The Transfer of Dissolved CS-137 from Soil to Plants

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ABSTRACT

Rapidly maturing plants were grown simultaneously at the same experimental sites under natural conditions at the Chornobyl Exclusion Zone. Roots of the plants were side by side in the soil. During two seasons we selected samples of the plants and of the soils several times every season. Content of Cs-137 in the plant and in the soil solution extracted from the samples of soils was measured. Results of measurements of the samples show that, for the experimental site, Cs-137 content in the plant varies with date of the sample selection. The plant:soil solution Cs-137 concentration ratio depends strongly on the date of selection and also on the type of soil. After analysis of the data we conclude that Cs-137 plant uptake is approximately proportional to the content of dissolved Cs-137 in the soil per unit of volume, and the plant:soil solution Cs-137 concentration ratio for the soil is approximately proportional to the soil moisture.

INTRODUCTION

The problem of transition of radionuclides to plants is significant for many countries, because there is always the possibility of an accident "a la Chornobyl". Moreover, soil may be polluted with radionuclides from other sources. Now one of the most acute problems is the transport of the long-life isotopes of cesium – Cs-137.

Many investigations of cesium uptake by plants have been performed during last few years. Different theories have been postulated for those parameters that affect the uptake of Cs-137 by plants. Many researchers consider K as an analog of Cs-137 in respect of plant uptake. (See the review [1,2]).

Johanson K.J., Dolgilevich M.I., Vasenkov G.I. [3] state that one of the main parameters for plant uptake of Cs-137 is the content of organic substance in the soil (especially humic acids).

Association of Cs-137 transfer soil-to-plant factor and organic matter content in soil was absent in the data analyzed by Nisbet, A.F., & Woodman, R.F.M. [4].

Àgapkina G.I. et al. [5] affirm that the greater part of the radionuclides at soil solutions are associated with different molecular fractions of the organic matter. These researchers affirm that the association of the radionuclides with molecules of the soil solution is very important for transition of the radionuclides from the soil to the plants. Plant:soil solution Cs-137 concentration ratio for the different molecular weight fractions differ sometimes by two mathematical orders. And these researchers assert, that the most "bioavailable" is the fraction with mass 1000 atomic units. This is not the accepted opinion.

The plant:soil solution Cs-137 concentration ratio significantly relates to the K concentration in the soil solution (Smolders E., Van den Brande K.[6], and Merckx R**.**, and Absalom J.P. et.al. [7]).
The theory of Prister B.S., Biesold H., Devill-Cavelin G. [8], is founded on consideration of soil as a three phase system where soil solution reaction (pH), absorbing capacity (E) and content of organic matter (OM) are the main characteristics. The area of an effective section (relative units) calculated as an area of the triangle with apexes lying in coordinates of normalized vectors pH, E and OM in three-dimensional space is used for complete estimation of Cs-137 transition to plant.

According to data of Prister B.S., Perepelyatnikov, G.P., Perepelyatnikova, L.V. [9], plant Cs-137 uptake increases with increasing of the soil moisture. However the data of Ehlken S., Kirchner G. [10], show that plant Cs-137 uptake decreases with increasing of the soil moisture.

Thus it is very complicated problem to predict plant uptake of Cs from the soil on the base of the soil chemical, physical and other parameters. Investigations continue because no reliable theory for prediction of transition of cesium to plants has been developed.

In this paper we are looking for the main parameter that, in the first order, determines Cs-137 plant uptake. We think that Cs-137 uptake is determined by parameters of the soil solution. We investigated dependence of plant Cs-137 uptake on content of dissolved Cs-137 in the soil per unit of volume. To obtain content of dissolved Cs-137 in the soil per unit of volume of the soil, we multiply content of Cs-137 in the soil solution by water content at a unit of volume of the soil. We do not consider the connection between chemical, physical and other parameters of soil and content of the soil solution.

EXPERIMENTAL

We carried out our investigations at field conditions and investigated growth at three experimental field sites with different types of soil using different plants. The sites are situated in 30 km from Chornobyl Nuclear Power Plant in the North-East direction.

