 4 – Lelchitsi district, village of Svidnoye; 5 – Lelchitsi district, village of Zamoshie; 6 – Belovezhskaya Pushcha; 7 – Chechersk district; 8 – Gorky district, village of Palenka; 9- Gorky district, village of Ruditsa; 10 – Mogilev region, village of Dubrovno; 11– Mogilev region, village of Kalinovka; 12 – Mogilev region, village of Ovsjanka; 13 – Shklov district; 14 – Dubrovno district; 15 – town of Khoiniki; 16 – Khoiniki district, village of Masani; 17 – Bragin; 18 – Narovlia district; 19 – Cherikov district; 20 – Vetka district; 21– town of Gomel; 22 – Myadel district, Naroch lake; 23 – Dokshitsy district, Medzozol lake; 24 – city of Minsk

Particular attention was given to the study of the north-western part of Belarus found in the immediate vicinity of the Ignalina NPP. The plant is located on the western side of the Drisvyaty Lake and equipped with the "Chernobyl type" reactors (**Fig. 2**). The territory around the nuclear power plant is generally characterized by a rather technogenic load on the landscape (Novikov *et al.*, 1994). Two canals were dug to connect the plant and the lake that are used to take water for reactor cooling. A heat anomaly was noted in the lake water.

The radiocarbon concentration was determined using the scintillation method in benzene (Mikhailov *et al.*, 2003). The benzene synthesis from carbon-bearing sample (vegetation, biocarbonates, arbonic acid of air, dissolved compounds of water samples) was performed at the Institute of Geological Sciences of the National Academy of Sciences of Belarus using an installation which permits synthesis of most purified benzene in amounts sufficient for counting. ^{14}C beta-particles were counted using a highly sensitive counter LKB 1219 RACKBETA, "Guardian".

3. RESULTS OF INVESTIGATION AND DISCUSSION

In regions distant from the Ignalina and Chernobyl NPPs (**Fig. 3**) the ^{14}C content of natural objects is characterized by pMC values ranging from 92 to 107. These values are correlated with data on radiocarbon concentration in "pure" air of Central Europe (Levin *et al.*, 1985). In these data sampled a rather high concentration of radiocarbon in a moss sample collected in the Volozhin district is worthy of attention, as it is not accidental because of a contamination spot by the Chernobyl-born heavy radioisotopes located there. The same may be also true for the Medzozol lake region where high radiocarbon concentrations were noted in the lake vegetation and water.

Radiocarbon in the Ignalina NPP region

Radiocarbon concentrations were determined in samples of water, vegetation, biocarbonates and surface water (**Figs 4** and **5**). An increase of ^{14}C concentration to 150-190 pMC against the background values was noted in vegetation. Rather high values were noted however in not all the plant species studied. Maximum ^{14}C accumulations were noted in biennial cones of pine growing near the village of Pashevichi, as well as in rush samples collected near the Castle island shore in the Drisvyaty lake. High radiocarbon concentrations are typically noted in lake water (pMC 127-154) connected by canal with the Ignalina NPP. At the same time ^{14}C concentration in the Muysata Lake located 10 km northeastward of the nuclear power plant varied in different years from the present-day level (102 pMC) to rather high value (132 pMC). An increase of radiocarbon concentration (120-150 pMC) was also noted in carbonate of *Anadonta cygnaea* and *Dreissena polymorpha* shells sampled on the Drisvyaty and Muysata Lake coast (**Fig. 5**). An accumulation of excess radiocarbon was revealed in eggshell of poultly fed by grains grown on personal plot at the village of Pashevichi (152 pMC).

An analysis of the ^{14}C distribution in rings of pine trees growing nearly the Drisvyaty lake (**Fig. 6**) generally supports the distribution pattern of radiocarbon in natural objects investigated by the authors within 1994-2000. These data are well correlated with ^{14}C determinations in the Drisvyaty Lake water performed by Banis (1988). Within 1978-1986 radiocarbon concentrations in the lake water varied between 115 and 150 pMC.

The investigations carried out permit a conclusion that the ^{14}C concentration variations or temporal increases in natural objects of the Ignalina NPP region are associated with this radioactive isotope discharges into the atmosphere and the Drisvyaty Lake water in the course of the nuclear reactor operation.

