

Regulation of Transfer of ^{137}Cs by Polymeric Sorbents for Grow Ecologically Sound Biomass

A. H. Tadevosyan, S. K. Mayrapetyan, N. B. Tavakalyan, K. I. Pyuskyulyan, A. H. Hovsepyan, S. N. Sergeeva

Abstract—Soil contamination with radiocesium has a long-term radiological impact due to its long physical half-life (30.1 years for ^{137}Cs and 2 years for ^{134}Cs) and its high biological availability. ^{137}Cs causes the largest concerns because of its deleterious effect on agriculture and stock farming, and, thus, human life for decades. One of the important aspects of the problem of contaminated soils remediation is understand of protective actions aimed at the reduction of biological migration of radionuclides in soil-plant system. The most effective way to bind radionuclides is the use of selective sorbents. The proposed research mainly aims to achieve control on transfer of ^{137}Cs in a system *growing media – plant* due to counter ions variation in the polymeric sorbents. As research object Japanese basil - *Perilla frutescens* was chosen. Productivity of plants depending on the presence (control-without presence of polymer) and type of polymer material, as well as content of ^{137}Cs in plant material has been determined. The character of different polymers influences on the ^{137}Cs migration in growing media – plant system as well as accumulation in the plants has been cleared up.

Keywords—Radioceaseum, Japanese basil, polymer, soil-plant system.

I. INTRODUCTION

THE largest concern on the ^{137}Cs deposition and its soil contamination due to the emission from the Fukushima Daiichi Nuclear Power Plant (NPP) showed up after a massive quake on March 11, 2011 [1]. Removal of ^{137}Cs contaminated soils or a land use limitation in areas where removal is not possible is, therefore, an urgent issue. It is demonstrated that greater than 86% of total radiocesium and 79% of total ^{131}I were absorbed in the upper 2.0cm in the soil profile [2].

A large portion of some radionuclides (RN) would remain in the root-zone soil for many years after their deposition. This means that a radioactive deposition onto soil can lead to a root uptake lasting for many consecutive years [3]. The ability to predict the consequences of an accidental release of RN

relies mainly on the level of understanding of the mechanisms involved in RN interactions with different components of agricultural and natural ecosystems [4].

One of important aspects of the problem related to contaminated soils remediation is elaboration of protective actions aimed at reduction of biological migration of RN in *water – soil – plant* system. The agricultural radioecology has acquired significant experimental information on modes for decreasing the transfer of RN from soils to plants [5]–[8].

Sorbents are used as a most effective way to bind RN. The main drawback of the available sorbents is rather low selectivity towards certain ions and the low strength of binding. It is not possible to reduce the concentration of water-soluble forms of RN to the background level. The problem of selective binding of radionuclides acquired special relevance due to the accident at the Fukushima nuclear power plant.

Recently the “Plastpolymer” Institute jointly with the Institute of Hydroponics Problems conducted research on application of high-water-expending polymers for plant production under hydroponic and soil conditions. The research team has developed new means for remediation of RN-contaminated soils through regulation of biological migration of anthropogenic RN particularly, ^{137}Cs and ^{90}Sr by adding water-retaining polymers to the ecosystems “water–soil– plant” and “water– nutrient solution–plant” in the zones of radio-ecological tension [9], [10].

Of special interest are compositional water-swellaable polymers with inorganic filler. Addition of different type filling materials and variation of their concentration will allow achievement of the desired balance of physical and chemical properties in the final composite material.

The aim of this research is to provide new procedures and development of technology for remediation of Cs-contaminated soils with low contamination level through regulation of biological migration of ^{137}Cs by polymeric compositions in the *soil – plant* systems.

This article proposes to achieve to the control of migration of radiocesium from soil to plant due to fillers in the polymeric compositions.

II. MATERIALS AND METHODS

The tests are being carried out under soil conditions with and without application of polymers in plant root-inhabited zone (PRIZ) in more radioecological tension zone (with the radius of 7km of Armenian NPP, v. Taronik, the Ararat Valley). For the investigations, Japanese basil – *Perilla*

A. H. Tadevosyan is with the Institute of Hydroponics Problems of the National Academy of Sciences, Yerevan 0082, Armenia (phone: +374-10-563015; fax: +374-10-565590; e-mail: anntadevosyan@yahoo.com).