- site A – based on glacial-water sand soddy-podzol middle podzol silt sand soil; average pollution with Cs-137 is near 980 kBq/m^2;
- site B – based on decomposed middle thick sedge, rush and wood peat low-lying type peat soil; average pollution with Cs-137 is near 13 000 kBq/m^2;

site C – based on glacial-water sand loam soddy-podzol middle podzol sand loam with the signs of temporal excess humidification soil; average pollution with Cs-137 is near 620 kBq/m^2.

Dimensions of every experimental site were 2x4 meters. Every experimental site was dug in spring of 2002 and 2003.We sowed, mixed together, at the sites A, B, C rapidly maturing plants that are as divergent from each other as possible. These are radish (raphanus sativus), salad (lactuca sativa), and watercress (lepidium sativum). The plants were sowed within every experimental site several times during each season. Parameters of the soil solution for all plants were the same, as they were growing jointly at the same site at the same time.

We selected samples several times every season. All dates of samples selection are pointed below. We took soil with a sampler with diameter 5cm and length 20 cm for five times at the every land. All soils selected at the same time at the same land soil were mixed. Every time we harvested samples of soils, samples of the sowed plants, (if they had grown), and samples of wild plants. These wild plants are elytrigia repens, phleum pratense, polygonum hydropiper, urticaceae dioica, barbarea vulgaris, convolvulus arvensis, rumex acetosa, stachys palustris. As a rule, we selected whole plants. But sometime it was only part of plant (leafs or roots). We harvest all plants at the every experimental site at every sample selection. But we did not harvest sprouts of plants so they can continue to grow. The mass of selected plants of every kind was in rang 5g – 500g.

The plants were sampled before blossoming and were washed and dried.

The soil solution was extracted from the soil using the centrifuge RS-6 (former USSR) with a centripetal acceleration of 3,000**g**. We extracted soil solution from some samples immediately. If a sample of soil was too dry for extraction of soil solution, we extracted soil solution after addition of distilled water to the sample and treating it with a rotator for an hour. Our estimation for all our investigated soils show that addition of water to the soil changes ^{137}Cs concentration in soil solution by not more than 5%, if soil moisture do not exceed moisture of water saturated soil. We used our experimental data on Cs-137 concentration in soil solution, as well as on soil moisture, Cs-137 concentration in the soil and literature data on content of exchangeable Cs-137 in Chornobyl soils (Ageyetz V.Yu. [11]) to estimate the change of Cs-137 concentration in the soil solution due to water addition to the soil. We can describe this concentration $\mathbf{A_1}$ as $\mathbf{A_1=A_s/K_d^{ex}}$, where $\mathbf{A_s}$ is concentration of solid exchangeable Cs-137 in the soil, $\mathbf{K_d^{ex}}$ is Cs-137 exchangeable sorption distribution coefficient for the soil from Konoplev A.V., Konopleva I.V. [12].
Our experimental data for site A confirm that addition of water at so low a concentration does not change Cs-137 concentration in soil solution within experimental error.

As a rule, we extracted near 200 ml of soil solution at every extraction. The centrifuged solution was filtered through the glass filter Whatman GF/A and through the filter TU 6-09-1678-86 (former USSR). The soil solutions after filtration were clear. To conserve the obtained soil solution, we added to the soil solution nitric acid in proportion 1 ml of concentrated nitric acid per 500 ml of the soil solution and heated it to boiling.

We determined moisture of soil, **h,** as the ratio of mass contained in the sample of soil water to the mass contained in the sample dry soil within experimental error 10%.

We determined Cs-137 content (activity) per unit of mass in dried plants and in soil solutions with gamma-spectrometer ORTEC to be within experimental error 5% - 15%.

RESULTS AND DISCUSSION

We represent obtained data for all our experimental field sites in the Tables I-III. We used this data for our analysis. Some data in our tables is absent because of difficulties of field experiments.

Table I. Site A. Content of Cs-137 in Dried Plants and in the Corresponding Soil Solutions. Soil Moisture – at the Time of the Plant Selection for Plants and at the Time of Beginning of Extraction of the Soil Solution for Soil Solutions.