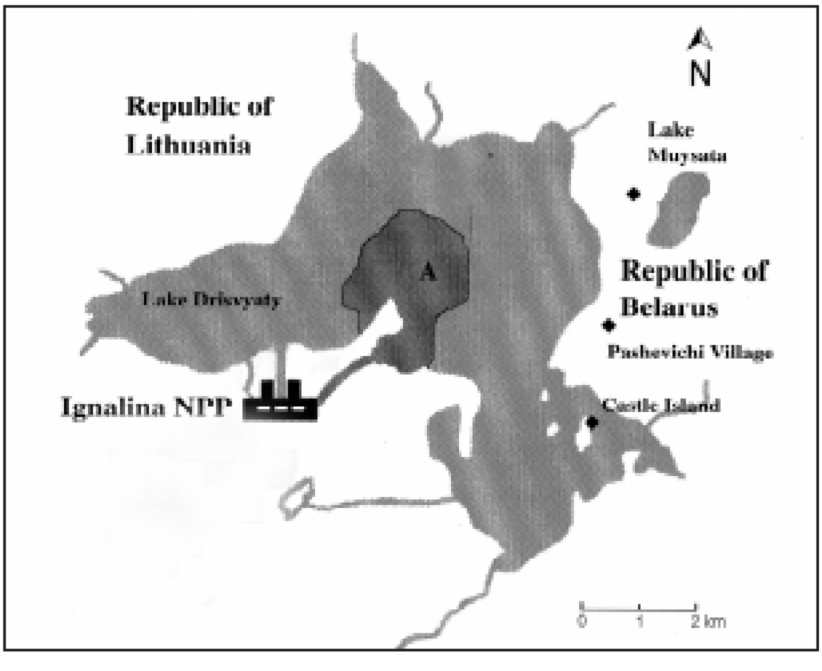

Fig. 2. Map showing sites of sample collection for radiocarbon analysis of – Ignalina NPP area.
◆ – sites of vegetation, lake water, air and biocarbonates sampling (Muysata Lake, Pashevichi Village, Castle and Island)

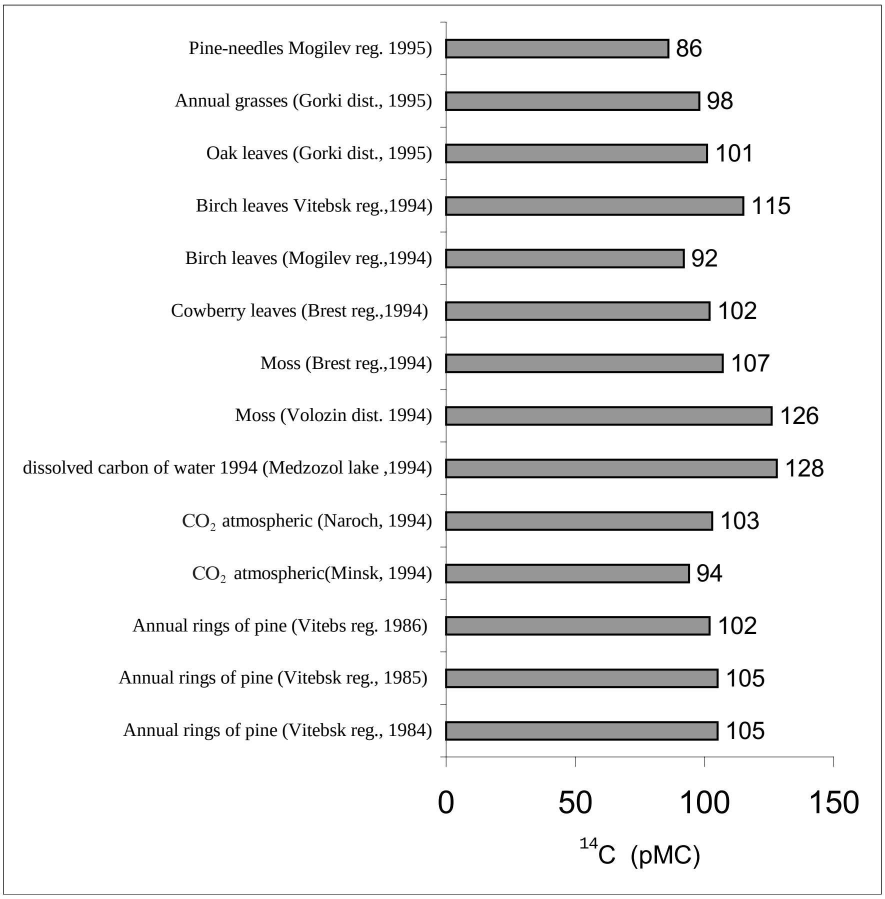

Fig. 3. *Radiocarbon concentration distribution in natural objects from Belarussian regions remote of any NPP*

