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Analysis of long-term dynamics of radiocaesium mobility and plant availability in peat systems.

Journal article submitted.

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Executive Summary

Journal article submitted.

Dynamics of bioavailability of freshly added ^{137}Cs has been studied in the 2-year laboratory experiments for the peat-bog soils of Rokytne district of Rivne region. The obtained parameters of dynamics of root uptake of ^{137}Cs by herbaceous plants in the peat-bog soils with abnormally high bioavailability differ from those previously reported in the literature. Fast half-loss period of the ^{137}Cs concentration ratio has been estimated as 0.2-0.3 y. After the fast decrease at the beginning of the experiments, the concentration ratios reached the stable level in 1-2 years. Ratios of the fast to slow-loss components of the concentration ratio at the moment of radiocesium injection into soil were estimated as 3-5.

Journal article have been published with reference to the COMET project (http://jnpae.kinr.kiev.ua/16.3.html):

Maloshtan, I., Polischuk, S., Khomutinin, Yu., Kashparov, V. 2015. Dynamics of ^{137}Cs uptake to herbaceous plants at the peat-bog soils with abnormally high radiocesium bioavailability. *Nuclear Physics and Atomic Energy*, v.16, No.3, p. 263-272. (Ukr.) (Annex : Journal article submitted - pdf file).

Introduction

The reason of the high ^{137}Cs specific activities in milk at the comparable low terrestrial contamination density (about 100 kBq m^{-2}) is utilization of the natural meadows as the pastures and hayfields. Such meadows are formed at peat and peat-bog soils. Specific physical-chemical and agrochemical properties of these soils facilitate high migration ability and bioavailability of ^{137}Cs in soil-plant system and, respectively, high transfer of radiocesium into milk (Prister et al., 1989).

Monitoring of ^{137}Cs in milk was performed in the Rovno region of Ukraine after Chernobyl accident for wet peat ecosystems (density of ^{137}Cs contamination about 100 kBq/m^2) was continued (http://www.uiar.org.ua/). Effective half-lives for ^{137}Cs to locally produced food products that are in used for radiological assessments in wet peat environments are T_{eff2}=28 years (Fig.1).