Soil humidity h	**Date**	**Sample name**	**Sample №**	**Cs-137 Bq/kg**	**K mg/g**
0,23	12May 2002	*Phleum pratense*	57	10880	28,65
0,11	09June2002	*Soil solution*	26	8	0,0070
-- " --	09June2002	*Phleum pratense*	58	5276	20,96
0,062	22June 2002	Roots of *raphanus sativus*	55	1746	64,70
-- " --	22June 2002	Leaf of *raphanus sativus*	65	2018	43,49
-- " --	22June 2002	*Lepidium sativum*	67	4540	48,30
0,65	14July 2002	*Soil solution*	22	7,9	0,0282
0,094	14July 2002	*Phleum pratense*	75	5725	12,15
-- " --	14July 2002	*Stachys palustris*	76	10206	27,67
0,29	08June 2003	*Soil solution*	5	4,9	0,0214
0,016	08June 2003	Leaf of *raphanus sativus*	35	216	52,34
-- " --	08June 2003	*Lepidium sativum*	36	265	38,28
-- " --	08June 2003	*Lactuca sativa*	37	672	52,617
-- " --	08June 2003	*Stachys palustris*	38	606	29,774
0,30	13July 2003	*Soil solution*	8	3,2	0,0217
0,035	13July 2003	*Lactuca sativa*	21	585	47,39
-- " --	13July 2003	*Polygonum hydropiper*	22	679	34,07
-- " --	13July 2003	*Stachys palustris*	24	1722	32,35
0,17	07Oct 2003	*Soil solution*	10	1,1	
-- " --	07Oct 2003	Roots of *raphanus sativus*	44	1529	18,70
-- " --	07Oct 2003	*Rumex acetosa*	45	3138	13,81
-- " --	07Oct 2003	Leaf of *raphanus sativus*	49	2226	21,43

Table II. Site B. Content of Cs-137 in Dried Plants and in the Corresponding Soil Solutions. Soil Moisture – at the Time of the Plant Selection for Plants and at Time of Beginning of Extraction of the Soil Solution for Soil Solutions.

Soil humidity h	**Date**	**Sample name**	**Sample №**	**Cs-137 Bq/kg**	**K mg/g**
2.35	09 June 2002	Soil solution	24	61	0,0045
2.35	09 June 2002	*Urticaceae dioica*	59	609400	11,42
-- “ --	09 June 2002	*Polygonum hydropiper*	60	606600	23,56
3.20	22 June 2002	Soil solution	19	40,8	0,007
2.06	22June 2002	Roots of *raphanus sativus*	54	240760	39,29
-- “ --	22 June 2002	Leaf of *raphanus sativus*	64	414700	26,84
-- “ --	22 June 2002	*Lepidium sativum*	68	758800	39,8
-- “ --	22 June 2002	*Polygonum hydropiper*	69	305700	18,71
3.20	14 July 2002	Soil solution	23	95	0,0047
1.46	14 July 2002	*Urticaceae dioica*	72	230900	6,704
-- “ --	14 July 2002	Motley grass	73	617500	9,522
1.38	08 June 2003	Soil solution	6	44,3	0,0217
0.45	08 June 2003	*Lactuca sativa*	39	35911	31,28
-- “ --	08 June 2003	Blades of *elytrigia repens*	41	47741	17,03
-- “ --	08 June 2003	*Urticaceae dioica*	42	55260	12,79
-- “ --	08 June 2003	*Polygonum hydropiper*	43	118888	17,5
0.72	13 July 2003	Soil solution	9	27	0,020
-- “ --	13 July 2003	*Lactuca sativa*	26	64924	40,25
-- “ --	13 July 2003	Leaf of *raphanus sativus*	27	26130	49,78
-- “ --	13 July 2003	*Polygonum hydropiper*	28	54348	24,06
-- “ --	13 July 2003	Blades of *elytrigia repens*	29	36505	27,18
1.88	07 Oct 2003	Soil solution	11	22,3	
1.88	07 Oct 2003	Blades of *elytrigia repens*	46	56897	6,63
-- “ --	07 Oct 2003	Raphanus sativus	48	82121	11,70

Table III. Site C. Content of Cs-137 in Dried Plants and in the Corresponding Soil Solutions. Soil Moisture – at the Time of the Plant Selection for Plants and at the Time of Beginning of Extraction of the Soil Solution for Soil Solutions.

