Projects Summary Reports

ISTC/STCU Technical Review Committee Meeting of Fukushima Initiative 'On the environmental assessment for long term monitoring and remediation in and around Fukushima

5-6 November 2015

Hitotsubashi University Hall

Tokyo, Japan

I S T C

М Н Т Ц

SCIENCE &TECHNOLOGY
CENTER IN UKRAINE

Contents

Preface

Overview of the ISTC/STCU Fukushima Initiative 'on the environmental assessment for long term monitoring and remediation in and around Fukushima'

ISTC/STCU Secretariat

Tohoku Earthquake of 2011

On March 11 of 2011, an earthquake measuring 9.0 (Mw) occurred off the Pacific coast of Tohoku, Japan. This was the most powerful earthquake to hit Japan in its recorded history and is the fourth largest recorded earthquake on world record. In addition to in-land damage caused by the seismic activity, including knocking out the local electrical grid, the earthquake triggered a powerful tsunami that swamped the back-up electrical supply to the emergency power systems at the Fukushima Daiichi Nuclear Power Plant. This chain of events caused a severe nuclear accident with substantial damage to three of the reactors. The resulting release of radionuclides and the severe environmental contamination resulted in the immediate evacuation of the surrounding populations and the creation of the Fukushima exclusion zone surrounding the plant.

ISTC/STCU Response

Taking into account the severity of this nuclear event and the need for practical solutions to address the environmental impact, the International Science and Technology Center (Moscow, Russia – Astana, Kazakhstan) and the Science and Technology Center in Ukraine (ISTC/STCU) formed the Fukushima Initiative through the direct support and encouragement of the Japanese government along with the state parties to the ISTC.

Drawing on almost 20 years of research knowledge in their fields of operation, ISTC/STCU were well placed to provide certain support to the Japanese authorities related to cumulative world experience in remediation and monitoring. Beginning from the middle of 2011, collaboration began with the Japanese authorities to provide expertise and consulting on issues of severe nuclear accident mitigation. In December 2011 the ISTC/STCU Initiative 'on the environmental assessment for long term monitoring and remediation in and around Fukushima' was created.

The November 5-6, 2015 meeting of the ISTC/STCU Technical Review Committee marks the culmination of the almost three year project/research phase of the Fukushima Initiative.

Chronology

The primary events that have been taken in support of Japan by the ISTC/STCU through the Fukushima Initiative are outlined below:

- June 2011 – At the ISTC Governing Board (GB) meeting #53, the decision was taken on a formal project review related to severe accidents management, decommissioning and environmental remediation to understand the implications and benefits of past projects experience in light of Japanese issues in coping with the aftermath of the accident and its long terms ramifications.
    - The initial program was designed to facilitate exchange with Japanese experts in areas of specific technical relevance. Japanese authorities identified 22 project of interest in the areas of decontamination, remediation, and rehabilitation and 18 projects in severe accident response for additional information.
- October 11-12, 2011 – A meeting took place in Moscow, Russia at the ISTC of the 'Contact Expert Group on Severe Accidents Management' with in-depth expert discussion on the situation at Fukushima. 16 projects were selected for formal review as well as discussion was undertaken related to organizing two international symposiums to be conducted in Japan in early 2012.
- December 1-2, 2011 and December 5-6, 2011– Two workshops were respectively held in Moscow in the areas of severe accident response and decontamination with the participation of Japanese and regional experts to address immediate technical questions related to the Fukushima situation.
- December 8-9, 2011 – ISTC formed the 'Fukushima Initiative' in December 2011 at ISTC GB meeting #53 in support of the Japanese party and the Japanese people. Official coordination with the STCU on a broader initiative was established.
- February 3-4, 2012 - ISTC/ STCU Symposium and Seminar: The Experience and Technology of Russia, Ukraine, and Other CIS Countries on Remediation and Restoration of Environments were conducted in Tokyo and then Fukushima City.
    - Review was undertaken of seven selected projects related to land and soil remediation in the Fukushima area.
    - Two formal reports in English and Japanese language are published.

- March 8-10, 2012 – International Science Symposium on combating radionuclide contamination in the agro-soil environment was conducted in Koriyama, Japan.
    - Reviews of a further 11 projects/technologies were made.
    - One formal report in the English and Japanese languages was published
- April 12-13, 2012 - ISTC/STCU held a project results review meeting of 17 remediation projects in support of 'Environmental Remediation after the Fukushima Accident/ II' in Moscow, Russia at the ISTC.
- June 1, 2012 - ISTC/STCU issued a joint 'Call for Proposals for projects related to land decontamination and monitoring in view of the Fukushima Nuclear Power Plant Accident'.
    - August 2012 - 53 short proposals were collected and subjected to independent review.
    - 11 short proposals were invited to provide full submissions for review and funding consideration in Dec 2012.
- December 11-12, 2012 – The 'ISTC/STCU Technical Working Group Meeting: on the environmental assessment for long term monitoring and remediation in and around Fukushima' was conducted in Tokyo, Japan including review of 11 proposals.
    - ISTC/STCU Technical Review Committee for monitoring and review of the research effort was formed,
    - Six projects were selected for funding in January 2013:

Studies of secondary migration of radionuclides and waste treatment

1. STCU 5954 – Sept. 2013-Aug. 2015) - Monitoring of radioactive pollution of forest ecosystems after accident on ChNPP
2. STCU 5953 – Sept. 2013-Aug. 2015 - Methodology for long - term radiation monitoring to dose assessment using radiological zoning and modeling of radionuclides migration in environmental and food chains
3. STCU 5952 – Sept. 2013-Aug. 2014 (1yr) - Compaction of radioactive waste produced by decontamination of territories polluted due to the accident at Fukushima Daiichi Nuclear Power Station
4. ISTC A-2071 – Jul. 2013-Jun. 2015 - Advanced polymeric systems providing deactivation of different Surfaces and soil from radioactive pollutions with the use of polymers

Studies to minimize transfer of radionuclides in soil-plant system

5. ISTC K-2085 – Sept. 2013-Aug. 2015 - Development of a set of measures for production of assured quality agricultural goods under radioactive contamination conditions
6. ISTC A-2072 – Jun. 2013-May 2015 - Project Remediation of Cs-contaminated soils through regulation 134Cs and 137Cs soil-plant transfer by polymeric sorbents

- April 14-17, 2014 - ISTC/STCU Technical Review Committee project mid-term review meetings are held including a visit to the Fukushima exclusion zone to survey local activities of the Japanese authorities.
    - April 14, 2014 – Visits were made to the following locations in the Fukusuhima area: Agricultural Radiation Research Center, Fukushima Branch and Tohoku Agriculture Research Center, National Agriculture and Food Research Organization (NARO); National Forest at Litate village (forest studies); and Kawamata town geographical area (4 separate sites).
    - April 15-17, 2014 – Technical Review of the six funded projects took place in Tokyo, Japan including critical recommendations to maximize the applicability of the work in the Fukushima context.
- November 5-6, 2015 – An ISTC/STCU Technical Review Committee review is conducted following fulfillment of the first phase of the Fukushima Initiative.

Contributing Parties

Many organizations have contributed to the ISTC/STCU Fukushima Initiative. The below list is a general reference of the principals involved from the initiative's inception and is not exhaustive.

Organizers

- ISTC - International Science and Technology Center
- STCU - Science and Technology Center in Ukraine

Sponsors

- Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT)
- United State Department of Energy (US DOE)
- European Commission Directorate-General for International Cooperation and Development (DEVCO)
- Ministry of Foreign Affairs of Japan (MOFA)

Technical Contributors

- Japan Atomic Energy Agency (JAEA)
- International Atomic Energy Association (IAEA)
- Ministry of the Environment of Japan (MOE)
- Ministry of Agriculture, Forestry, and Fishery of Japan (MAFF)
- Nuclear Safety Research Association (NSRA)

Among many others

Session 1

Waste Interim Storage and Waste Volume Reduction

Organic Waste Volume Reduction with Incineration

STCU-5952

Compaction of radioactive waste produced by decontamination of territories polluted due to the accident at Fukushima Daiichi Nuclear Power Station

Dr. Volodymyr Tokarevskyy
Institute of Chernobyl Problems (Ukraine)

ISTC/STCU Fukushima Expert Meeting

Tokyo, Japan
November 5-6, 2015

Summary Report – project #5952

on work performed from *date: September 1, 2013* to *date: August 31, 2014*

Name of the Leading Institute:
Institute for Chernobyl Problems of the Chernobyl Union of Ukraine

Project Manager:	**Name**	**Tokarevsky V.V.**
	Scientific degree	**Professor, doctor PhD**

Title of the Project:	Volume Reduction of Radioactive Waste Produced by Decontamination of Territories Polluted due to the Accident at Fukushima Daiichi Nuclear Power Station
Commencement Date:	September 1, 2013
Duration:	12 months
Project Manager	Tokarevsky V.V.
phone number:	+380506812407
e-mail address:	tokarevsky@voliacable.com
Leading Institute:	<Name> Institute for Chernobyl Problems <Director> Tokarevsky V.V. <Address> Prospect Nauki, 46, Kiev, Ukraine <Phone> +380445240478 <Email> tokarevsky@voliacable.com <URL> if exist
Participating Institutes:	<Name> EKSIS Ltd, Ukraine <Director> Rovensky A. I. <Address> 61040, 43, Skorostnaia str, Kharkiv, Ukraine <Phone> +380577556901 <Email> guandr@mail.ru <URL> if exist
Foreign Collaborators:	<Institute> Pacific Northwest National Laboratory <Name> Yasuo Onishi <Address> P.O. Box 999, Richland, WA 99352, USA <Phone> +15093757203 <Email> Yasuo.Onishi@pnnl.gov <URL> if exist

1 Brief description of work: objectives, and expected results

After the accident on the NPP Fukushima Daiichi a very large volume of radioactive contaminated materials was formed during the time. First of all, the nuclear fuel, and radioactive damaged spent nuclear fuel or partly melted in the core of reactor after release of products due to the loss of hermetic encapsulation into the atmosphere. At following time after the next hydrogen explosions has happened the reactor building disruption and radioactive contamination of environment of the NPP site (on-site) and over than their border (off-site). On the NPP site the ocean water was used for cool the spent nuclear fuel and was formed the very large volume of solid radioactive waste. In a whole the range of radioactive contamination in the dismantled reactor buildings and in a close vicinity of reactor was be higher the level acceptable. Repair and construction works generated a big volume of the designated waste. Beside a big number of buildings and equipments destroyed by tsunami were contaminated with the radioactivity.

The main objects that are subject to remediation and rehabilitation after the accident at the Fukushima Daiichi are contaminated buildings, roads, forests, arable land and soil. Interim storage of radioactive materials removed and the subsequent disposal of the expected volume of radioactive waste will need significant current and capital expenditures. The aim of the project is to develop a feasibility study of the possibilities of using technology reduce the volume of radioactive waste from / generated during decontamination of areas contaminated after the accident at Fukushima Daiichi, based on the experience of the treatment of radioactive waste of Chernobyl origin.

The principal goal of the Project is the achievement in reduction of radioactive waste (RW) amount. The amount of RW, i.e. the weight and volume of waste, during the remediation time has requirement to decrease the total expansion of the *sum amount* on the whole process of collection waste and for the disposal of waste, is achieved by three ways.

The first way consists in collection of *only* radioactive contaminated materials, where segregation is achieved on the level of the weight concentration of the main radionuclides. This level is streamed from the value of clearance which is defined as the safe level for all population.

The second way is connected with the amount of RW and has influence on the additional cost for construction, operation and close up of storage which depends directly on the amount of waste. The time of temporary storage is defined by the radiation characteristics of waste.

The third way consists in the further *reduction* of RW amount after temporary storage, which demands of additional cost. It is defined by set of technological processes for treatment of waste and obtains in result the requirements of criteria acceptance for disposal (lack of fires, explosions and different dangers) for the level of exemption of given type of radionuclide.