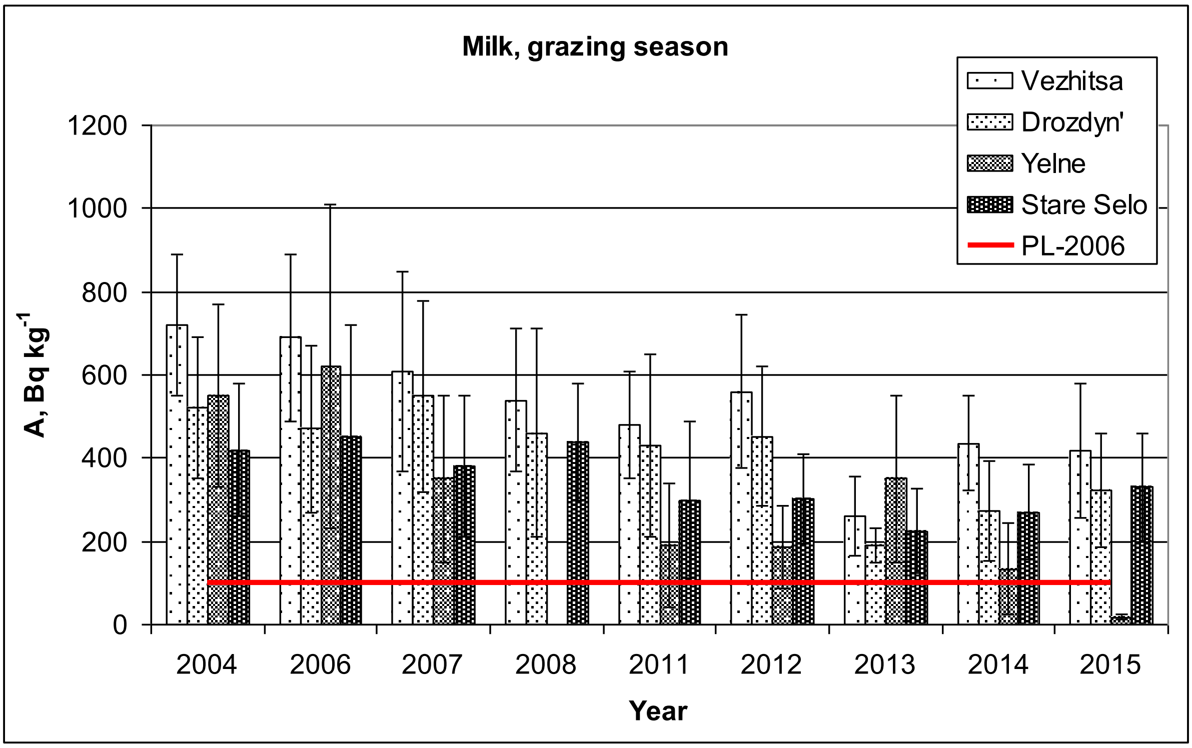

Fig. 1 – The dynamics of the milk contamination by 137Cs which is produced in the private farms of the most critical settlements during the grazing period (arithmetic mean, standard deviation, n> 20) (UIAR, 2015)

For the first time the abnormal high bioavailability of ^{137}Cs in the peat-bog soils of Polissya was noticed long before the Chernobyl accident, in the 1970-es, by Marey et al. (1974). After the accident the high values of transfer factors (TF_{ag}) and concentration ratios (CR) of ^{137}Cs into natural grass at the peat-bog soils that were one-two orders of magnitude higher, than at the mineral soils, were reported by Perepelyatnikov and Ilyin (1991), Ilyin (1993, 1996), Perepelyatnikov (2001), Prister et al. (2001) and were reflected in the reports of the international projects (The behaviour of radionuclides in natural and semi-natural environments, 1996; IAEA, 2006).

Beside the waterlogged peats, abnormal high bioavailability of radiocesium that slowly changes in time is also known for arctic and alpine ecosystems due to the low rate of decay of soil organic matter at the high moisture content and low temperature. As a result, such soils are characterized by high acidity of soil solution, high concentration of ammonia and nutritional deficiency. High values of ^{137}Cs TF also are caused by the species composition, low productivity and quality of the natural meadows, and by the soil properties: high content of organic matter (> 70 %) in peat-bog soils formed at sands and low contents of clay minerals, phosphorus, exchangeable potassium and calcium (IAEA, 2009).

Dynamics of radiocesium TF_{ag} into plants are usually described with the two-exponent dependence that reflects irreversible fixation of radionuclide by soil, or ageing effect (IAEA, 2010):

$$TF_{ag}(t) = TF_{ag}^{f}(0)\cdot \exp(-\ln(2)\cdot t/T_e^{f}) + TF_{ag}^{s}(0)\cdot \exp(-\ln(2)\cdot t/T_e^{s})$$

where $TF_{ag}^{f}(0)$ and $TF_{ag}^{s}(0)$ are the contributions of the fast and slow loss components, respectively, into the value of TF_{ag} in the moment of deposition, and T_e^{f} and T_e^{s} are the periods of the fast and slow half-loss. The same expression is also applied for dynamics of radiocesium CR.

According to the National Report of Ukraine (20 years after…, 2006), for hay of natural meadows on peat-bog soils the values of the fast and slow components of ^{137}Cs TF_{ag} at the moment of deposition (1986) differed by one order of magnitude ($TF_{ag}^{f}(0) = 218$ and $TF_{ag}^{s}(0) = 22$ (Bq kg^{-1}) (kBq m^{-2})$^{-1}$), while the half-loss periods were 0.89 and 6.6 years for the fast and slow components and 6.6 year for the slow components, respectively. The presented slow component half-loss period for peat-bog soil was lowest among those for soddy-podsolic, grey forest and chernozem soils (20, 44 and 112 years, respectively). However, it contradicts to the observed dynamics of the radiocesium concentration in milk of the cows grazing at the natural pastures at peat-bog soils in the critical settlements of Rokytne district of Rivne region (Fig. 2), from which T_e^s can be estimated as 26 years. From Fig. 2 $TF_{ag}^s(0)$ for ^{137}Cs in hay of natural grasses at the peat-bog soil can be obtained as

$$TF_{ag}^s(0) = \frac{15}{8 \cdot 0.01} \approx 190 \ \ (\mathrm{Bq\,kg^{-1}})\,(\mathrm{kBq\,m^{-2}})^{-1},$$

where

15 is extrapolated to 1986 value of transfer factor for milk, $(\mathrm{Bq\ L^{-1}})\ (\mathrm{kBq\ m^{-2}})^{-1}$,