S. K. Mayrapetyan is with the Institute of Hydroponics Problems of the National Academy of Sciences, Yerevan 0082, Armenia (phone: +374-10-565162; fax: +374-10-565590; e-mail: hydrop@netsys.am).

N. B. Tavakalyan and S. N. Sergeeva are with the Scientific Research Institute Erplastpolymer, Yerevan 0007, Armenia (phone: +374-10-481920; e-mail: polyfa@yandex.ru, sybnina@yandex.ru).

K. I. Pyuskyulyan is with the Armenian Nuclear Power Plant, Metsamor 0910, Armenia (phone: +374-10-255880; fax: +374-10-280669; e-mail: pyuskyulyan@bk.ru).

A. H. Hovsepyan is with the Institute of Hydroponics Problems of the National Academy of Sciences, Yerevan 0082, Armenia (phone: +374-10-562805; fax: +374-10-565590; e-mail: hydrop@netsys.am).

frutescens was chosen. Japanese basil or Shi-so is herb of Japanese extraction and commonly cultivated as a vegetable in Japan. The green leaves can be served as tempura, in salads and with sashimi. It is also used to flavor sauces, added to ponzu and daidai, used in Japanese-style spaghetti dishes. The leaves can be soaked in soya sauce for a year and wrapped around rice to make a tasty rice ball.

The objectives of investigations are: determination of both quantitative and qualitative productivity of plants depending on the presence (control-without polymer), type of polymer material, water regimen; determination of ^{137}Cs content in a system *soil – plant*; determination of migration and accumulation of ^{137}Cs in soil layers in different depth (0-5; 5-10; 10-20), below- and above-ground parts of plants depending on application of the polymers; revealing the dependence of redistribution of ^{137}Cs in systems *soil – plant* on the type of a polymer material.

The plants were planted with a density of 20plant/m² in the experimental field. Different polymer compositions (1g/plant) were applied in plants' root-inhabited zone (PRIZ) during planting of seedlings.

The polymer compositions with bentonite (Sample 12 and 13) and zeolite (Sample 4) were synthesized in the Scientific Research Institute Erplaspolymer.

The experiments were carried out with the following variants:

1. Control - without polymer, irrigating frequency (IF) once three days;
2. PRIZ + Sample 13, IF once four days;
3. PRIZ + Sample 12, IF once four days;
4. PRIZ + Sample 4, IF once four days.

Content of ^{137}Cs was determined by Low-background gamma spectrometer with pure Ge detector (Canberra production) and supporting "Linx" and "GENIE-2000" software (for gamma spectrometric measurements).

The contents of vitamin C and A in fresh leaves were determined by titration method [11]. The water concentration in the leaves was determined by the refractometric method [12]. The content of essential oil in dry leaves was determined by steam distillation method [13].

A statistical analysis of collected data was carried out using GraphPad5 software program.

III. RESULTS AND DISCUSSION

The results showed (Table I) that adding Sample 13, 12 and 4 polymers in PRIZ exceeded dry yield of Japanese basil leaves 1.7-2.0 times compared to the control (without polymer) at the same time saving irrigating water about 25 %. Roots of control variants by dry mass conceded all polymer variants by 15-20%.

In the above-ground mass, independently of polymer sort and use, the relation of leaves and stems was not changed significantly: it was about 2/1 (Fig. 1).