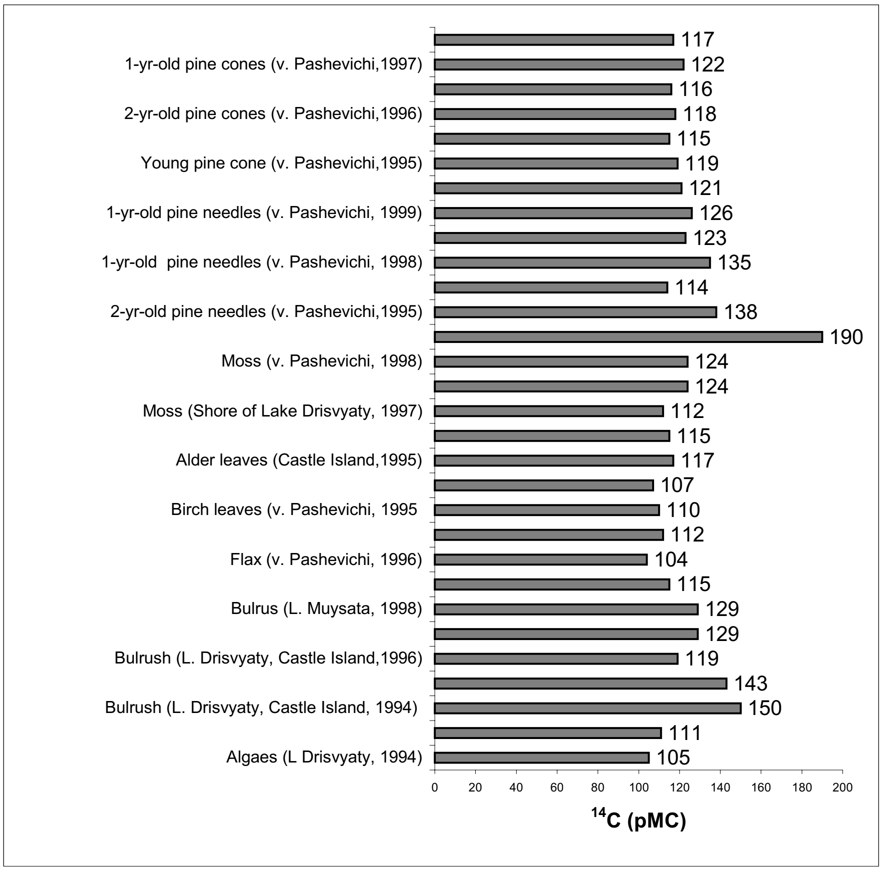

Fig. 4 *Radiocarbon concentration distribution in vegetation growing on shore of the Drisvyaty and Muysata Lakes, at the Pashevichi Village, in the Castle Island adjacent to the Ignalina NPP*