2 Modifications of work after April 2014 meeting

The project considers only radioactively contaminated materials (RCM) and radioactive waste (RW), which are localized in the territory, which is located outside the industrial area Fukushima Daiichi (off-site). Information provides an assessment of waste parts in special decontamination area (area countermeasures). It has dependences on the weight in ton after cleaning of buildings and after tsunami destruction. All data shows the distribution of the assigned (precisely defined) waste after cleaning of buildings descent in 12 prefectures of Japan (Iwate, Miyagi, Yamagata, Fukushima, Ibaraki, Tochigi, Gumma, Chiba, Tokyo, Kanagawa, Niigata , Shizuoka). This information does not give a complete picture of the number of radioactively contaminated material that will appear of object processing in the Project (soil, combustible materials). Therefore, we present several forecast estimates of numbers. So, includes information to calculate the amount of waste from decontamination, and combustible number and non-combustible waste in countermeasures and their specific activity and other parameters.

From the above brief analysis, we can conclude that:

1. Contribution of amount of soil and combustible materials in the total amount of radioactively contaminated materials and radioactive waste is decisive;
2. Purification of soil and returning large part of them to re-use will significantly reduce the cost of storage in storage sites;
3. Burning of combustible materials is, firstly, the requirements of combined wet technology regulations, and secondly, will also minimize the costs of storage and disposal of radioactive waste later on.
4. Special attachment must be regarded to the purification of cesium release in atmosphere.

The current practice of decontamination activities and subsequent treatment of radioactively contaminated materials and radioactive waste is schematically shown in Fig. 1. It should be noted that the actions in the aftermath of the accident (AA) at Fukushima Daiichi compares favorably to the action by AA of Chernobyl disaster. The main differences are the following:

1. The organization of high culture and decontamination measures.
2. Rapidly and efficiently develop legal and regulations that govern all stages of the work, namely the «Law on Special Measures» and «Guidelines relating to waste» , which includes the following sections:

- Guidance on the methodology to determine the level of contamination,
- Guidance associated with specific household waste and industrial waste,
- Guidance associated with the designated waste,
- Guidance of waste associated with waste of deactivation,
- Guidance on the methodology of measurement of radioactivity,
- Guide for specific wastes.

3. Allocate an attention and funding on development of new methods and technologies for decontamination area and minimize the RE and RW whose purpose was:

- checking the availability and effectiveness tested and new methods;
- study of the value, creation period, the labor force;
- estimate the amount of secondary waste generated, as well as exposure to radiation of workers;
- the creation of a full cycle of waste management including reducing their volume and treatment of secondary waste;
- radiation protection of workers;
- establishment of optimal radiation control.

4. Allocate special attention to the collection and analysis of quantitative and qualitative characteristics of radioactively contaminated materials and radioactive waste and creating a unified information-analytical system for radioactive waste management. Treatment of radioactive contaminated materials and radioactive waste can be divided into five stages:

1. Collection of radioactively contaminated materials and radioactive waste.
2. Transportation to temporary storage facility.
3. Temporary storage.
4. Transportation of radioactive waste to an intermediate storage facility.
5. Intermediate storage.

In the *first stage* performed: radiation exploration and determination of vertical migration of radionuclides in order to minimize the number of radioactively contaminated materials; collection of the type of materials to large flexible containers and labeling containers (using either strong identifiers or electronic chips). These rules allow the collection to further preserve the history of all radioactive - contaminated materials and waste management to account for radioactive waste. Radioactively contaminated material to the level of specific activity below 8 kBq/kg but above the threshold, which is set legal acts for this type of terrain is subject treated as household waste. It should be noted that all schemes of treatment of various wastes indicated that residues of specific activity levels below 8 kBq/kg to be treated as household waste. But this is wrong. The lower threshold of depends on the specific activity of gathering places radioactive material that, in our opinion, is performed in Japan.

The *second stage* is formed by the route of transportation. The vehicle is provided radiation monitoring and radiation protection system driver. Itinerary tracked automatically, which ensures a quality system for transportation.

At the *third stage* the unloading of the vehicle in temporary storage constructed for different types of material.

The *fourth stage* radiation monitoring is performed when loading the waste into the truck. Transportation is done the same way as in the second stage.

At the *fifth stage* on constructed an intermediate storage facility (ISF), which will be equipped with well-developed infrastructure with the ability to: radioactive - contaminated materials' segregation and waste management; processing; radiation monitoring and environmental

monitoring of the environment; conducting research and laboratory tests; input register of radioactive material at all stages of the input control to an interim storage and inventory of radioactive waste; public awareness.

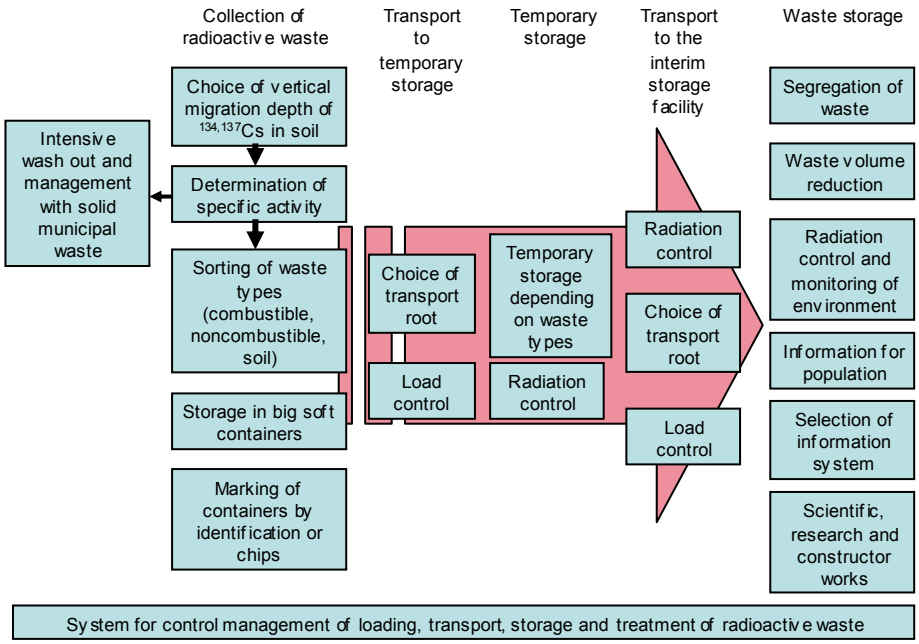


Fig. 1. Scheme of current practice decontamination area and the subsequent management of radioactively contaminated materials and radioactive waste treatment.

3 Technical approach, method, experiments, theory, etc

Based on the information provided in the previous sections, we propose the following scheme for radioactive contaminated materials and radioactive waste that will be sent to intermediate storage facility (ISF). Please note that the project is considered only for radioactive contaminated soil and combustible materials (Fig. 2).

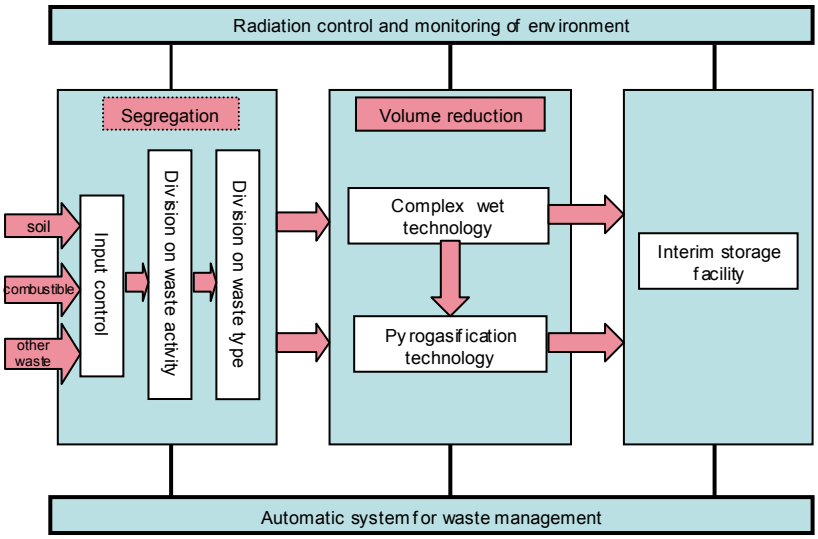


Fig. 2. Scheme for management of radioactive contaminated soil and combustible materials at ISF.

Recall that the intermediate storage facility (ISF) consisting of objects with different functions as storage, facility for segregation, facility for radioactive waste compaction, equipment around the clock monitoring, Centre for Research and experimental development and Centre for Public Information.
Based on the practice of decontamination activities and management of radioactive waste, as described above, on the subject of segregation should be placed several technologies that carry out the separation of radioactive waste. First of all, a complex of separating radioactive waste specific activity that will separate the waste into two streams: 8 ÷ 100 kBq/kg and > 100 kBq/kg. The basic principles underlying the concept of the complex are as follows:
1. Maximum mechanization and automation of processes to reduce exposure doses;
2. Saving the entire history of the origin and treatment of radioactive waste, as well as all the information necessary for keeping the register of radioactive waste and storages of radioactive waste cadastre.
These principles are achieved through the use of remote-controlled mechanisms, and mechanisms that have the ability to control in automatic mode with feedback.
In addition, for each technology processing of radioactive waste presented their claims to the homogeneity of the composition of the material processed. So pyrolysis - gasification technology is not critical to the composition and combination wet technology does not allow the soil composed of processed including organic fine particles of concrete and metal products. Therefore, it is necessary to expose the soil mechanical fractionation, eg by vibro -separation to separate the mixture into its constituent components, but after grinding.

3.1. Combined wet technology.

The method combined decontamination of soils developed and tested in the Chernobyl exclusion zone to solve the problem of decontamination of radioactively contaminated soil in order to reduce its volume before disposal. The difficulty of solving this problem is due, above all, a high degree of "dilution" of radionuclides in the contaminated soil. Thus, the contents of the main short-lived radionuclides (^{137}Cs, ^{90}Sr,) throughout the volume of soil in the temporary location of radioactive waste (> 700,000 m^3), is only about two kilograms. This fact leads to the solution of technical features of the soil decontamination. It is obvious that method of decontamination should include processing all the soil and concentration of radionuclides in a small volume of waste generated.
The technical solution is the basis of the proposed method, based on the uneven distribution of radionuclides in soil granulometric fractions.
Such character of distribution is due on the one hand, the developed surface of fine soil fractions and on the other - a relatively high sorption capacity of minerals that are part of such fractions. Therefore, as indicated, for example, separation from the main mass soil low by weight and volume of fine fractions allows for decontamination of soils by ~ 60%. Fractionation of dispersed particle size (classification) can be made as a result of dry sieving (dry classification) or by suspending the previous material in a liquid, usually water, followed by separation and drying of a suspension of fine fractions, which according to law Stokes sediment at a slower rate

than coarse particles dispersed fractions (hydroclassification). In most cases, the fractionation of soil-sized particles carried by his hydro classification. Moreover, hydroclassification prior separation of large mechanical impurities, included stones, elements of concrete and metal structures. This preliminary fractionation of soil carried out by, respectively, screening and magnetic separation. Soil decontamination method was tested in the laboratory. It has been shown that the combination of leaching and hydraulic classification can be achieved by a significant effect of decontamination. Thus the radioactive impurities are concentrated in a small volume (5-10% of the soil). In the laboratory experiments with samples of radioactively contaminated soils were tested, sampled in the area near Chernobyl.

Deactivating effect, resulting in a technological experiment is of the order of 80% and is not inferior to the results obtained in the laboratory. Given that the minimum ratio of volumes of soil and liquid phase solution process by which you can still homogeneous suspension is 1:3, the performance of industrial plants for soil decontamination at the specified equipment can reach 10 tons of soil/hr. Thus the use of Ferro - Ferrocyanides sorbent provides effective decontamination waste solution process. Especially pronounced is the effect on ^{134}Cs and ^{137}Cs. Technological scheme for decontamination of soil comprise the steps of:

Step 1. Chipping mixture of soil and large mechanical inclusions (fragments of building structures) in the crusher, which are produced industrially, to a state of loose material (particle size less than 4 cm).

Step 2. Mechanical fractionation got loose material, e.g. by vibration separation to separate a mixture into its constituent components:

- combustible materials (wood, biomass plants, plastic, etc.);
- crushed concrete building structures, metallic inclusions;
- soil.

Step 3. Thermal processing of combustible materials of pyrolysis gasification technology.