Soil humidity **h**	Date	Sample name	Sample №	Cs-137 Bq/kg	K mg/g
0,34	09June 2002	Soil solution	12	4,8	0,0475
0,069	09June 2002	*Barbarea vulgaris*	61	5212	32,26
-- “ --	09June 2002	*Convolulas arvensis*	62	7772	36,52
0,15	08June2003	Soil solution	4	1,9	0,0475
	08June2003	Leafs of *raphanus sativus*	31	82	40,203
	08June2003	*Lepidium sativum*	32	65	25,675
	08June2003	*Convolulas arvensis*	34	93	34,455
0,12	13July2003	Soil solution	7	7,1	0,0528
	13July2003	Leafs of *raphanus sativus*	16	164	50,7
	13July2003	*Convolulas arvensis*	19	165	38,09
	13July2003	*Barbarea vulgaris*	20	105	31,61

The data on Cs-137 content in plants for sites A, B and C we can see in Fig.1.

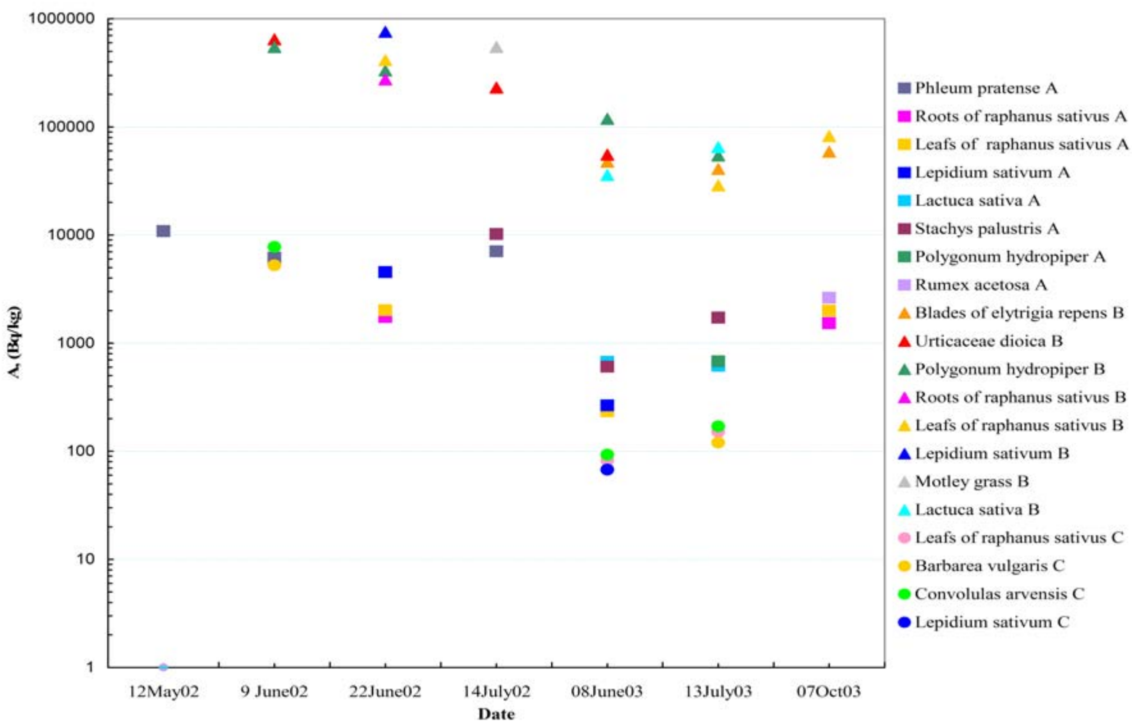

Fig. 1. Content of ^{137}Cs in dried plants **A**(Bq/kg) for site A, site B, site C. The plants' names are on the right from the diagram. A – site A, B - site B, C – site C. The dates of the plant selection are represented below from the diagram.