TABLE I
INDICES OF JAPANESE BASIL PRODUCTIVITY (DRY MASS, G/PLANT)

Variants	Whole biomass	Above-ground part	Leaves	Stems	Roots
Control- without polymer	59.1	50.9	32.1 ^a	18.8	8.2
PRIZ + Sample 13	91.8	82.4	53.7 ^b	28.7	9.4
PRIZ + Sample 12	105.4	95.7	63.7 ^b	32	9.7
PRIZ + Sample 4	96.8	87.4	58.9 ^b	28.5	9.4

^{ab} Tukey's Multiple Comparison Test (P<0.05)

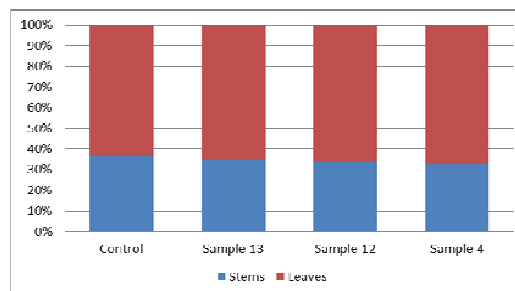


Fig. 1 Ratio of leaves and stems in the above-ground biomass of Japanese basil

The results of biochemical analysis had shown significant increases (1.2-1.4 times) only of β -carotene content in all variants with applying polymers. However, control plants by essential oil content in dry leaves exceeded polymer variants by 30-50% (Table II).

TABLE II
QUALITY INDICES OF JAPANESE BASIL DEPENDING ON THE USE OF POLYMERIC COMPOSITIONS

Variants	Essential oil content in dry leaves, %	Vitamin C content in fresh leaves, mg%	β -carotene content in fresh leaves, mg%
Control-without polymer	0.80±0.07	86.8±6.2	8.8±0.7
PRIZ + Sample 13	0.63±0.05	87.5±7.0	11.0±0.9
PRIZ + Sample 12	0.54±0.04	92.6±8.8	12.6±1.0
PRIZ + Sample 4	0.54±0.05	96.3±8.9	10.4±0.8

Some physiological indices of Japanese basil leaves were introduced in Table III.

Use of polymer (with the decreasing of irrigating water by 25%) increased the content of bound water in basil leaves about 1.2-1.6 times and colloid bound water 1.6-2.2 times (Table III).

TABLE III
PHYSIOLOGICAL INDICES OF JAPANESE BASIL DEPENDING ON THE USE OF
POLYMERIC COMPOSITIONS

Indices	Control-without polymer	Sample 13	Sample 12	Sample 4
Total water content, %	72.1	72.4	70.9	72.6
Free water content, %	46.0	39.8	29.6	37.7
Bound water content, %	26.1	32.6	41.3	34.9
Free and bound water ratio	1.76	1.22	0.72	1.08
Osmotic pressure of cell sap, atm.	7.02	6.90	8.11	6.65
Osmotic bound water, %	15.1	14.6	17.2	14.4
Colloid bound water, %	11.0	18.0	24.1	20.5

During the vegetation period, as a result of soil irrigation, ^{137}Cs migrated from the upper (0-5cm) layers to the down (5-10 and 10-20cm). It was more intensive in Sample 13 polymer variant (Table IV). Thus, in September compared with June (before planting ^{137}Cs concentration was: 0-5=29.4±2.2, 5-10=6.1±0.4 and 10-20=5.8±0.4), ^{137}Cs content was reduced in the upper 0-5cm layer 1.6 times in control, 1.4-1.5 times in Sample 12 and 4, and 2.0 times in Sample 13 variants. On the contrary, in September ^{137}Cs content increase is observed in 5-10 and 10-20cm layers (in control, in Sample 12 and 4 variants – 3.3-3.9, in Sample 13 polymer variant - 3.0-3.1 times).

TABLE IV
CONTENT OF ^{137}Cs IN THE GREY CARBONATE IRRIGABLE SOILS IN THE
VICINITY OF THE ARMENIAN NPP AFTER BASIL HARVEST (SEPTEMBER)

Variants	Depth of soil layer, cm	Bq/kg
Control-without polymer	0-5	18±4.0
	5-10	24±5.3
	10-20	21±4.6
PRIZ + Sample 13	0-5	15±3.3
	5-10	18±4.0
	10-20	18±4.0
PRIZ + Sample 12	0-5	19±4.6
	5-10	20±4.8
	10-20	19±4.6
PRIZ + Sample 4	0-5	21±5.0
	5-10	23±5.5
	10-20	21±5.0