Step 4. Intensive washing of concrete and metal inclusions. Dosimetric control washed materials. Depending on the results of dosimetric monitoring these materials released from regulatory control, or direct for disposal.

Step 5. Getting pulp soil. To do this, washout formed in step 4, add to the soil. And add an additional number of process water to form a stable suspension.

Step 6. The pH of the pulp was adjusted to pH1 value in this case pH1 = 7.0 units pH.

Step 7. To separate the finely dispersed fraction the pulp is subjected to gravity separation method to obtain purified soil as sediment of large dispersed particles and flood - a suspension of fine particles of soil. Gravitational separation of pulp (hydro classification) is in the spiral classifier. Application of spiral classifiers due to the fact that these devices provide maximum compared to other types of classifiers, dehydration of large dispersed fraction of soil that is unloaded and, accordingly, the minimum content of the fractions contaminated with Cs process solution. The precipitate of large dispersed fraction accumulate, the suspension was centrifuged and pumped. The result obtained by finely dispersed sediment fraction (concentrate radionuclides) and illuminated technological solution.

Step 8. After centrifuging the suspension accumulated sediment of large dispersed fraction was subjected to leaching in lighted technological solution at pH = pH 2 (in this case 3 units pH). Leaching is carried out with stirring vigorously, for example, concrete mixer. This results in additional decontamination of soil due to desorption of radionuclides from the surface of large particles dispersed in a technological solution. In addition, as a result of repetition of washing soil particles are removed from it finely dispersed fraction remaining.

Step 9. After the leaching solution is drained from the mixer into the receiving container, bring the pH to a value PH3 (in this case 7 units pH) and resuspended in a solution of hydrological sorbent at 1:100 (1kg of sorbent in 100 liters of solution). The suspension was then subjected sorbent centrifuged to obtain a precipitate of spent sorbent (concentrate radionuclides) and clarified deactivated technological solution returned to the work cycle for reuse.

Step 10. The precipitate was dispersed of large fraction taken from the mixer, dehydrated and subjected dosimetry control. Depending on the results of dosimetric monitoring the material released from regulatory control, or direct for burial. Concentrates radionuclides (precipitation finely dispersed fraction and spent sorbent) subjected dosimetry control, conditioning and storage. The order of the method explained diagrams (Fig. 3, 4).

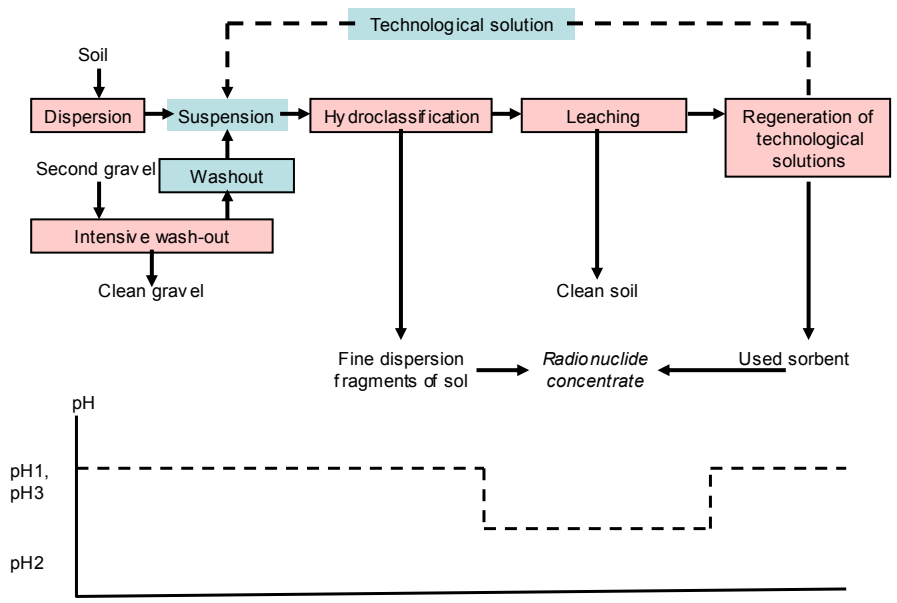


Fig. 3. Block diagram of the method (dashed line - pH - a series of technological solution)

As shown in the diagram (Fig. 4) combined wet technology associated with the pyrolysis gasification technology. Because of the design of ISF preferably placed near each other.

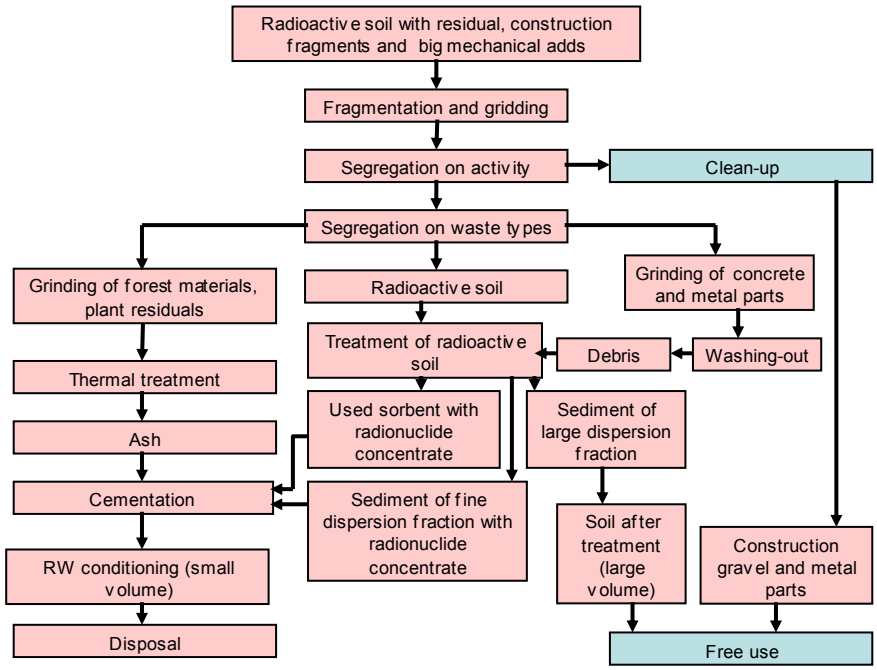


Fig. 4. Diagram of treatment of radioactively contaminated soils (combined wet technology)

3.2. Pyrolysis gasification technology

Pyrolysis - gasification technology developed and tested in the Chernobyl exclusion zone to solve the problem by reducing the number of combustible radioactive contaminated materials. Batch furnace from a mixture of different materials of solid radioactive waste (Fig. 5), after solar drying to remove moisture and crushed to a size of 60×60×60 mm, mixed with powdered additives that the combustion charge interact with Cs to form insoluble compounds. With the help of hermetic devices batch furnace is loaded into shaft furnace (periodic or continuous current mode). Thermal processing batch furnace is in its dense layer deterioration in the upward flow of air - oxidant. This emerging area of drying, pyrolysis, gasification of carbon, recovery combustion ash, cooling, in which the destruction organics and not organic; coagulation and adsorption, immobilization Cs aluminosilicate compounds in the interaction with impurities clay, zeolite, etc. The main products of thermal processing (pyrolysis gas and ash) are derived from the shaft furnace. Derivation of ash accumulated in it radionuclides performed using airtight container and gateway. When processing a mixture of organic not organic (concrete, brick, etc) are not organic residues are eliminated and can be used as fillers in concrete construction of radioactive waste storage facilities and roads. To that obtained in the furnace ash aerosol joins dropouts ash derived from gas cleaning filters.

There are two options for the treatment of immobilized ash:

- place the ashes for intermediate storage in sealed steel drums or containers or
- translate ash in the monolith, while it is mixed with a solution of binding materials (cement, sand, and water) and filled with monolithic ash containers arriving at an intermediate storage.

Obtained pyrolysis gas is burned additionally obtained heat is converted into heat and electricity. Gas emissions are cleared in multistage gas cleaning system.

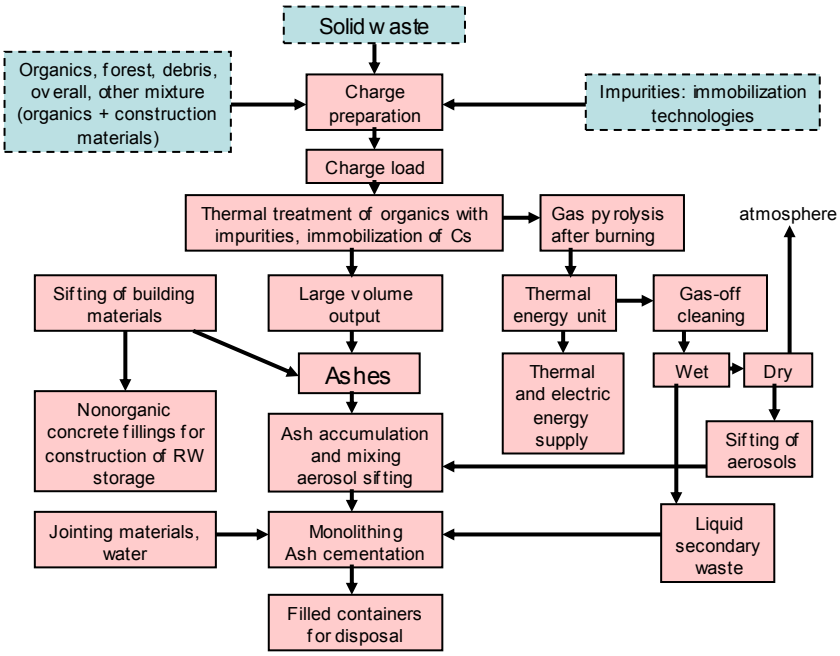


Fig. 5. Scheme of management of radioactively contaminated organic (pyrolysis gasification technology)

3.3. Alternative options integrated wet technology

Evaluation of alternative approaches (including Chernobyl experience) the main alternative approaches are:

1 The passive strategy when clearing the territory is due to natural radioactive decay of radionuclide contaminants.

2 Bioremediation using plants and fungi when clearing is not only due to natural radioactive decay of radionuclide contaminants, but also due to root uptake of radionuclides, which is enhanced in the presence of symbiotic fungi.

A comparative analysis of alternative approaches showed that the passive remediation strategy area (for ^{137}Cs) will last 300 years. The use of bioremediation technologies will reduce this period to 100 ÷ 150 years (the Chernobyl zone). However, removal of the surface layer of soil and its processing using combined wet decontamination technology will solve the problem in the course of several years (performance industrial facility

decontamination of soil about 10 t/h). An approximation to the proposed method is a method based on the fractionation of soil contaminated with radionuclides. The main stages of the process:
1. Withdrawal of the upper soil layer.
2. Remove the large inclusions and biomass plants by screening on sieves.
3. Soil dispersion in the aquatic environment to form a pulp.
4. Ultrasonic treatment pulp to a homogeneous suspension state and destruction of waterproof materials.
5. Hydro suspension classification by size and density of particles of soil.
6. Separation suspension of fine fractions containing most of the radionuclides.
7. Processing of suspension poly electrolytes to accelerate the deposition of fine fraction.
8. Regeneration of poly electrolytes for reuse by washing the precipitate with concentrated solutions of salts and acids.
9. Dehydration and subsequent disposal of sludge.
10. Back purified water solution for reuse in obtaining pulp output ground.
Attention is drawn to that this method involves the use of potentially toxic chemicals (flocculants), as well as mineral acids and salts on the stage of regeneration flocculants. As a result, significantly increases the amount of liquid chemical wastes and risk of converting radioactive waste in addition to chemical waste due to residual mineral acids. Also characteristic of this method is the high content of residual waste process solution in the final product - the soil after treatment. As follows from the presented data of large dispersed soil fraction obtained by hydro, containing up to 30% moisture in a technological solution. When washing the soil more than 50% of the initial amount of radionuclides goes in a technological solution that increases its specific activity. Consequently, the residual specific activity of the soil after treatment may not be less than 20% of the initial value.
Experimental data presents the results of testing different methods of decontamination of soil directly "on the spot". Emphasis is placed on high performance (deactivating effect, reducing waste) resulting test - experiments. However, it should be noted that these results were obtained during the processing of a limited number of samples specific soil.
At the same time, deactivating effect and the indicator "volume loss" is largely determined by the relative content in soil clay, silt, sand, and mineral composition of these components. Therefore, our work aimed to quantify the effect of deactivating and indicator "volume loss" for all soil types (according to the International Society of Soil Classification soil (SSSA) and company agronomists (ASSA) USA, which differ in particle size composition and content of clay, silt and sand.