We can see from these results that content of Cs-137 in the same plant from the same land changes with the date of its collection. Content of Cs-137 in all selected at the same site at the same date different kinds plants differ not more than twice from some average content for the land and for the date of selection. These differences are due to inhomogeneity of ^{137}Cs distribution in the soil plus due to change of soil solution parameters during the plants growing plus due to different Cs-137 uptake by these different kinds of the plants and may also be due to other factors. So, we can conclude, that as a rule, for every our experimental sites Cs-137 content in the plant varies due to inhomogeneity of Cs-137 distribution in the soil not more than twice from some average value. For the majority of cases, for every investigated site, the difference in Cs-137 content between two selected at different times the same kinds of plants is more than difference in ^{137}Cs content between two selected at the same time different kinds of plants. We can see from Fig.1, that content of Cs-137 in the selected in different times the same kind plants from the same site differ sometime in more than 10 times. Such a big deviation in Cs-137 content in the same kind plants (10 times) due to inhomogeneity of Cs-137 distribution in the soil and due to other above pointed factors has a low probability. So deviation for several plants simultaneously has negligible probability, because the probability of several random events is equals to product of probabilities of all these events, according to calculus of probabilities. Consequently, so deviation in plants Cs-137 content for the experimental sites for different dates

cannot be determined by inhomogeneity of Cs-137 distribution in the soil. We must look for other reasons of the differences in this case.
In Fig.2 plant:soil solution Cs-137 concentration ratio for all the investigated sites and plants