8 is an average daily consumption of hay by lactating cow, kg,

and 0.01 is transfer coefficient from hay to milk, $\mathrm{L^{-1}}$.

Derived value is in good accordance with the ^{137}Cs TF_{ag} value of 130 $(\mathrm{Bq\ kg^{-1}})\ (\mathrm{kBq\ m^{-2}})^{-1}$ obtained at peat soils near village Yelne in 2004 (Fesenko et al., 2013), but exceeds almost by order of magnitude the above mentioned value of 22 $(\mathrm{Bq\ kg^{-1}})\ (\mathrm{kBq\ m^{-2}})^{-1}$ from the National Report of Ukraine (20 years after…, 2006). IAEA (2009) recommends for organic soils $T_e^f = 1.8$ years and $T_e^s = 11$ years, which also does not correspond to the observed dynamics of radiocesium contamination of natural grasses at the peat-bog soils of north-western Polissya of Ukraine at the late phase after the Chernobyl accident.

Our study is aimed to fill this gap and to estimate in the long-term laboratory experiment the values of ^{137}Cs CR and Tf in the peat-bog soils with the abnormal high bioavailability.

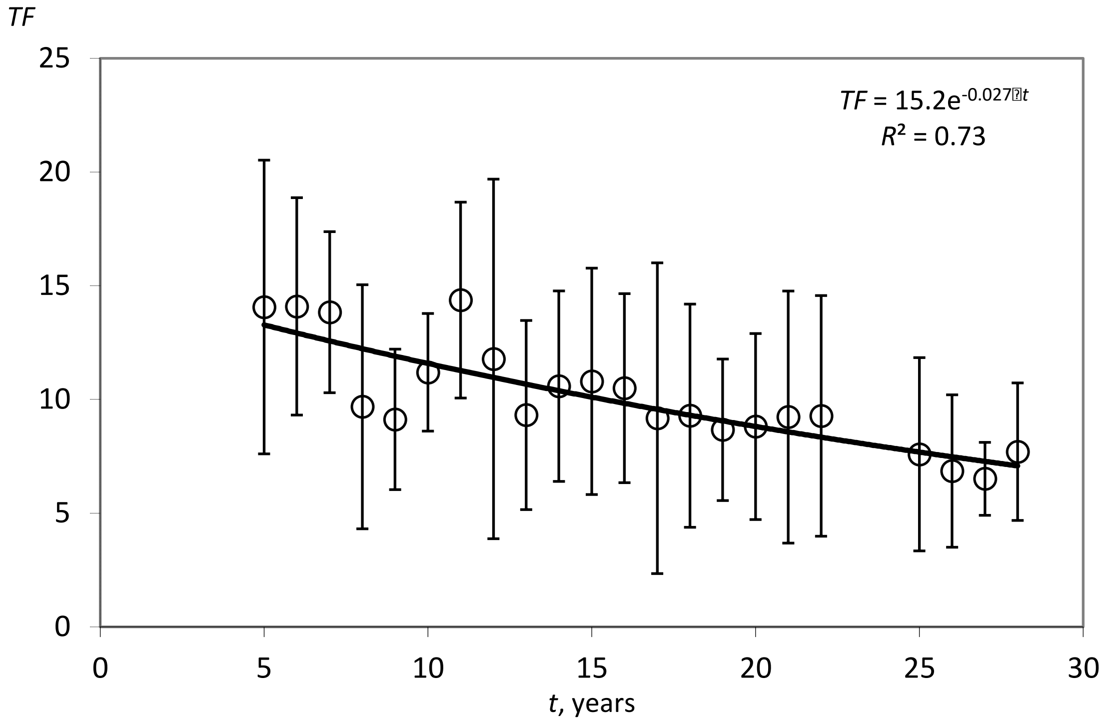

Fig. 2. Dynamics of ^{137}Cs TF, (Bq L^{-1}) (kBq m^{-2})$^{-1}$, in cow milk in the critical settlements of the north-western Polissya of Ukraine (Vezhytsya, Grabun', Drozdyn', Yelne, Zabolottya, Perekhodychi, Stare Selo) according to the dosimetric surveys of 1991-2012 (Data on ^{137}Cs specific activities in milk…) and results of monitoring performed by UIAR NUBiP of Ukraine in 2013-2014 (http://www.uiar.org.ua/).

Material and methods

Methods

Dynamics of bioavailability of freshly added and Chernobyl ^{137}Cs were studied in the long-term experiments in controlled laboratory conditions for two peat-bog soils. As a model plant, we used spiny rush (*Juncus acutus* L.), undemanding to soil and climate conditions perennial grass which is widely spread at the peat-bog soils and is easily identified.