3.4. Alternative options pyrolysis gasification technology.

When performing work on the technical and economic evaluation of the pyrolysis gasification technologies for radioactive waste from 16 selected screening technology systems by compacting the volume of radioactive waste combustion systems with 2 groups of settings that can be used for emergency combustion of organic radioactive waste - pyrolysis gasification immobilization of ^{137}Cs and direct combustion in the installation of municipal type EKSYS Ltd. In recent years attention-grabbing alternative composite techniques that extend to Japan.

1. Technology of decrease volume and weight of organic waste (grass, vegetation, mud water treatment plants, Japanese cedar, farm waste, etc.) by aerobic fermentation of organic matter with subsequent combustion residue. Due to the volume reduction achieved by fermentation and organic weight to 95%. Firm Mishimax of Mikuniya Corporation made installations for processing organic waste chemical decomposition, by the action of microorganisms, ranging from 100 kg/day to 1000 kg/day. In Ukraine such works for the utilization of organic biomass fast pyrolysis method from the combustion residue is introduced into scientific and engineering center "Biomass" Institute of Engineering Thermal Physics.

2. Volume reduction technology of organic radioactive wastes from biomass combustion in the boiler with hydraulic fire grate, including waste containing pollutants wet. The special design, which is equipped with a modern gas-allows processing organics with minimal emissions, the production of heat and electricity. Processing capacity of one block of 4 t/h., three blocks of 12 t/h., with production of 15 MW and 4 MW of thermal electricity. The excess energy allows dry a little charge of water pollutants. When humidity ~ 60% charge combustion temperature does not exceed 1000˚C that it is important to reduce the mass transfer of ^{137}Cs in gas-increasing transfer factor of 137Cs in ash. Such systems "turnkey" produced by the Austrian company Polytechnik Biomass Energy and its Japanese branch of Japan Matsubo Corporation (installation Biomasse Marusen power 4000 kW). The cost of a capacity of 10 MW - 30 million €.

3. Volume reduction technology of organic wood and biomass combustion in the boiler gasification charge in an upward flow of gas; heat output of 6 MW is considered a promising method for processing contaminated biomass from wood Chernobyl while obtaining energy. Equipment firm produces Bioner (Finland).

4. Technology of separation of ^{137}Cs from soil and additional combustion of organic impurities in it by continuous supply of soil into the installation with rotating heated drum (similar installation EKSYS Ltd), evaporation of ^{137}Cs and its condensation in the cooling system. Installation can also burn additional organic soil, capture aerosols in gas-cleaned soil in place for the collection of follow-up and use. This alternative wet soil decontamination technology was developed by National Agriculture and Food Research Organization (Japan). Depending on the temperature evaporation of ^{137}Cs and impurities in the soil, accelerating, rate of removal of ^{137}Cs in soil is 80% ÷ 99.9% (at 1000 ÷ 1300°C).
In Fig. 6 is a diagram of thermal separation technology ^{137}Cs from soil.

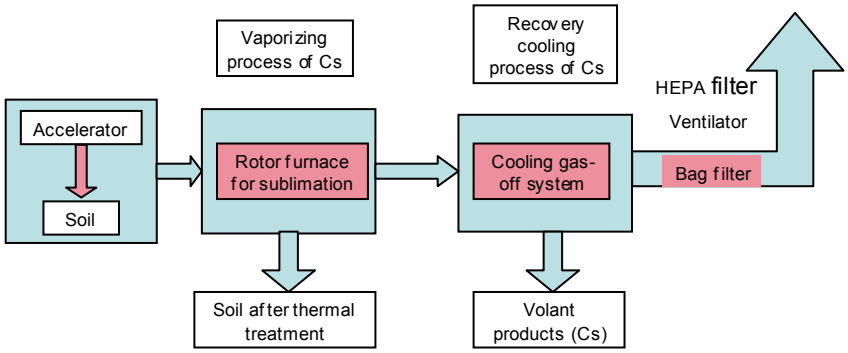


Fig. 6. Scheme of thermal separation technology ^{137}Cs from soil.

Marked technology and thermal processing complement detailed comparative analysis of systems, they are close to the municipal type. Technology section 1, 2, 4 are tested in Japan. With proper additional equipment gas cleaning with HEPA filters - they can be used in combination with the selected and recommended to achieve the ultimate goal - installation of Institute for Chernobyl Problems and EKSYS Ltd.

3.5. Justification of combined wet technology.

Indicator "reduce volume" (K_1), which was defined as the ratio of the total soil (M_1) to the amount of solid radioactive waste arising from decontamination (M_2):

$K_1 = M_1/M_2$.

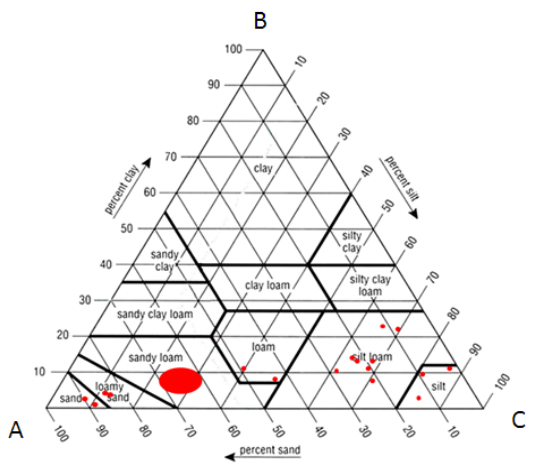
Deactivating effect (K_2), which was determined at the initial specific activity of soil (A_0) and the residual specific activity of soil (A_2) after deactivation:
$K_2 = (A_0 - A_2)/A_0$.
Suitability of combined soil decontamination technology was evaluated for its effectiveness for all types of soil according to the SSSA and company agronomists ASSA depending on the particle size distribution of soil (clay content, silt, sand). A database index suitability and performance for all types of soil was created.
To assess the applicability of the technology classification chart SSSA/ASSA with parametric coordinates (Fig. 7) transformed to the form of a triangular matrix without changing the basic requirements parametric coordinates - the constancy of the sum of the coordinates. In this case, this requirement is expressed as follows:

C(%) clay + C(%) silt + C(%) sand = 100%,

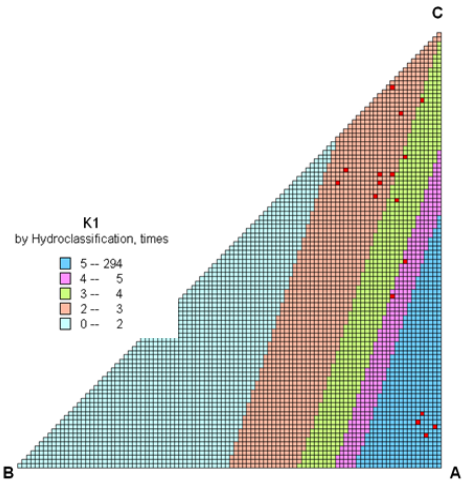
where C (%) clay content, silt, sand in the soil.
Each point classification diagram and, accordingly, each element of the triangular matrix correspond to individual components of the value content of the soil and thus grain size. For example, for the vertices of a triangle chart and classification of triangular matrices have the following value content of soil components:

Peak A (%) clay = 0%;	C (%) silt = 0%;	C (%) sand = 100%
Peak B (%) clay = 100%;	C (%) silt = 0%;	C (%) sand = 0%
Peak C (%) clay = 0%;	C (%) silt = 100%;	C (%) sand = 0%

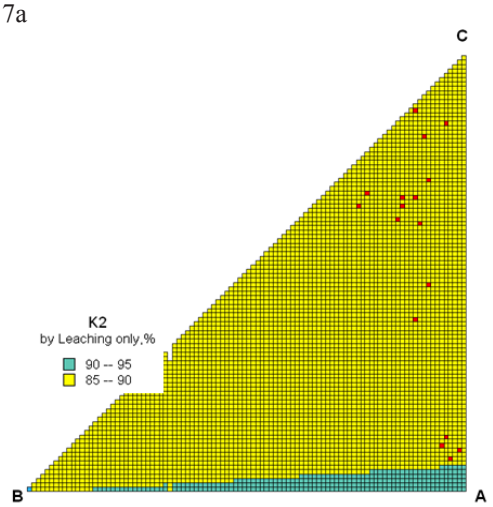
Triangular matrix, except percentage concentration of clay, silt and sand in the soil also contain information about the contents of the main minerals of the soil, as well as information about their specific sorption capacity at pH1 and pH2. The combination of these data was used as the source. Further, the methods of computer modeling values were calculated performance criteria (K_1 and K_2) for the whole set of values. We performed calculations of step changes by 1% for each of the variables C (%) clay, C (%) silt and C (%) sand.
To visualize the results of calculations (Fig. 7b, 7c, 7d), we used the method of constructing thematic maps software system MapInfo.



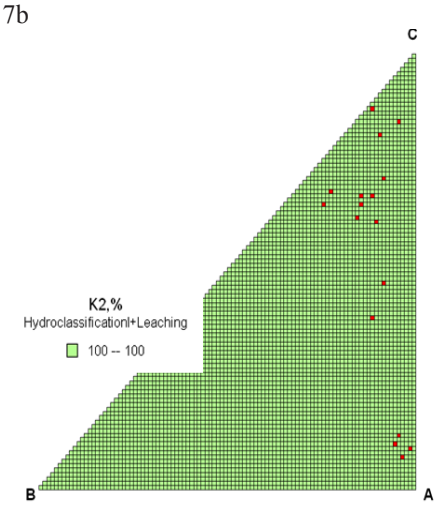
7a



7b



7c



7d

Fig. 7a. Classification chart of the SSSA and company agronomists ASSA.
Fig. 7b. Dependence of K_1 "reduce volume" for hydraulic classification depending on the composition of the soil.
Fig. 7c. Dependence of K_2 decontamination effect only for leaching depending on the composition of the soil.
Fig. 7d. Dependence of the total decontamination effect K_2 (hydroclassification + leaching) depending on the composition of the soil.

The points in the diagrams - size distribution of soils California, recommended us to use as analogues of soil in Fukushima prefecture. As can be seen from Fig. 7b, an optimized version of the technology has the potential to provide a significant reduction of radioactive waste (contaminated soil). It should be noted that even if the solid waste generated will relate waste sorbent (after decontamination process solution), even in this case, the processing of soils similar soils Fukushima prefecture, the rate of K_1 ("reduce volume") will vary limits 4 ÷ 100. Deactivating effect of leaching (Fig. 7c) varies from 80 ÷ 90% and almost the same for soils - analogues Fukushima Prefecture. However, as shown in Fig. 7d, the total deactivating effect (hydroclassification + leaching),

according to the results of computer simulation, close to 100% and should not depend on the composition of soil processed.

4 Conclusions

Scientific Results of Project:
- Semi-empirical model to quantify the distribution of radionuclides ^{134}Cs, ^{137}Cs between particle size fractions of soil.
- Results of calculations of the equilibrium distribution of ^{134}Cs, ^{137}Cs between the dispersed phase (particle size fractions of soil) and dispersed environment (technological solution) in the decontamination of soil.
- Analytical and experimental model of mass transfer of radionuclides in organic thermo processing of radioactive waste.

Environmental Results of Project:
- Transfer of radioactive waste into a form suitable for disposal, excluding their migration in the environment.
- Concentration of ^{134}Cs, ^{137}Cs in a small volume of waste that facilitates and accelerates the process of their disposal.
- Accelerate the process of decontamination areas that will contribute to the normalization of the radioecological situation in the regions affected by the accident at the Fukushima Daiichi.
- Minimization of gaseous radioactive emissions and liquid radioactive waste processing products decontamination.