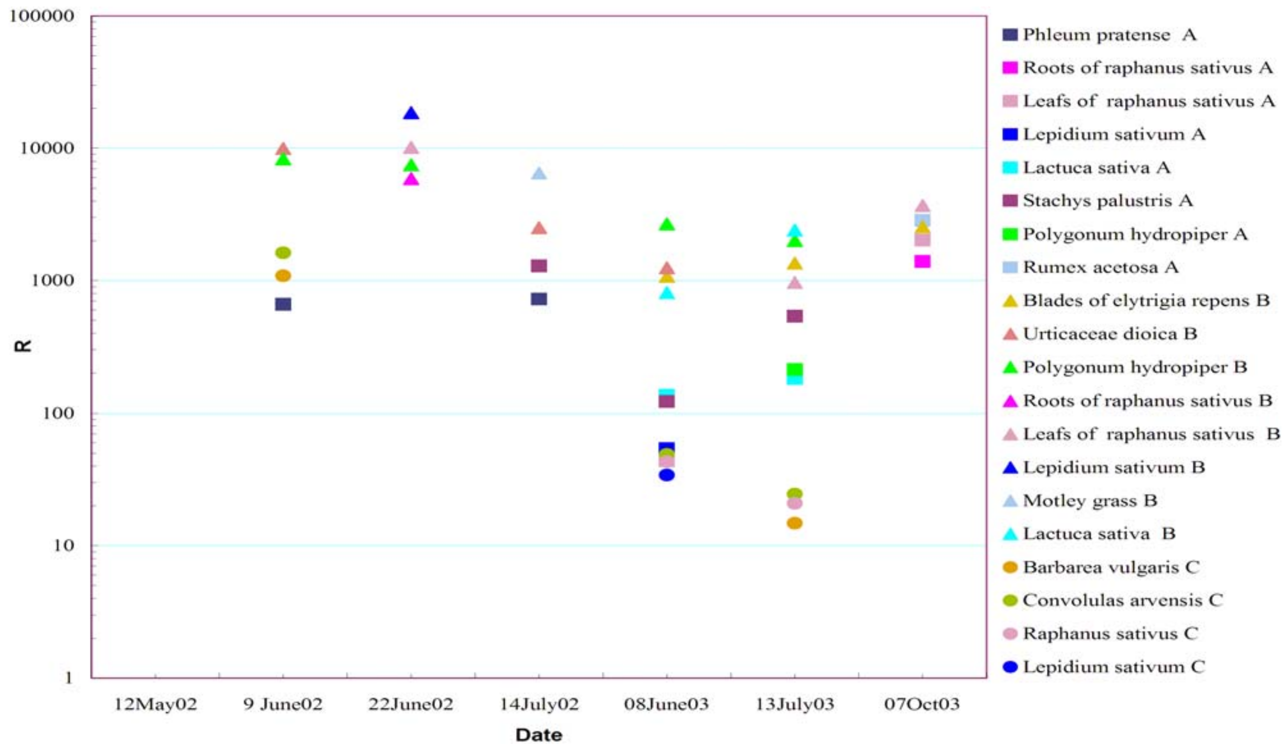

Fig. 2. Plant – soil solution Cs-137 concentration ratio **R**. Designations – see legend under Fig.1.

$R=A/A_1$, where **A** – Cs-137 concentration in the dried plant, **A_1** - Cs-137 concentration in the corresponding soil solution, are shown.

We can see from Fig.2, that represented coefficients **R** vary in range of three mathematical orders. Similar to Cs-137 content in plants, plant:soil solution Cs-137 concentration ratio depends strongly on the date of selection and on type of the soil, but it is practically invariable for the selected at the same date different plants at the site.

Ratio $(Cs\text{-}137/K)_p/(Cs\text{-}137/K)_{ss}$, where $(Cs\text{-}137/K)_p$ is value for plant and $(Cs\text{-}137/K)_{ss}$ is value for the corresponding soil solution, for all investigated plants and experimental sites is represented in Fig. 3. We can see from Fig.3 that the ratio varies in interval of more than two mathematical orders for the investigated plants and soils during two seasons. We can conclude that for our experiment plants mechanisms of plants uptake of Cs-137 and K uptake are different. So, we cannot consider K as an analog of Cs-137 in respect of plant uptake.

Let us introduce normalized by moisture content in a unit of volume of the soil plant: soil solution Cs-137 concentration ratio **c** as

$$c=A/A_1hp=R/hp, \qquad \text{(Eq. 1)}$$

where **h** - moisture of the soil at the time of selection of the samples, **p** – density of dry soil.

Density of dried soil is equal 1.12 kg/dm^3 for site A, 0.39 kg/dm^3 for site B and 1.38 kg/dm^3 for site C.