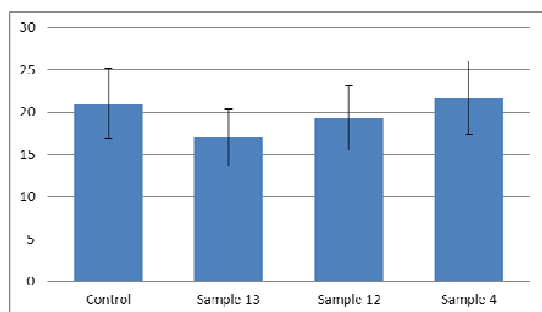


Fig. 2 Concentration of ^{137}Cs in 0-20cm soil layer at the end of vegetation period (Bq/kg, cover crop-Japanese basil)

By ^{137}Cs content in soil (0-20cm) there was no significant difference between control and polymer variants at the end of vegetation period (Fig. 2).

It turned out (Table V) that basil organs according to ^{137}Cs content have the following trend: leaves>roots>stems in control and roots>leaves>stems in all polymer variants. In control ^{137}Cs concentration exceeded polymer variants: in leaves 2.1-2.3, in stems – 1.2-3.1 times. At the same time roots of control variant distinguished with the minimum concentration of ^{137}Cs and conceded polymer variants 1.3-4.0 times. It means that all polymer compositions prevented ^{137}Cs migration from roots to stems and leaves.

TABLE V
CONTENT OF ^{137}Cs IN DIFFERENT ORGANS OF JAPANESE BASIL, BQ/KG

Variants	Leaves	Stems	Roots
Control-without polymer	2.71±0.27	0.94±0.16	1.08±0.27
PRIZ + Sample 13	1.29±0.26	0.30±0.14	1.41±0.25
PRIZ + Sample 12	1.25±0.4	0.50±0.2	3.29±0.6
PRIZ + Sample 4	1.16±0.5	0.79±0.3	4.31±0.8

It is known that the migration of RN from soil into plants depends on a number of factors: from the quantity of atmospheric precipitations, from biological peculiarities of culture, from agrochemical peculiarities of soil (humus, hydrolytic acidity, K and Ca content in soil).

^{137}Cs Accumulation Coefficient (AC, RN content in plant, Bq/kg ÷ RN content in soil Bq/kg) in different organs of Japanese basil are introduced in Table VI.

TABLE VI
ACCUMULATION COEFFICIENTS OF ^{137}Cs IN DIFFERENT ORGANS OF
JAPANESE BASIL

Variants	Leaves	Stems	Roots
Control-without polymer	0.13	0.04	0.05
PRIZ + Sample 13	0.08	0.02	0.08
PRIZ + Sample 12	0.06	0.03	0.17
PRIZ + Sample 4	0.05	0.04	0.20

It found out that AC of ^{137}Cs in roots for all polymer variants exceeded AC of ^{137}Cs in stems 4.0-5.7, in leaves – 2.8-4.0 times (for Sample 12 and 4 variants). On the contrary, AC of ^{137}Cs in leaves of control exceeded AC of ^{137}Cs in roots and stems 2.6 and 3.2 times, accordingly.

The data of Table VI showed that AC of ^{137}Cs in leaves of control exceeded all polymer variants 1.6-2.6 times, but AC of ^{137}Cs in roots conceded all polymer variants 1.6-4.0 times. One could conclude that use of polymer compositions in PRIZ can prevent transfer of ^{137}Cs from roots to leaves.

IV. CONCLUSION

- The existence of polymer compositions in PRIZ gave an opportunity to decrease the expense of irrigating water in about 25% at the same time provided high yield of Japanese basil leaves (1.7-2.0 times) compared to the control.
- Applying of polymer compositions in PRIZ promoted the decrease of ^{137}Cs concentration in basil leaves 2.1 -2.3 times compared to the control.

- During the vegetation period ^{137}Cs migration with soil vertical slit (from upper 0-5 cm to down 5-10 and 10-20 cm soil layers) was more intensive in Sample 13 polymer variant than in control.
- Despite usage of polymer compositions didn't promote significantly decreasing of ^{137}Cs concentration in soil, but they give an opportunity to grow more ecologically sound biomass for food.

ACKNOWLEDGMENT

The researches were implemented in the context of project A-2072 ISTC.