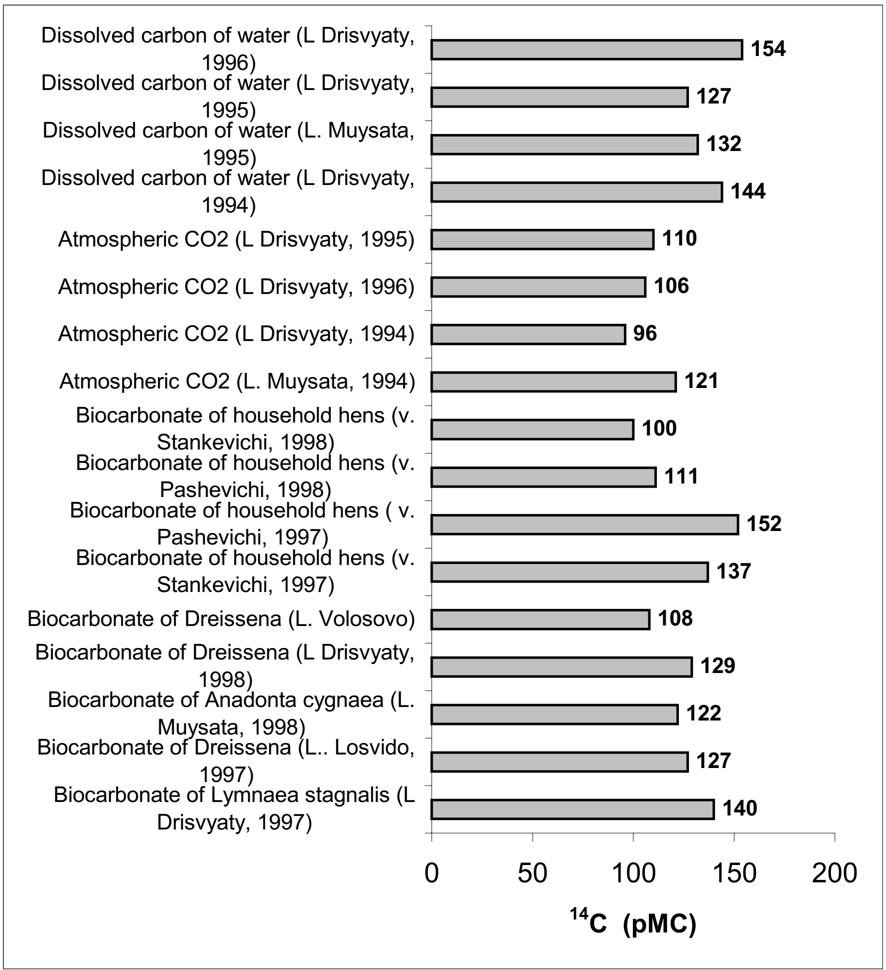

Fig. 5 *Radiocarbon concentration distribution in lake water and biocarbonates – from the Drisvyaty and Muysata Lakes, Pashevichi Village, Castle Island adjacent to the Ignalina NPP*

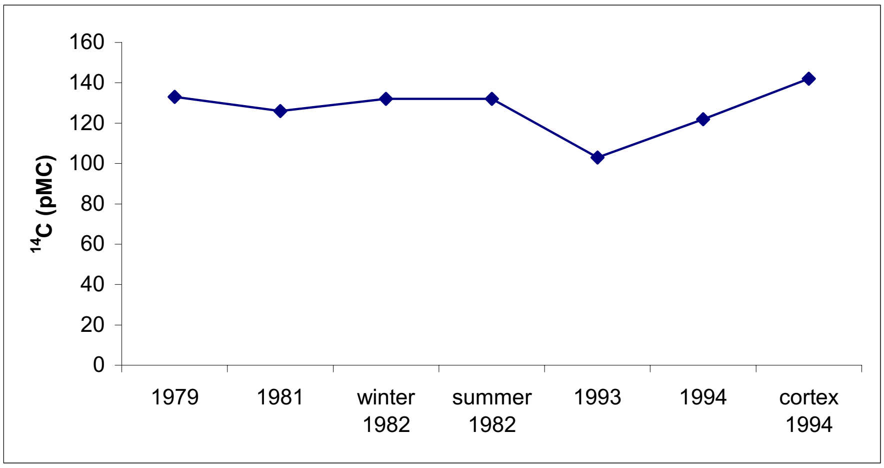

Fig. 6 *Radiocarbon concentration distribution in annual rings of pine growing along the Drisvyaty Lake shoreline*

Radiocarbon released by the Chernobyl NPP under accident and operating conditions

Vegetation sampled in 1994-1999 showed rather high ^{14}C concentrations (**Fig. 7**) as compared to the present-day values, and, at first sight, may reflect an impact of the Chernobyl NPP in operation. The radiocarbon content of samples collected in the Chernobyl and Ignalina NPPs regions shows a distinct similarity. However, our investigations showed that the Ignalina NPP impact on the ^{14}C concentration in natural objects does not manifest itself over large area, whereas in the Gomel region high radiocarbon concentrations in vegetation were noted in areas more than 100 km distant from the nuclear power plant. In this context an important question arise of whether high radiocarbon concentrations in vegetation are an impact of the Chernobyl NPP in operation (1994-1999), or the soil and vegetation medium still helds an excess ^{14}C fallen during the Chernobyl NPP accident. Data on the radiocarbon concentration in vegetation and air at the accident instant in 1986 (Mikhailov *et al.*, 1996) and in the soil cover of regions contaminated by heavy isotopes (Sr, Cs and others) can serve as a definite answer to the above question. For this purpose ^{14}C concentrations were determined in a collection of plants and in air at the instant of the