Typical for north-western Polissya of Ukraine peat-bog soils were collected at the marshes near villages Vezhytsya (N 51.5752º, E 27.1309º, soil #1) and Yelne (N 51.4847º, E 27.0629º, soil #2) in Rokytne district of Rivne region, at the territories where the values of ^{137}Cs transfer factors into the natural grasses reach 100-200 (Bq kg^{-1}) (kBq m^{-2})$^{-1}$ (Kashparov et al, 2008 and 2011; Khomenko and Polischuk, 2014). Soil was collected from the upper 20-cm root inhabited layer. After excavation with the shovel, roots were removed from the soil.

Physical-chemical and agrochemical properties of collected soils and specific activities of ^{137}Cs of Chernobyl fallout were determined using the standard methods

Results

Ratio of CR_{fresh} to $CR_{Chernobyl}$ decreased in time for both soils due to decrease of bioavailable forms of freshly added radiocesium in soil. For *CR(t)* an expression similar to (1) for $TF_{ag}(t)$ can be applied, which takes into account irreversible fixation of radiocesium by soil (IAEA, 2010). However, such expression predicts that radiocesium $CR(t) \to 0$ at $t >> T_e^s$, while with approach to equilibrium between radioactive and stable cesium in soil their CR become similar, i.e. radiocesium $CR(t) \to CR_{stable}$ (Yoshida et al., 2004). Therefore,

$$CR(t) = CR^f(0) \cdot \exp(-\ln(2) \cdot t / T_e^f) + CR^s(0) \cdot \exp(-\ln(2) \cdot t / T_e^s) + CR_{stable}$$

Then ratio $CR_{fresh} / CR_{Chernobyl}$ at при $T_e^f << T_e^s$ and $t << T_e^s$ can be approximated as

$$\frac{CR_{fresh}(t)}{CR_{Chernobyl}(t)} = \frac{CR^f(0) \cdot \exp(-\ln(2) \cdot t / T_e^f) + CR^s(0) \cdot \exp(-\ln(2) \cdot t / T_e^s) + CR_{stable}}{CR^f(0) \cdot \exp(-\ln(2) \cdot (26.8 + t) / T_e^f) + CR^s(0) \cdot \exp(-\ln(2) \cdot (26.8 + t) / T_e^s) + CR_{stable}} \approx$$

$$\frac{CR^f(0) \cdot \exp(-0.693 \cdot t / T_e^f) + CR^s(0) + CR_{stable}}{CR^s(0) \cdot \exp(-0.693 \cdot (26.8 + t) / T_e^s) + CR_{stable}} \approx A_1 \cdot \exp(-0.693 \cdot t / T_e^f) + A_2$$

Due to the ^{137}Cs injection in peat-bog soils (reconstruction of Chernobyl fallout in the first years after the accident) the dynamics of 'fresh' T_e^f = 0.2-0.3 y from the peat-bog soils with unusually high transfers to the grass was obtained for the first time (Fig.3)