REFERENCES

- [1] J. Koarashi, M. Atarashi-Andoh, T. Matsunaga, T. Sato, S. Nagao, H. Nagai, "Factors affecting vertical distribution of Fukushima accident-derived radiocesium in soil under different land-use conditions", *Sci Total Environ.*, 2012, 431, pp. 392-401.
- [2] H. Kato, Y. Onda, M. Teramage, "Depth distribution of ^{137}Cs , ^{134}Cs , and ^{131}I in soil profile after Fukushima Dai-ichi Nuclear Power Plant Accident", *Journal of Environmental Radioactivity*, 2011, <http://dx.doi.org/10.1016/j.jenvrad.2011.10.003>.
- [3] Y. H. Choi, K. M. Lim, I. Jun, D. K. Keum, M. H. Han, "Time-Dependent transfer of ^{54}Mn , ^{60}Co , ^{85}Sr and ^{137}Cs from a Sandy Soybean Plants", *Nucl Sci Technol*, 2011, 1, pp. 392-395.
- [4] C. Tamponnet, A. Martin-Garin, M. A. Gonze, N. Parekh, R. Vallejo, T. Sauras-Yera, J. Casadesus, C. Plassard, S. Staunton, M. Norden, R. Avila, G. Shaw, "An overview of BORIS: Bioavailability of Radionuclides in Soils", *J Environ Radioact.*, 2008, vol. 99, no. 5, pp. 820-830.
- [5] A. A. Bulgakov, "Simulation of fixation ^{137}Cs in soils," *Soil Science*, 2009, vol. 6, pp. 726-732.
- [6] A. V. Panov, S. V. Fesenko, R. M. Alexakhin, A. D. Pasternak, P. V. Prudnikov, N. I. Sanzharova *et al.*, "The radioecological situation in the agricultural sphere in the contaminated regions of Russia during the long term after the Chernobyl accident," *Radiation Biology, Radioecology*, 2007, vol. 47, no. 4, pp. 423-434.
- [7] A. G. Podolyak, I. M. Bogdevich, V. Yu. Ageyets, S. F. Timofeyev, "Radiological estimation of the protective measures used in agriculture Republic of Belarus in 2000-2005 (20 years after the accident on the ChNPP)," *Radiation Biology, Radioecology*, 2007, vol. 47, no. 3, pp. 356-370.
- [8] H. Shimura, K. Itoh, A. Sugiyama, S. Ichijo, M. Ichijo, *et al.* "Absorption of Radionuclides from the Fukushima Nuclear Accident by a Novel Algal Strain", *PLoS ONE*, 2012, 7(9): e44200. doi:10.1371/journal.pone.0044200.
- [9] A. H. Tadevosyan, M. P. Schellenberg, S. Kh. Mayrapetyan, L. M. Ghalachyan, "Opportunity of Remediation of Radionuclide-Contaminated Soils and Growing Ecologically pure Plant Material via Water-Retaining Polymer" In Book: *Advanced Bioactive Compounds Countering the Effects of Radiological, Chemical and Biological Agents* (Eds: Pierce, G.N.; Mizin, V.I.; Omelchenko, A.), Springer, Dordrecht, 2013, p. 278-290.
- [10] A. H. Tadevosyan, S. K. Mayrapetyan, M. P. Schellenberg, L. M. Ghalachyan, A. H. Hovsepyan, K. S. Mayrapetyan, "Migration and Accumulation of Artificial Radionuclides in The System Water-Soil-Plants Depending on Polymers Applying", *World Acad Sci Eng and Technol*, 2011, vol. 78, pp. 656-660.
- [11] A. I. Ermakov, V. V. Arasimovich, M. I. Smirnova-Ikonikova, I. K. Murri, *The biochemical experimental method of plants*, Moscow: 1952.
- [12] N.A. Gusev, *The experimental methods water cycle of the plants*, Kasan Institute issue, Kasan, 1989.
- [13] A.S. Ginsberg, "Reductive method for determination the quantity of essential oil in the volatile-oil-bearing plants", *Chemical Pharm Industr*, 1932, no. 8-9, pp. 326-329.