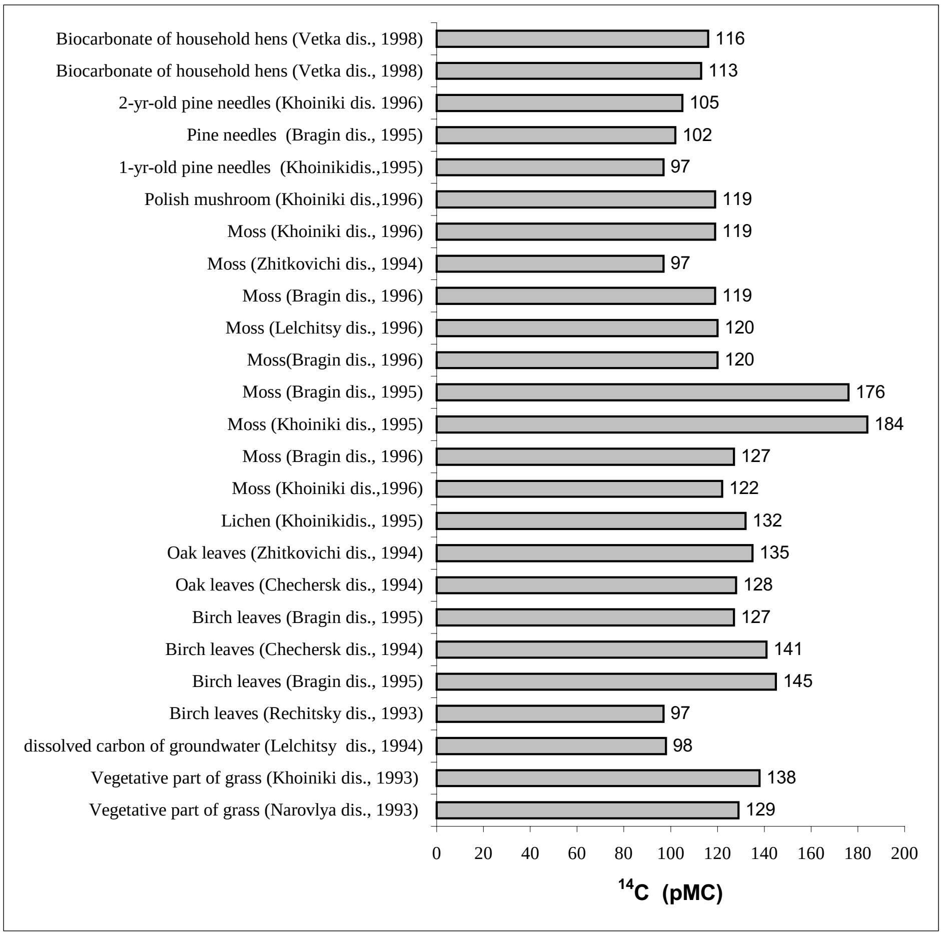

__Fig. 7__ Radiocarbon concentration distribution in natural objects from the Gomel region

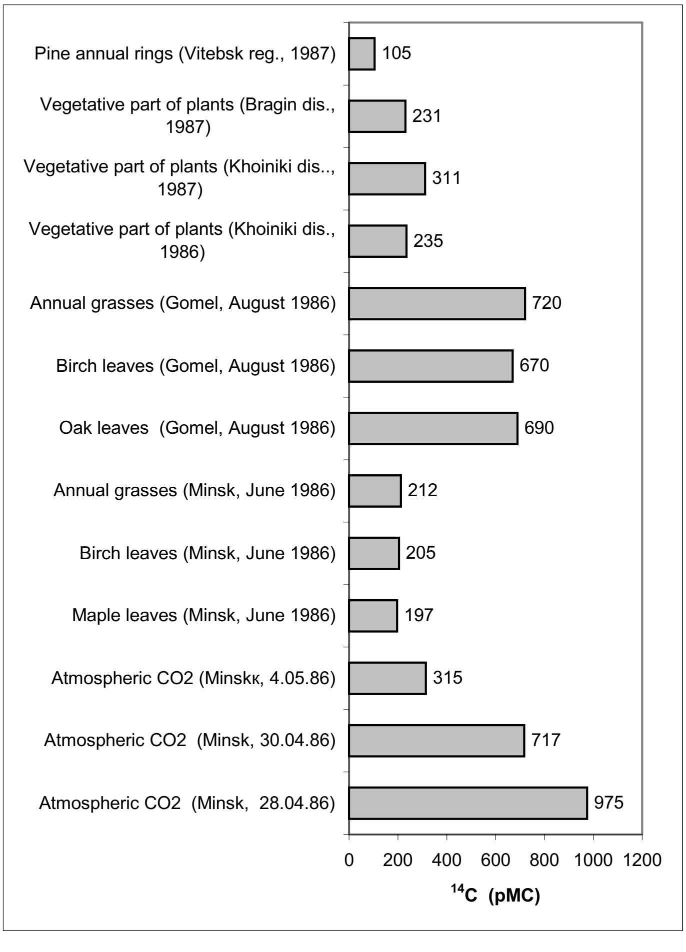

__Fig. 8.__ Radiocarbon concentration distribution in natural objects from the territory of Belarus immediately after the Chernobyl NPP accident (1986) and in the first year after the accident

accident in 1986 (Mikhailov *et al.*, 1996), and a possibility of ^{14}C fallout and fixation in soil together with the heavier radioisotopes (Sr, Cs, etc.) was studied.

Since the accident at the Chernobyl NPP took place in the vegetative season (end of April-beginning of May) a considerable portion of radioactive carbon abundant in the air was absorbed by plants (**Fig. 8**). This is indicated by ^{14}C activity of birch and oak foliage, annual grasses, which increased toward the nuclear power plant. So, in the Minsk region ^{14}C in vegetation