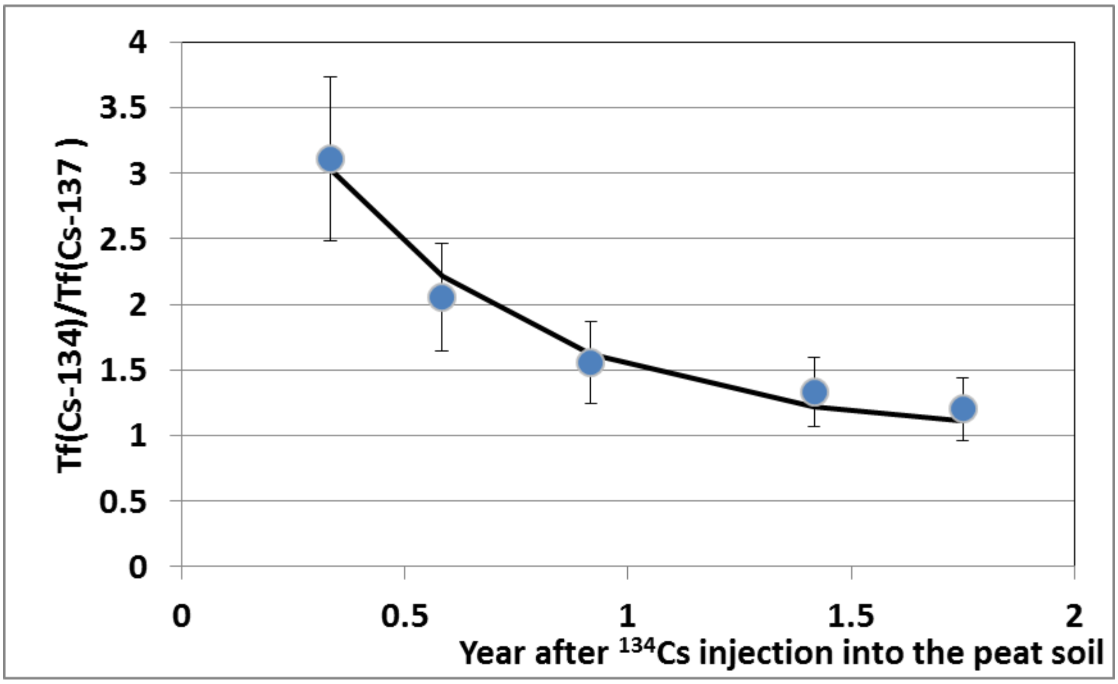

Fig. 3. The dynamics of the relationship 'fresh' **Tf'$_{Cs\text{-}137}$(t)** to "Chernobyl" **Tf$_{Cs\text{-}137}$(t+26.8 year)** radiocesium from peat-bog soils to grass (solid line - theoretical dependence:

Conclusions

Obtained in the laboratory experiments parameters of dynamics of the ^{137}Cs root uptake by grass plants at the peat-bog soils with the abnormal high radiocesium bioavailability differ from those presented in the literature sources.

Fast half-loss period of the ^{137}Cs concentration ratio can be estimated as T_e^f = 0.2-0.3 years. In 1-2 years after the deposition concentration ratio of radiocesium decreased to the equilibrium level. Ratio of the fast component of concentration ratio $CR^f(0)$ to the sum of slow and stable components $CR^s(0)+CR_{stable}$ in our experiments was 3-5.

For specification of parameters of dynamics of the radiocesium soil-to-plant root uptake at the peat-bog soils, especially in the early period after deposition, additional experiments are needed. These experiments should be performed in the close to natural conditions with account to redistribution of the radionuclide in soil profile and litter, changes of temperature and moisture regimes, seasonal variations of the biomass growth and activity of the soil microbiota.

Acknowledgement

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ANNEX

Annex: Journal article submitted "**Maloshtan, I., Polischuk, S., Khomutinin, Yu., Kashparov, V. 2015. Dynamics of ^{137}Cs uptake to herbaceous plants at the peat-bog soils with abnormally high radiocesium bioavailability. *Nuclear Physics and Atomic Energy*, v.16, No.3, p. 263-272. (Ukr.)**." (pdf file). http://jnpae.kinr.kiev.ua/15.2.html