The Economic Results of the Project
- Cost savings due to reduced volumes of radioactive waste processing resulting sort of radioactively contaminated materials.
- Return to reuse clean of radioactively contaminated materials resulting complex sorting.
- Reducing the cost of disposal of radioactive waste due to reduced volumes of the primary radioactive waste.

Other Results of the Project:
- Removal of organic and combustible impurities in radioactive waste destined for disposal.
- Bringing characteristics of waste after processing in accordance with the eligibility criteria for disposal.

5 Applicability and Recommendations for Japan

5.1. Pyrolysis gasification technology

When using pyrolysis gasification technology (Institute of Chernobyl Problems) combustion in the shaft furnace in a dense layer of much the band up flow air oxidant possible recycling of rare-earth organic and 100% of a mixture of inorganic to 80%, humidity to 60 ÷ 80%.

The design capacity of processing is for 1 reactor 500 kg/h, and the block for 4 reactors of 2000 kg/h. Compaction ratio of organic 90 ÷ 100 times. Combustion technology is performed with simultaneous immobilization of Cs introduction of the charge impurity clays and other materials (1 ÷ 2% by weight), which also leads to a fall in pressure in the gas phase Cs. Low temperature in the combustion zone - to 1000˚C. Coagulation and immobilization of Cs provide its minimum mass moved in gas cleaning and minimal leaching of ash. Recycling of contaminated wood compaction was performed with the charge that contained ^{137}Cs specific activity of 10 kBq/kg.

Experience processing contaminated wood of Chernobyl Exclusion Zone to install capacity of 50 kg/h showed the following results:

Compaction ratio of the charge	100 times (wood) 25 to 80 times (from organic and not organic, which subsequently discarded from the ashes).
The transition rate Cs in ash	95 ÷ 98% (Cs in the ash mainly in the form of insoluble compounds aluminosilicates, carbonates, sulfates).
Coefficient of leaching Cs from ash	$<10^{-5}$ g/cm^2 per day.
Modern gas cleaning provides: cleaning of solid particles and aerosols quality of emissions	to 99.99% according to the standard requirements
The concentration of ^{137}Cs in the air torch output (D)	$< 8 \cdot 10^{-1}$ Bq/m^3.
Costs: capital (in 2004 prices). block of 4 reactors	 20 million €. (1 main reactor 10 million €, 3 auxiliary 10 million €).
Exploitation	1 million € per year.
Cost of processing 1 ton	166 €

5.2. Technology of Ltd. EKSYS

When using the rotary combustion technology experiments were studied processing of radioactive waste (organic - 87.4% plastic - 0.5%, building materials - 5% and others).
The technology is based on the principle of rotary combustion charge in revolving drum and requires no prior grinding charge and branches.
Experience processing of contaminated with toxic materials charge of municipal waste. When processing was used as a label isotope ^{133}Cs, which was introduced into the charge in an amount that simulates a specific activity of 8 kBq/kg. This shows the following results:

Compaction ratio of the charge	90 ÷ 95 times
The transition rate ^{133}Cs in ash	to 80%
6 stepped gas cleaning with thermo-catalytic combustion and hermetic ash capture provides cleaning of solid particles and aerosols	to 99.99%
Quality emissions	according to the requirements
The experiments also showed technology provides clean the aerosols ^{133}Cs	to 1×10^{-1} Bq/m^3 at a rate of D $8\cdot10^{-1}$ Bq/m^3.

Has license and positive decision of the State Ecological Expertise Ukraine and Russia. Stationary produced commercially - Research Enterprise "Energosteel", Kharkov (Ukraine).
Costs (in prices 2006):
Capital 199 million UAH;
Operational 12.87 million UAH;
Cost of processing 1 ton of 288 UAH.

Based on the positive results of the first phase of the project and coordination of stakeholders Japan, we recommend the following stages of this project after the end of this report..

5.3. Step 2 – Preproject study.

As a continuation of the feasibility study will be carried diligence volume reduce of radioactive waste containing radionuclides ^{134}Cs and ^{137}Cs. This phase will be based on the recommendations of stakeholders from Japan on site selection for the operation of a pilot demonstration plant in order to reduce the volume of radioactive waste containing radionuclides ^{134}Cs and ^{137}Cs. The results of a feasibility study will be determined by taking into account the proposed location and characteristics of waste streams. At the beginning of the report will be released preliminary assessment and survey the location of the pilot plant. Preproject research will also include:
- Viewing detailed design criteria for the development and take account of changes in the project which is the result of a preliminary assessment report and survey the location of the pilot plant.
- Requirements for the infrastructure.
- Preliminary assessment of radioactive emissions and discharges.
- Assessment of health risks, including a quantitative description of the sources of emissions and discharges, identification and evaluation of pollutant pathways of, and preliminary assessment of doses from harmful pollutants.
- Preliminary project schedule based on information available at the stage of preliminary design.

In cooperation with Japanese stakeholders will develop design criteria for conditioning of radioactive waste at the end of their treatment to meet the admissibility criteria at the disposal of radioactive waste.

5.4. Step 3 - Conceptual design.

As a continuation of preproject research will develop a conceptual design of a pilot demonstration plant in order to reduce the volume of radioactive waste containing radionuclides ^{134}Cs and ^{137}Cs, and will be released a report on the conceptual project. Conceptual project will include:
- The characteristics of the project needed to assess the impact on the environment, emergency plan and report on the environmental impact.
- Preliminary design drawings, which are sufficient for understanding the design features of each facility.
- Estimates of doses to operating personnel.

In cooperation with Japanese stakeholders:
- It will be produced and agreed with the Japanese regulators preliminary safety analysis report with sorting and processing waste.
- Preliminary cost estimate of the project and other evaluation required for comparison of alternative projects, prepared for the preliminary project.
- A short list of specifications for the construction and purchase of equipment.

- Identification of future long-term purchases.

Inorganic Waste Volume Reduction with Polymer

ISTC-A-2071

Advanced polymer systems providing deactivation of different surfaces and soil from radioactive pollutions

Dr. Zoya Farmazyan
CJSC "Yerevan SRI Plastpolymer" (Project Manager) (Armenia)

ISTC/STCU Fukushima Expert Meeting

Tokyo, Japan
November 5-6, 2015

Summary Report – project ISTC #A-2071

on work performed from July 1, 2013 to July 1, 2015

CJSC "Yerevan SRI "Plastpolymer"

Project Manager: Farmazyan Z.M.
Ph.D

Title of the Project:	Advanced Polymeric Systems Providing Deactivation of Different Surfaces and Soil from Radioactive Pollutions
Commencement Date:	1 July 2013
Duration:	2 years
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Participating Institutes:	-
Foreign Collaborators:	Dr. Upendra Singh Rohatgi Brookhaven National Laboratory, USA Dr. Sang Don Lee, US Environmental Protection Agency EPA Dr. Mitsuru Akashi, professor, Osaka University, Department of Applied Chemistry, Graduate School of Engineering, Japan

1. Brief description of work: objectives, and expected results

Project goals and objectives

At the beginning of the project two main problems were the subjects under study:

1. Development of polymer systems for decontamination of porous surfaces as strippable films;

2. Study of the behavior of polymer solutions in soils in the following aspects :

- As solidification agents for top soil layers on fields.
- Examinations of polymers solutions for decontamination of soils from radionuclides ^{137}Cs and ^{134}Cs ;
- Study an effect of combining polymers with microscopic soil fungi (MSF) on radionuclide fixation;

Expected Results and Their Application

1. *Deactivating strippable films:* Development of new polymer systems implies the possibility of their use for decontamination of porous surfaces as strippable films instead of the commonly used high-pressure water.

2. *Soil decontamination*

Several variants for application of polymer systems were foreseen for soil decontamination:

- To fix the radionuclides in soil to prevent dissemination by creating an accumulating and retaining polymer layer.
- To concentrate the radionuclides into narrow layer of soil before soil is being removed from fields.
- To use polymeric systems for decontamination of the removed soil wastes.

On April 13-18 of 2014, the review and discussions of the project implementation results (for 1-3 quarters) was held at the ISTC / STCU Technical Review Committee on the environmental assessment for long term monitoring and remediation in and around Fukushima, Fukushima and Tokyo, Japan.

As a result, in view of current situation on Fukushima, Japanese side noted that cleanup of hard surfaces with polymers is of no interest at this time, but mechanism of trapping and moving of ^{137}Cs in soil is of great interest and importance.

Therefore, work with hard surfaces was stopped, and all further efforts were aimed to investigations behavior of polymer solutions in soils

Nevertheless, by the end of the third quarter the principal variant of polymer synthesis was found which provide removing polymer films from porous surfaces. Also a part of works on soils was performed.

The main results of the works, performed before April 2014 are summarized below.

Deactivating strippable films

- New polymeric systems for using as strippable films for decontamination of porous surfaces from ^{137}Cs and ^{134}Cs were developed
- The basic technological process of the new polymers was designed. Parameters of the technological process ensure films quality with desired operational properties such as adhesion, strippability, plasticity-elasticity, depth of penetration into the porous surface.
- Properties of the films were studied on 4 porous surfaces, differing in chemical composition and porosity. The tested samples penetrate into pores; plasticity/elasticity of films, formed after drying, allows pulling the film from the surface layers. Due to this percent of removing ^{137}Cs from porous surface reached up to 35%.

- The developed technological process and properties of the products are competitive with one of the known compositions for decontaminating coatings -water dispersion ALARA-1146 of Carboline production, which we used as an analogue.

Soil decontamination

Objects under study

Soil. The first soil sample (sANPP) was sampled at the area around the Armenian nuclear power plant ANPP (Ararat plain) and corresponds to type of a soil around Fukushima (Gray Lowland soils)

Accordingly to XRD analysis, the clay fraction consists mainly from montmorillonite MMT $(Al_{1.67}Mg_{0.33})Si_4O_{10}(OH)_2\,Na_{0.33}$. Data of mechanical composition are provided in Table 1.

Preparation of soil, contaminated with radionuclides. All soil samples throughout the projet were artificially contaminated with radionuclides ^{137}Cs and ^{134}Cs. The source of radioactive contamination was radioactive water.
The sieved soil with particle size 2-3 mm was mixed in several doses with radioactive water in ratio 1:0.4 by weight, and then was air-dried. The entire process of sample preparation lasted 8-10 days.

Polymers

Initially for soil experiments two groups of polymers were used and investigated (as screening of samples):

1. Polyvinyl alcohols samples (PVA), in two variants: the known industrially produced grades and our own elaborations as modified polyvinyl alcohol (MPVA). PVA-s contains mainly hydroxyl and acetate functional groups in different ratio; MPVA additionally have carboxyl containing monomers.
PVA and MPVA samples are water-soluble powders, for soil tests their solutions (0.5-3.0%) were used.

2. New developed VA-AA polymer system, based on vinylacetate (VA-AA copolymers). These are types of water-dispersion products. Variations in ratio of comonomers and synthesis conditions allow obtaining of polymers in a wide range of properties, from solutions to dispersions. For soil tests diluted solutions/dispersions (0.5-3.0%) were used.

Hereafter the both polymeric system will be referred as "polymer solutions"

Methodology of soil experiments

Initially, to solve the project tasks on soils, study of **downward** stream of polymer solutions in soil was foreseen, i.e. polymer solutions should be sprayed over the top layer of soil. Height of experimental vessels (6-7 cm) was chosen reasoning from knowledge that the main amount radioactive Cs is distributed in soil in upper layers of soil (up to 3-5 cm).
A plastic tube (height 6 cm, D-6, 2 cm) has been filled with contaminated and moistened soil sample (~250 g).
Then the certain volume of polymer solution (15-20 ml) was poured over the topsoil.
After 24 hours the upper layer of the plastic tube of 1-1,5 cm height was cut, and the second portion of polymer solutions was applied over the second soil surface.
After 24 hours the second layer of the plastic tube of 1-1.5 cm height was cut. Radioactivity of the initial soil, all the layers and the remaining part of the soil were measured. Further this volume of experiments is referred as Stage **1.**

The main results and conclusions from the soils experiments (before April 2014)

Solutions of several samples PVA, MPVA and VA-AA copolymers were tested by the above described methodology.
Two series of experiments were also carried out with the same solutions, to which were added water-spore suspensions of soil fungi (Aspergillus niger, Aspergillus flares, Aspergillus apricalis, Alternaria alternata, Rhizopus stolonifer)

The main important effects were discovered at this stage of experiments:

1. Increasing of activity ^{137}Cs, ^{134}Cs in the upper soil layers was noted, in case when some polymer solutions (two grades of polyvinyl alcohol) were poured over the top soil layer.
2. For all the other tested solutions the opposite effect was observed: increasing of Cs activity in the lower layer, in parallel to solutions movement.
3. The additives of fungal spores to the polymer solutions showed a significant change in the dynamics of motion of Cs. Presence of fungi led to increasing Cs mostly in the lower layers.