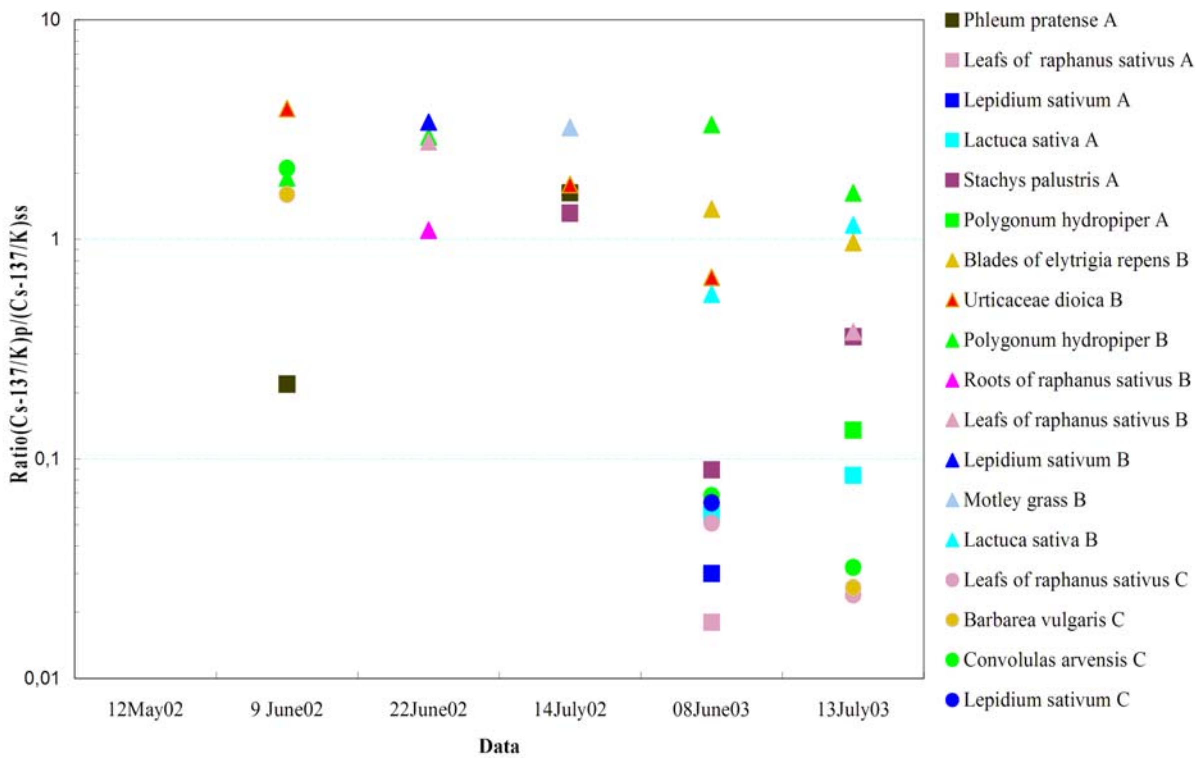

Fig. 3. Ratio $(Cs\text{-}137/K)_p/(Cs\text{-}137/K)_{ss}$. Designations – see legend under Fig. 1.

In Fig. 4 the values **c** for all our experimental sites and plants are represented.

We can see, that this value does not depend on the date of selection, on the type of the soil, on the soil moisture and on the kind of plant. Average value **c** for all our experimental field sites and plants is $\mathbf{c_0}$ = 9300 dm^3/kg. Average deviation from average value is 4300 dm^3/kg. Deviations of our experimental values **c** from average value $\mathbf{c_0}$ may be caused by inhomogeneity of Cs-137 distribution in the soil at the experimental site, by changes of soil moisture and Cs-137 concentration in soil solution during growing of the plants, by different biomass accumulation rates of different kinds of the plants and maybe by other factors. But it is obvious that the value **R** for different lands and dates of sample selection varies far more than value **c.** We can see from Fig. 3 and Fig. 4 that at our experiment range of value **c** change in 100 times lesser than range of change of value R. So, we can conclude, that the most important factor for magnitude of plant:soil solution Cs-137 concentration ratio is moisture content of the soil.

We can obtain from **(1):**

$$R=A/A_1=chp, \qquad \text{(Eq. 2)}$$

i.e. plant:soil solution Cs-137 concentration ratio for the land is roughly proportional to the soil moisture.

We can get from **(1)**:

$$A=chpA_1 \qquad \text{(Eq. 3)}$$

Let us see at (Eq. 3). The value hpA_1 is equal to quantity of dissolved in soil solution Cs-137 at a