measured 200 pMC, and in the Gomel region was as high as 600-700 pMC. Close by the Chernobyl NPP radiocarbon concentrations in vegetation even in 1987 were about 300 pMC and more (see **Fig. 8**). These data permit a realistic assessment of the radiocarbon fallout consequences, as its concentrations may be redistributed mainly due to biochemical processes. This may be proved by the results of studying the ^{14}C distribution along the vertical profile of the soil and bog vegetation medium (*Sphagnum* and Shreber moss) sampled in bog soils with Cs contamination >40 Ci/km^2 (village of Aravichi). The radiocarbon content of bog soils determined there was found much similar to the ^{14}C content of plant samples collected within 1986-1987 (**Fig. 9**).

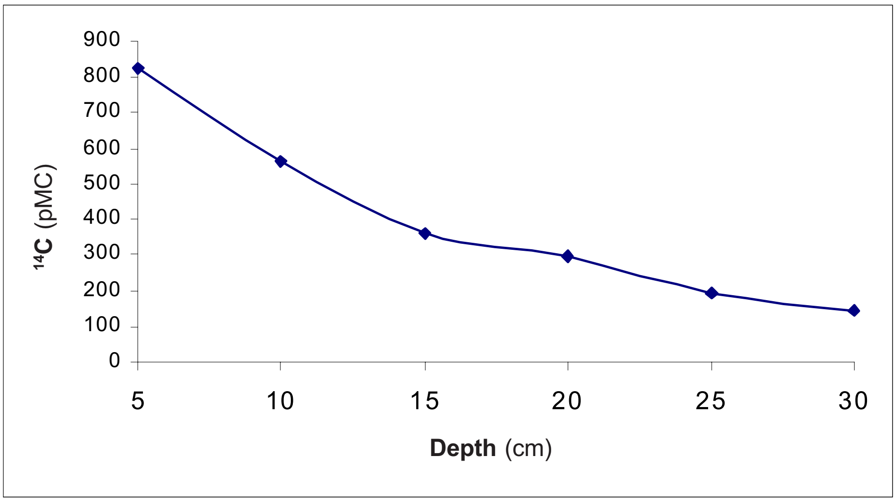

Fig. 9. *Radiocarbon concentration distribution along the bog land vertical section (Khoiniki district, village of Aravichi)*