The basic conclusions:

1. Behavior of polymer in soil is similar to water, with manifestations of capillary and gravitational movement.
2. In the carried experimental conditions (solutions flow top-down) the capillary and/or gravitational movement of radionuclides in the soil under the influence of polymer solutions is occur.
3. The investigated polymer solutions have a number of parameters that may determine their gravitational and capillary behavior in the soil. Given that these two opposite movements in the soil determine increase of ^{137}Cs in the upper or the lower layers due to binding with polymers, further studies would be conducted to identify the role of each parameter of polymeric systems.
4. The fact of increasing ^{137}Cs in the topsoil as a result of capillary movement of polymer solutions has shown the need for a separate study of capillary and gravitational behavior of polymers.

A series of experiments was carried out under experiments Stage1, but polymer solutions were applied so that penetrate into soil **upward** (from down to top).

Results of all experiments with different polymers solutions (on one type of soil, sANPP) have identified the main factors regulating efficiency of redistribution ^{137}Cs in soils :

- Initial soil moisture (ISM),
- Volume of polymer solutions,
- Ratio of hydrophilic and hydrophobic functional groups in polymers, presence of inorganic groups,
- Morphological state of polymer systems (solutions or dispersions).

All these findings were analyzed, the conclusions were made, and the basis of detailed studies after April 2014 was formed.

2. Modifications of work after April 2014 meeting

The main task after April was formed as:

1. Study of capillary and gravitational movement Cs in soil influenced by the polymer solutions;

2. Possibility to use results of these studies within the common problem to reduce the amount of radioactive soils wastes which were taken off from the fields and stored in bags.

To solve the new tasks, the methodology of experiments was being corrected according to the previously obtained results. Also number of objects under studies was increased.

Soils.

1. In addition to sANPP (enriched with MMT), two other soils types containing muscovite and illite, were sampled from different places, located around the Tsaghkunyats mountains- Meghradzor (sMdz) and Tsaghkadzor (sTh).

According to XRD analysis, these soils contain:

Mica-montmorrilonite – $((Al_9Ti_9Fe)Al_{0.68}Si_{9.32})O_{10}(SOH_2)(K_8Na)_{0.43}X_{0.17}$
Illite- $(K_9Na_9Ca)_{1.36}(Al,Mg)_{4.17}(Si,Al)_8(O,SOH)_{24}$
Muscovite - $(K_9Na)(Al_9Mg_9Fe)_2 \quad (Si_{2.1}Al_{0.9})O10(SOH)_2$
Illite- $(K_9Na_9Ca)_20_{3.33}(Mg_9 Mn)O.4.3(Al_9Fe_9Ti)_2O_2 16(Si_9Al)O_2 4H_2O$

Soils sMdz и sTh are very similar in mineralogical composition, but differ in mechanical composition (Table 1)

Table 1 . Mechanical properties of soils

Place of soil sampling	Content of particles of different size (mm) in soil suspension (%) (Kachinski method)					
	1-0.25	0.25-0.05	0.05-0.01	0.01-0.005	0.005-0.001 5-1 μm	<0.001 < 1 μm
Ararat plane (sANPP soil)	4.2	21.84	23.1	12.0	33.58	5.28
Meghradzor, sMdz	16.81	29.37	27.24	4.9	14.74	6.94
Tsaghkadzor, sTh	1.35	6.06	20.12	11.52	28.4	32.56

2. Throughout the project the results of investigations were discussed and analyzed by experts Dr. U.Rohatgi and Dr.Y.Onishi.

It was suggested to refine mechanism Cs binding by polymeric solutions on pure clay minerals (illite, vermiculite). The reasons for that: 1-these minerals are known to be mostly fixed ^{137}Cs and ^{134}Cs in soils; 2- Fukushima soils (clay fractions) mostly contain namely these minerals; 3- Japanese specialists plan to sieve contaminated soils to separate highly contaminated silt and clay fractions.

Vermiculite (Vm) was purchased as raw material with large particles of 1-2 cm, which were crushed and sieved through meshes 0.25 mm (Vm-0.2 mm).

The illite-enriched fraction was separated from soil sMdz- 2 mm by sieving through a sieve 0.2 - 0.25 mm (sMdz-0.2 mm).

All soil samples were contaminated with ^{137}Cs, ^{134}Cs as described above. Level of Cs activity in soils was within 6000-10000 Bq/kg (sANPP) and 5000-6000 Bq/kg (sMdz).

Only to contaminate sMdz-0.2 mm and Vm-0.2mm, weighted samples were placed into packets from filtration material, and immersed into radioactive water. 24 hours later the contaminated soil and Vm were took out of the bags and air dried. Cs activity in these samples was ~6000 Bq/kg and ~2500 Bq/kg respectively.

RIP

In order to assess the similarity between soils used in the project and soils around Fukushima NPP, RIP (radiocesium interception potential) values of tested soils (sANPP-2 mm, sMdz-2 mm, sTh-2 mm) were determined (Table 2).

Table 2 RIP for different soil

Soil	RIP (K) for ^{137}Cs, mmol/kg	RIP (K) for ^{134}Cs, mmol/kg	∑ RIP (K mmol/kg
sANPP-2mm	1296 ± 94.5	1234 ± 91.6	2530
sMdz-2mm	646 ± 6.4	650 ± 19.6	1296
sTh-2mm	773 ± 60.5	728 ± 60.3	1501

The data obtained correlate with data of RIP values for various soil and clay minerals in Fukushima, provided on Page 77 in the 81-pages document (in Japanese) from Dr.Onishi.

Polymers systems

Based on the results before April 2014, a set of studied polymer systems was changed.
1. Number of PVA samples was supplemented with new grades, which together with the previously selected samples allowed to estimate impact on the behavior of polymer solutions in soil such polymer characteristics as a ratio of hydrophilic / hydrophobic functional groups, viscosity and molecular weights MM.

2. Based on the PVA and VA-AA copolymers test results, methods for introducing hydroxyl groups into VA-AA copolymers were found.
As a result, new technology of synthesis the hydrolyzed VA-AA copolymers (VA-AA-H) was developed.
These copolymers have the most optimal properties to be used in soils, from point of view the project tasks.

3. The developed technology provides synthesis of polymers with reproducible characteristics. A pilot sample VA-AA-H-80 was synthesized by this technology, which was then used in all Batch tests (as Stage 4).

4. By the developed technology a series of new samples-analogs of VA-AA-H-80, but with lower viscosity was synthesized, for testing on sMdz-0.2 mm and Vm-0.2 mm

Stages of experiments
Experimental stages in soils throughout the project are defined as **Stage 1 –Stage 4**

Stage 1 Experiments in vessels (height 6 cm, D-6.5 cm), as described above

Stage 2 Experiments to study capillary rises of water and polymer solutions into soil in polymeric tubes (H=35-60 cm, D =1.9-2.1cm), soils samples ~100-200 g

Stage 3 Experiments in columns, height of 35-40 cm, similar to Stage 2. Soil weight 1.2-1.5 kg
Based on analysis of all the results, it was concluded that parameters influencing the processes in these volumes under the experimental conditions can not be evaluated in full.

Stage 4 Small Batch tests – These are the volumes of experiments where all regularities and findings revealed at the Stages 1-3 appear.

The height of the soil in vessels (Hmax) is determined by the pore size and was assessed in tubes (Stage 2) for each tested soil. Width of vessel was increased so that it is possible to take off three columns with soil for analysis within different time intervals. The first experiment at ANPP showed that in this volume both capillary and gravitational effects appear , the results are reproducible and allow to draw conclusions and predict subsequent experiments.

So, all experiments at ANPP within 7-8 quarters were done in conditions of stage 4 as the Small Batch tests. Both downward and upward experiments were carried out in sANPP, sMdz, sTh (soils particles size 2-3 mm), and also downward experiments in soil sMdz-0.2 mm and Vm-0.2 mm.

The resulting methodology was as follows:
The soils of a certain weight (2.5-4 kg) and initial moisture content (2-8%) loaded into a plastic vessel to a height of 25-40 cm; the bottom of the vessel is cut off and covered by two-layer gauze. The vessel is set in tray on a holder (ring or mesh with large pores) of height 0.5-1cm. In case of upward experiment the tray is filled with a required amount of polymer solution. In downward experiment a solution is poured over the top layer gradually. In both cases time of complete absorption of solution into soil is registered.

For this experiment, method of soil sampling (similar geological core) by a sharpened plastic tube with inner diameter 3.5 cm and a height of 30-40 cm was elaborated.
The tube is inserted into the studied soil to the bottom, pulled out and is cut into several pieces, of which a soil sample is taken for analysis (^{137}Cs, ^{134}Cs). Three sampling are done with interval 24, 48 and 120 hours.
After sampling the first tube with soil, an empty tube is inserted into the empty soil space to not disturb equilibrium of the system. The same is done for the second and third sampling.

Washburn equations
To compare capillary rise of polymer solutions into all the tested soils, Washburn equations was used that defines the kinetics and rate of liquid penetration into a cylindrical capillary, depending on liquid properties:

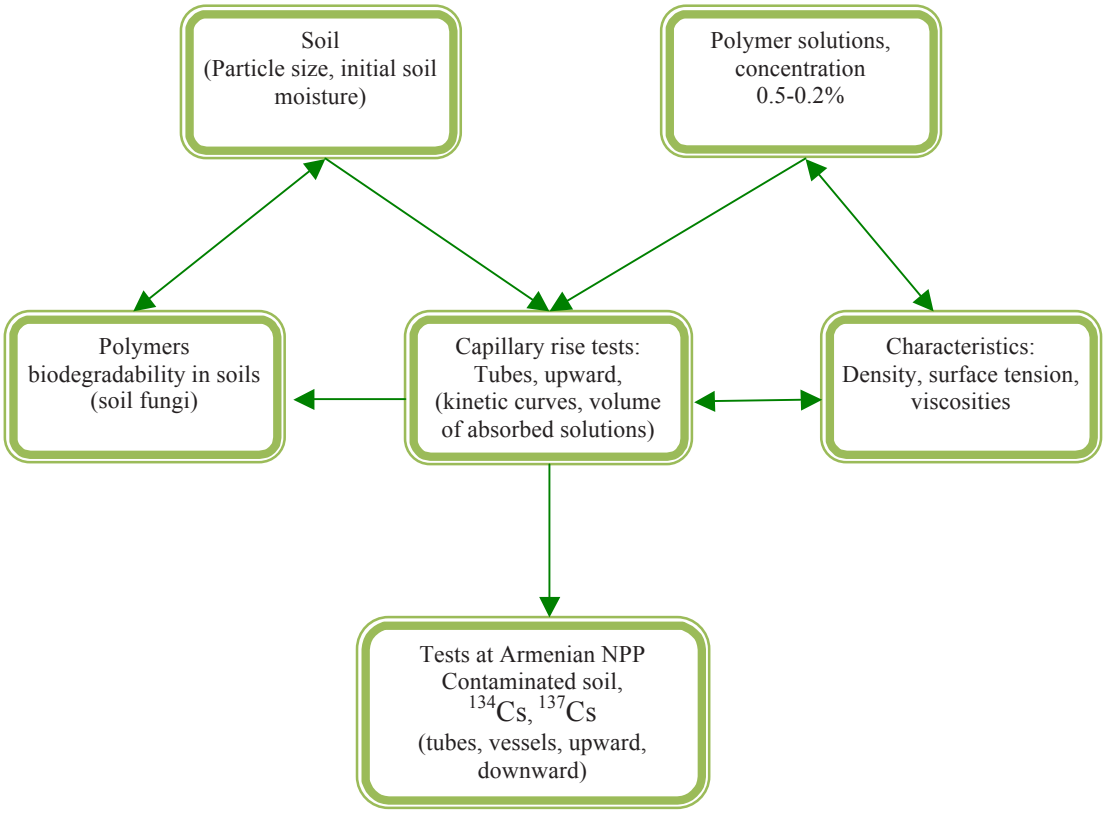
$$h(t) = \sqrt{\frac{\sigma \cdot r \cdot \cos\theta}{2\eta} \cdot t}$$

where σ is surface tension of liquid (polymer solution), θ – wetting angle, h – length of capillary region filled by solution at the current moment, η is viscosity of liquid, r – radius of pores.