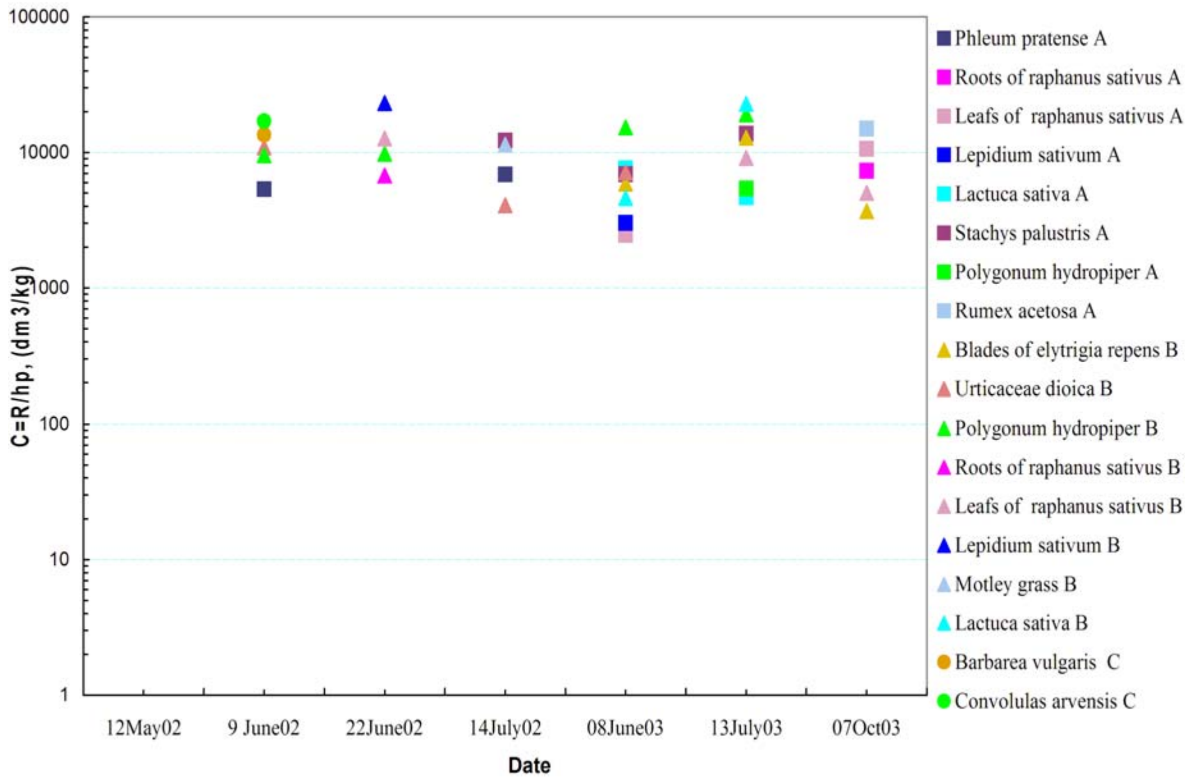

Fig. 4. Normalized by moisture content in a unit of volume of the soil plant – soil solution Cs-137 concentration ratio **c**(dm/kg). Designations – see legend under Fig. 1.

unit of volume of the soil. In other words, the value hpA_1 is the content of dissolved Cs-137 in the soil per unit of volume of the soil. The value **c** varies little. Consequently, for all investigated lands and plants Cs-137 uptake by plant depends roughly proportionally on concentration of dissolved Cs-137 in the soil.

Similar investigations must be carried out for other types of soil and plants. Unfortunately, the large majority of papers on Cs-137 plant uptake have no information about Cs-137 content in soil solution and about moisture of the soil, so we cannot use data from these papers for comparison with our formulas **(2)** and **(3)**.

Under natural conditions value **h** varies. We can see from the table that under natural conditions value A_1 varies, too. Consequently, plant Cs-137 uptake at the land depends on the time.

We investigated rapidly maturing plants. Soil humidity changes are not essential while the plants were growing. For this reason differences in Cs-137 content in selected plants at the same site at the same date plants are not big. We think that this difference would be smaller, if we can

account plant biomass accumulation rate and change of soil humidity and Cs-137 content at the soil solution during the plants growing.
For slower growing plants we consider for the land with parameters **p** and **c** that Cs-137 content **A** at the grown for the time interval $t_1 - t_2$ plant we can describe with formula:

$$\mathbf{A} = \frac{\mathbf{pc}\int_{t_2}^{t_1} \mathbf{A_1(t)U(t)h(t)dt}}{\int_{t_2}^{t_1} \mathbf{U(t)dt}}, \qquad \text{(Eq. 4)}$$

where:

A_1(t) - dependence of Cs-137 content at the soil solution on time;
U(t) - dependence of plant biomass accumulation rate on time;
h(t) - dependence of soil moisture on time.

There is Cs-137 uptake by the plant for the time interval $t_1 - t_2$ at the numerator and the plant biomass accumulation for this time at the denominator.

We have not completed a statistical treatment of our experimental data on Cs-137 sample content **A** and transition coefficients **R** for different dates of selection (average value, standard deviation and so on), because it would not be correct. These value change, not due to random experimental error, but according to formulas (Eq. 4) and (Eq. 2).

ACKNOWLEDGEMENTS

Financial support was obtained from Science and Technology Center in Ukraine (STCU project 2390).

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