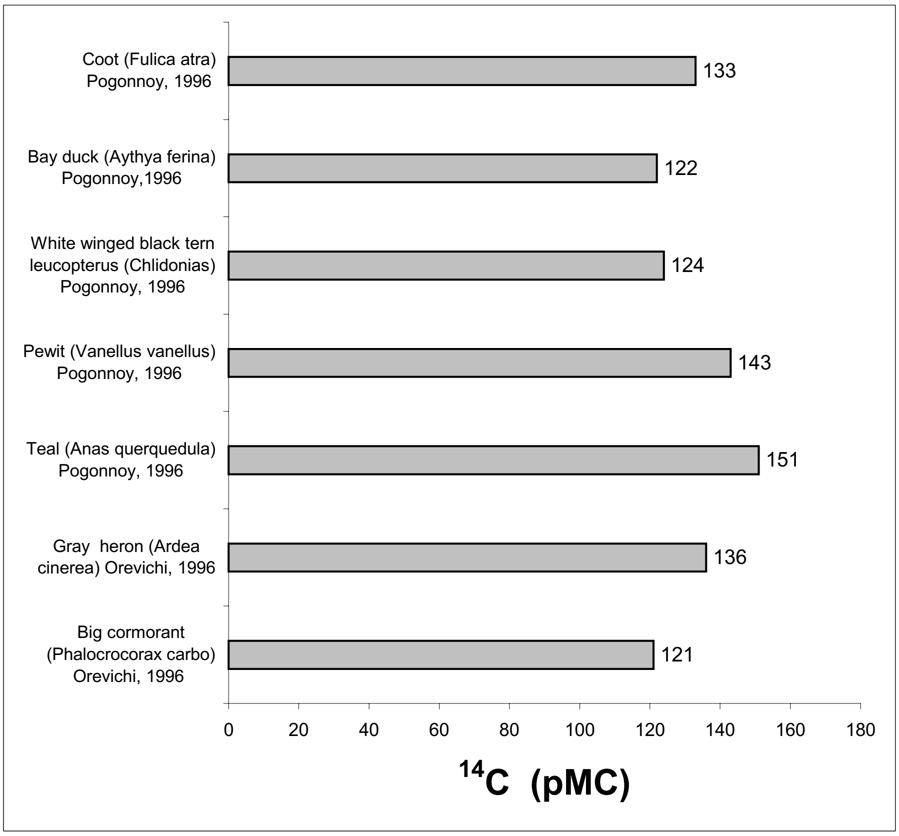

Fig. 10. *Radiocarbon concentration distribution in eggshell of wild birds inhabiting the Chernobyl 30 km zone (contamination over 40 Ci/km²).*

^{14}C concentrations in the upper part of the profile reach 825 pMC and regularly decrease with increasing depth to 145 pMC (25-30 cm layer). This supports the conclusion of Ukrainian radiochemists that maximum radiocarbon concentrations were fixed in natural objects situated very close to Chernobyl within nine years that followed the accident. Their investigation showed that the average values of this isotope concentration in vegetation growing in the 30 km zone were about 120 pMC (Kovalyukh *et al.*, 1996), as the accident-born ^{14}C migration and redistribution to the lower soil horizons took place. This permits a suggestion that the similarity between the radiocarbon concentration in vegetation sampled in 1993-1994 nearby the Chernobyl NPP and in regions well distant from the plant (Chechersk, Zhitkovichi and Volozhin) may be due to radiocarbon contamination of soils within these territories by the Chernobyl fallout.

A high radiocarbon concentration may be fixed not only in vegetation. It is seen in the ^{14}C distribution in eggshell of wildfowl dwelling in bogged area immediately in the Chernobyl zone where Cs contamination is 40 Ci/km^2. Among the studied kinds of birds there were: *Adrea cinerea* (gray heron), *Phalocrocorax carbo* (cormorant), *Chlidomias leucopterus*, *Fulka atra*, *Aythya ferina*, *Anas querquedula* (teal), *Vanellus vanellus* (lapwing) that are fed differently. Pochard, cormorant and lapwing are fed mainly in shallow-water reservoir and the top part of wet soil. The other kinds of birds mainly eat fish, but a mixed feeding is possible. Data obtained (**Fig.10**) show that high radiocarbon concentrations in eggshell are noted in the same regions where a high ^{14}C content of bog lands is observed. The highest concentrations of this radioisotope are characteristic of eggshell of wildfowl fed in closed shallow-water reservoirs and in the top wet soil layer.

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