Values (η,σ) for all the tested polymers solutions were experimentally determined. For assessment of pore size r and wetting angles θ^0 the kinetic curves of polymer solution rise was applied.
The method was implied to select the pairs of contact angle θ^0 and pores diameter **r** so that the theoretical kinetic curves of solution rise coincide with the experimental ones.
This approach allows estimating the pore sizes through which polymer solutions penetrate in soil capillaries.

3. Technical approach, method, experiments, theory, etc

After April 2014 the stages of work can be described with the following flowchart:



Methodology of experiments

In the course of works within last quarters, based on a comparative analysis of all carried out experiments in the polymer laboratory and at ANPP, a way of experiments is revealed, in which data of capillary tubes serve as guidance for scaled experiments. This methodology includes several steps:

1. First in tubes with soil (of known ISM) kinetics of polymer solutions capillary rise are determined along with humidity distribution and volumes of absorbed liquids.
2. Next reasoning from data of step 1, optimal samples (polymer solutions with needed surface tension and viscosity which provide capillary rise of solutions) are selected
3. Then the selected solutions are tested in tubes with contaminated soils at ANPP (upward experiments)
4. Based on the obtained data, polymer solution for scaled experiments (Batch tests) are selected.

All experiments in 7-8 quarters were conducted by this methodology, upward-downward experiments on soils sANPP, sMdz, and sTh (downward). The difference in the activity of ^{137}Cs between layers reached 40% in sANPP, and 30% in sMdz.

Below as an example this methodology is shown in case of the soil sMdz-0.2mm; ISM 6.5%, soil by weight 4 kg, polymer solutions applied top-down (downward).

A series of VA-AA-H copolymers with lower viscosity was synthesized for testing on sMdz-0.2mm and Vm-0.2mm

1.Table 3 shows a part of synthesized copolymers samples and their functional groups.

Sample VA-AA-H 80 earlier was investigated experimentally on all soils with particle size of 2-3mm.

Sample PVA GF is one of the reference samples in the development of a series of copolymers of VA-AA-H, so there is shown for comparison.

Table 3 VA-AA- H copolymers

Sample VA-AA-H (“Tests”)*	Content of functional groups, % weight		
	Esters	Carboxyl	Hydroxyl
80	19,8	19,3	60,9
83	53,5	21,3	25,2
87	23,7	18.6	57,7
PVA GF	15.8	-	84.2

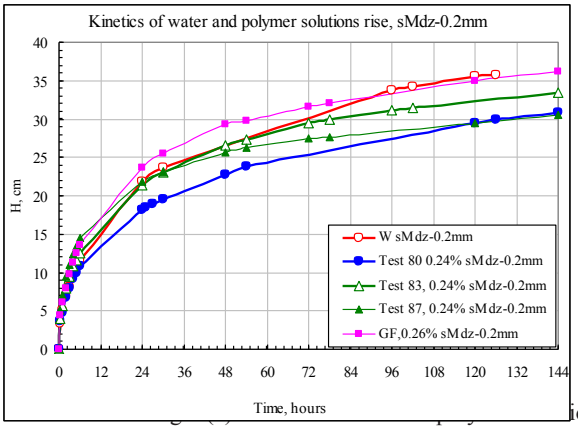
* designations "VA-AA-H" = "Tests"

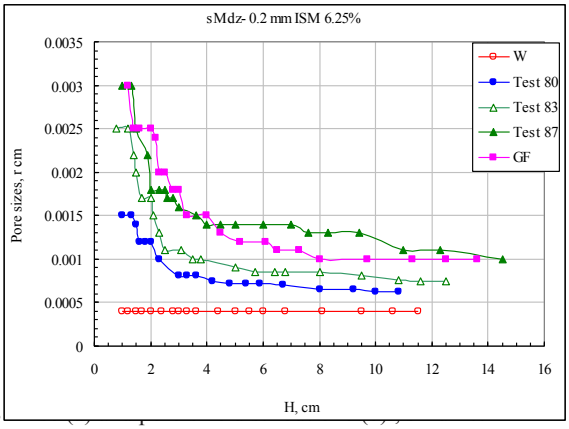
2. Table 4 summarizes the values of parameters in the Washburn equation, determined experimentally for solutions with concentration 0.23- 0.24%.

Table 4 VA-AA- H copolymers, parameters of Washburn

Parameters	Solutions, 0.23-0.24%			
	VA-AA-H 80	VA-AA-H 83	VA-AA-H 87	PVA GF
Surface tension, mN/m	58	71	62	51
Viscosity, mm^2/sec	1,42	1,57	1,38	1.25
Density, d g/cm^3	0,9986	0,9987	0,9989	0.9986

3. Kinetic curves of polymer solutions rise in soil Mdz- 0,2 mm were determined in tubes (Stage 2) (Fig.1a)



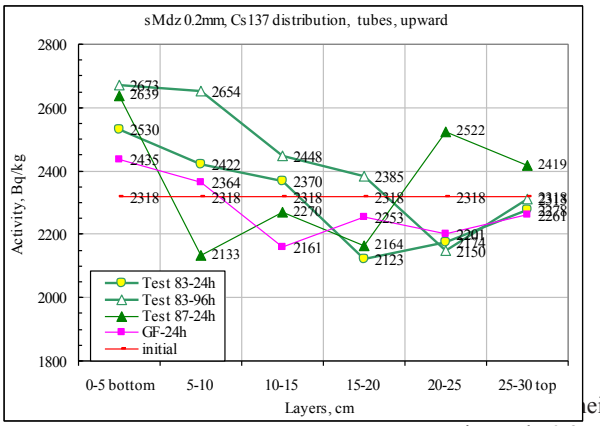


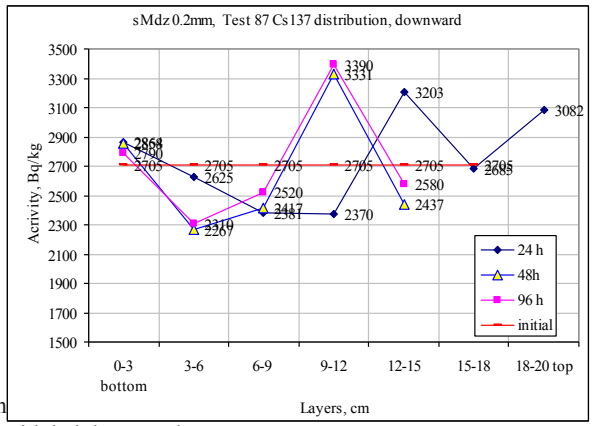
By the kinetic curves the radius of pores, through which capillary rise occurs are defined for each solution. To do this, we build a theoretical curve of the Washburn equation, substituting the experimental data from Table 4, add selecting values for ***r*** so that the experimental and theoretical curves coincided. The contact angle $\boldsymbol{\theta}$ is 89.8^0 (this value obtained according to our numerous definitions)

Pore-size distribution with height (Fig. 1б) shows that up to height of 20-25 cm kinetics rise of solutions 83 and 87 close to the water, however, the polymer solutions penetrate through the larger pores.

4. Polymer solutions 83 and 87, as well as PVA GF are tested in the identical conditions in tubes with contaminated soils at ANPP. 4 tubes were filled with soil; two of them were used for Test 83. Soils in tubes with Test 87, PVA GF and one tube with Test 83 were analyzed after 24 hours, the second one –after 48 hours from experiment beginning (Fig.2a).

Fig. 2(a) shows distributions of specific activity of ^{137}Cs in soil layers depending on duration of experiment.





in sMdz-0.2mm with height, vessel

An unexpected result was that sample 83 promotes a movement Cs down, and a tendency to capillary rise of Cs no observed even after 4 days. Sample PVA GF (low-molecular polyvinyl alcohol) is as similar to 83;

Unlike 83 and GF, sample 87 already in the first 24 hours shown tendency to both gravitational and capillary movement. Hence sample 87 was chosen for the Batch test experiment (Stage 4).

5. Solution VA-AA-H 87 (0.24%) was tested on contaminated soil at ANPP in the Batch experiment, results are plotted in Fig.2b.

Difference ^{137}Cs activity between layers is ~30%, the polymer solution penetrates into the pores only to 10mkm. The equilibrium in the system is observed after 48 hours.

Thus, a methodology was developed for assessing the behavior of polymer solutions in soils, which allows to determine kinetics and height of solutions rise, pore-size distribution, the efficiency of the capillary and/or gravitational Cs movement

To mechanism

1. The main conclusion resulting from the conducted researches: binding Cs with polymer solutions and movement in soil occurs via the same mechanism, regardless of type of clay minerals (montmorillonite, illite, vermiculite).

2. As a possible explanation of this phenomenon at this stage of investigations can be a similarity with the known processes of intercalation and exfoliation of the clays resulting from adsorption of the polymers molecules from its solution.

3. In order to include Cs into polymer solution, Cs connection with the actives centers of clay lattice should be weakened or broken. The roles of the polymeric macromolecules in Cs binding are:

- To weaken a binding Cs with the minerals due to binding with its own groups
- To keep Cs in solution by coordinating with its group and absorption; do not allow the reversible binding with clays;
- Pull out Cs from the inter-packet space and drag into the pores, where capillary and gravitational forces appear.

The tested polymers possess parameters which can compete with Cs for binding with active centers of clay minerals.
From this standpoint, knowledge of the specific location Cs in clay minerals at nano-scale level is one of important factors to enhance efficiency Cs removing from soil.

After Fukushima accident, the Japanese and the other authors more detailed examine the mechanisms of Cs adsorption, especially for pure minerals such as illite, vermiculite, with the assistance of new techniques, such as scanning transmission electron microscopy, energy dispersive X-ray spectroscopy and extended X-ray absorption fine structure spectroscopy (EXAFS).
These methods provide deeper insight into structure of Cs-containing clay particles at the level ~1 nm, and more precise determine active centers of clay minerals. Consequently a more accurate understanding of reasons for weak desorption Cs from illite, vermiculite comes.

The main conclusions from these investigations at present are that Cs cannot be easily removed from the interlayer spaces. Any effective remediation measures must degrade the illite/vermiculite clay fraction. One possibility would be to expand the interlayers by use of a large and highly charged cation or a bulky ligand strongly coordinating Cs.

Comparison of the project results and the latest data on the mechanisms of fixing Cs can promote ways to improve the efficiency of Cs removing from clay minerals.

Evaluation of polymers biodegradation in soil

In Japan, there is a strict classification of biodegradable polymers, presented in Japan BioPlastics Association (JBPA) as GreenPla and BiomassPla.

PVA is known as biodegradable polymer, and is included in the catalog Japan BioPlastics as GreenPla. At the same time, any modifications of PVA continue to be studied; moreover standardization of methods for determining biodegradability, including techniques ISO is in progress.
Samples of PVA and its carboxyl derivatives used in the project generally correspond to the classification GreenPla.

As to the new developed VA-AA-H copolymers systems, they have functional groups which are standard for biodegradable polymeric systems suitable for use in soil.

To assess the extent of biodegradation of the new developed VA-AA-H copolymers in soil we used one of the often conducted studies - effect of soil microorganisms (fungi and bacteria) on the samples. In this case a changes of amount of polymer in soil with time, as well as changes in the composition of soil fungi under the influence of the polymers are studied.
Several studies in the range 14-28-60 days were performed in soils sMdz-2 mm and sANPP-2 mm. In each series of experiments new developed VA-AA-H samples were tested in parallel with PVA grade "Mowiol", which is considered to be fully degradable polymer.

Based on the experiments results it was concluded that polymers undergoes degradation in soil (probably, stepwise conversion of the polymer and destruction), and in line with this a favorable conditions is created in soil for activation of specific species of fungi responsible for specific reactions. It was revealed a dependence of results on soil types as well as on seasons of soils sampling (summer, autumn).

Results for new developed VA-AA-H series are very close to Mowiol.
Reasoning from alterations in compositions of fungi, can be concluded, that there is no dramatic changes in soils from mycology point of view as a result of polymers “intervention”.

Results of experiments demonstrate that the newly synthesized copolymers, according to variation in compositions of fungi, undergo biodegradation by similar mechanisms as Mowiol. So at this stage of studies it can be accepted that VA-AA-H copolymers are not harmful for soil microorganisms.

4. Conclusions

1. Increasing activity of ^{137}Cs, ^{134}Cs in the upper layers of soils influenced by some polymer solutions was found in the radiation-contaminated soils. This phenomenon is due to the manifestation of the capillary movement of polymer solutions in soils.

2. Capillary rise of Cs under action of various polymer solutions was investigated in several soils of various mechanical and mineralogical compositions containing montmorillonite, illite, and vermiculite.

3. Based on these results it was concluded that the mechanism for Cs binding and movement with the polymer solutions is the same for all the studied minerals. The mechanism of this process is suggested.

4. The types of polymeric systems and parameters responsible for ^{137}Cs, ^{134}Cs binding were determined.

5. The technology for synthesis of such copolymers at the laboratory scale was developed and refined in the process of scaling of experiments volumes. A pilot sample was synthesized by the developed technology and used in several Batch tests at ANPP

6. The possibility of a practical application of the observed effects for redistribution Cs in soil with concentration in the narrower layers was examined in the various soils.

7. The methodology for preliminary assessment of polymeric systems in efficiency to bind and move Cs in soils is elaborated.

5. Applicability and Recommendations for Japan

1. The results of the studies have shown the principal possibility of using the revealed regularities of increasing of ^{137}Cs, ^{134}Cs in the top layers of soils under the influence of polymer solutions to reduce volume of contaminated soils, taken off from the fields.

2. Several series of experiments at the ANPP as “Small batch mode tests" defined the main technological parameters and their influence on the efficiency ^{137}Cs, ^{134}Cs redistribution in soil.

3. In Japan, first of all, the project results should be checked on the real contaminated soils. The developed methodology (polymers - tubes -batch mode tests) can be applied for the initial estimations.

4. It is supposed, that a further change in experiments volume (with small soil particles) should be regulated by increasing of vessel height without significantly widening. At this stage all parameters to ensure an effective process are expected to be defined. That is, the " Small batch tests” can be considered as the stage, results of which can be used to predict expected changes in case of further scaling of experiment volume.

5. Analysis of published works relating to mechanisms of Cs adsorption-desorption on illite and vermiculite shows that to improve the efficiency of Cs capillary rise, deeper studies of interaction of polymer molecules with clay minerals and Cs on the molecular level may be needed.

Session 2

Recovery of Agriculture

ISTC-A-2072

Remediation of Cs-contaminated soils through regulation ^{134}Cs and ^{137}Cs soil-plant-transfer by polymeric sorbents

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Inst. Hydroponics Problems (Project Manger) (Armenia)

ISTC/STCU Fukushima Expert Meeting

Tokyo, Japan
November 5-6, 2015

Summary Report – project # A-2072

on work performed from June 01, 2013 to May 31, 2015

Institute of Hydroponics Problems NAS RA
Yerevan, 0082, 108 Noragyugh, Republic of Armenia

Project Manager: **Tadevosyan Anna Hovhannesovna** Ph.D

Title of the Project:	Remediation of Cs-contaminated soils through regulation ^{134}Cs and ^{137}Cs soil-plant transfer by polymeric sorbents
Commencement Date:	01 June, 2013
Duration:	24 month
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List of abbreviations

AAc - acrylic acid
AC - Accumulation Coefficient
ANPP - Armenian Nuclear Power Plant
AP - ammonium persulfate
IF - irrigation frequency
IPN - interpenetrating polymer networks
NF - nourishment frequency
PRIZ - plants' root-inhabited zone
VA - vinyl acetate
VS - vegetation season
v. - village

1. Brief description of work: objectives, and expected results

Objective

The aim of project is to provide new procedures and development of technology for remediation of Cs-contaminated soils with different contamination levels through regulation of biological migration of polymeric sorbents in the *water – soil – plant* system.

The study includes (Flow Chart):

Task 1: Development of methods for the synthesis and study on the properties of highly effective polymer sorbents of radionuclides from the soil

Task 2: Assessment of the effectiveness of polymers on growing plants in the zones of low and high radioecological tension

Expected results

Project main outcomes will demonstrate development of new models for stage-by-stage remediation of agricultural lands in and around Fukushima contaminated by radionuclides, especially by ^{134}Cs and ^{137}Cs:

- In areas with low level of Cs-contamination by growing plants with compositions/polymeric sorbents which prevent migration of Cs in soil-plant system and give an opportunity to grow and obtain harmless and radioecologicaly sound biomass of Japanese basil-Shi-so.
- In areas with low and medium level of Cs-contamination by co-growing system of food-plants (Shi-so) and Cs-hyperaccumulator-plants applying corresponding compositions/polymeric sorbents and polymer types.
- In the areas with high level of Cs-contamination by phytoremediation with Cs-hyperaccumulator plants and applying polymer materials which contribute to Cs transfer from soil to plants.

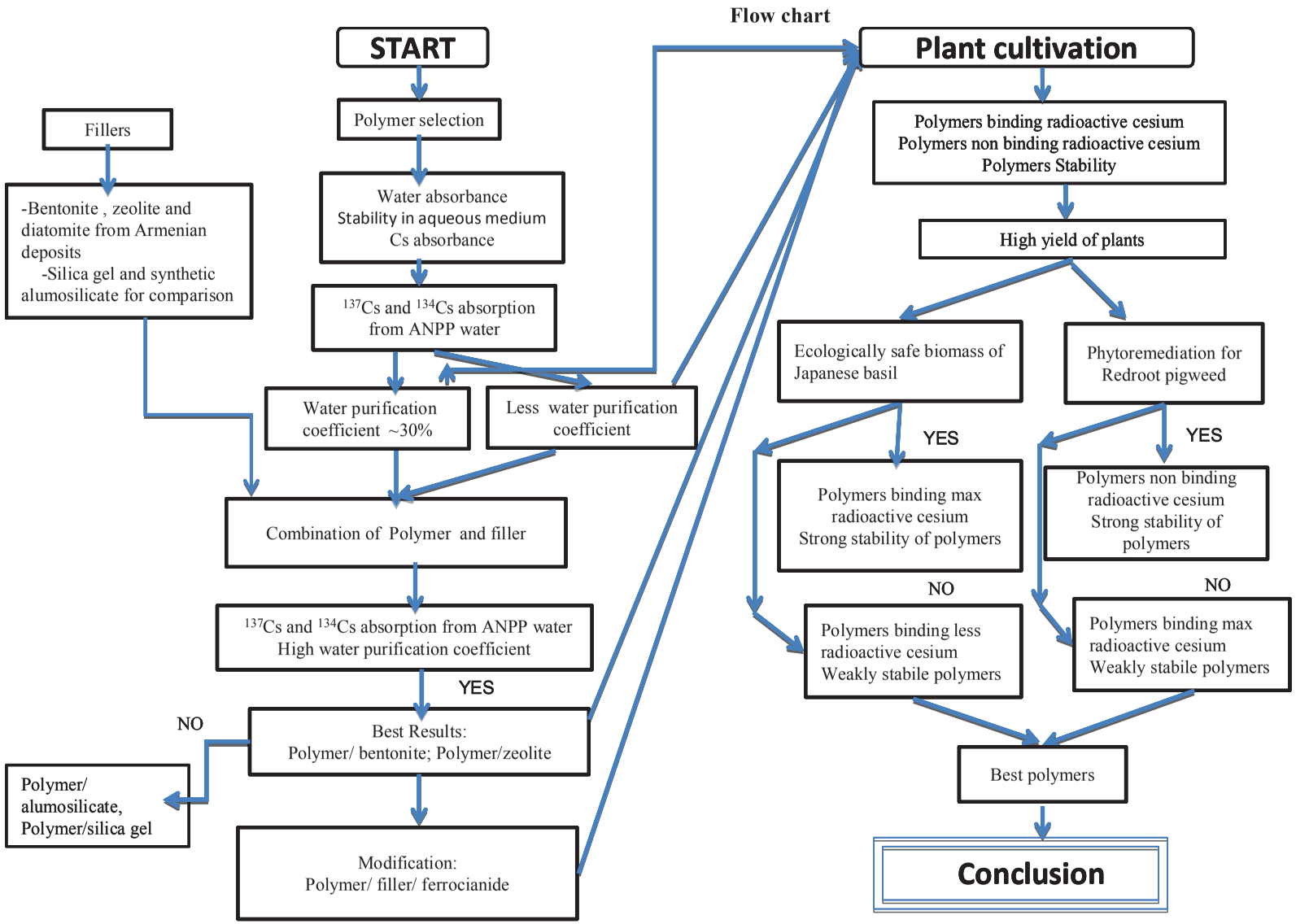
Modifications of work after April 2014 meeting

We have considered to the comments from experts.

1) **Toxicology of polymer should be addressed.**

The suggested polymer materials cannot show adverse effects or unintended consequences.

- Polymeric sorbents are: nontoxic; dust-free/non dust-forming; can be easily removed from the soil with the plant root system during harvest.
- Components (bentonite, zeolite, ferrocyanide) making the content of compositions are nontoxic.
- The synthesized polymeric materials have a three-dimensional/cross-linked structure, and they are insoluble in water and soil.
- For Sample 13, as the most thoroughly studied sorbent, the test for acute toxicity (ISO 10993-11) was performed.
  The Scientific Center of Radiation Medicine and Burns (Yerevan, Armenia) provided technical assistance in studying acute toxicity of Sample 13 at a dose of 2g/kg in white inbred rats (by 5 animals, oral administration). No unfavorable effects were observed during 24 hours after administration of the single dose. In 15 days after administration no difference in weight, behavior, or appearance was recorded between the animals of experiment and control groups.


Flow chart
START
Polymer selection
Water absorbance
Stability in aqueous medium
Cs absorbance
137Cs and 134Cs absorption from ANPP water
Water purification coefficient ~30%
Less water purification coefficient
Fillers
-Bentonite , zeolite and diatomite from Armenian deposits
-Silica gel and synthetic alumosilicate for comparison
Combination of Polymer and filler
137Cs and 134Cs absorption from ANPP water
High water purification coefficient
YES
NO
Best Results:
Polymer/ bentonite; Polymer/zeolite
Polymer/ alumosilicate, Polymer/silica gel
Modification:
Polymer/ filler/ ferrocianide
Plant cultivation
Polymers binding radioactive cesium
Polymers non binding radioactive cesium
Polymers Stability
High yield of plants
Ecologically safe biomass of Japanese basil
Phytoremediation for Redroot pigweed
YES
YES
Polymers binding max radioactive cesium
Strong stability of polymers
Polymers non binding radioactive cesium
Strong stability of polymers
NO
NO
Polymers binding less radioactive cesium
Weakly stabile polymers
Polymers binding max radioactive cesium
Weakly stabile polymers
Best polymers
Conclusion

2) The polymer absorption of Cs will also lead to absorption of potassium from fertilizer or soil.

Properties of polymers removed from the soil and hydroponic substance were evaluated at the end of vegetation period (Table 1).
Polymers retained the ability to absorb water; the degree of swelling was higher for all samples used in hydroponic conditions.
All polymer samples removed from the soil after vegetation period differed from the initial ones and were characterized by lower content of potassium (Table 1). The obtained results confirmed that the used polymers did not recover potassium from soil.
At hydroponics conditions (Table 1) a slight increase was observed for the potassium content of the Sample 73* and 13. This effect might be explained, because in hydroponics the nutrient medium contains an excess of potassium ions and in the aqueous medium there occurs a constant ion exchange.

Table 1. Properties of polymer compositions after basil harvest

Sample code	Q, g/g initial	Q, g/g		K ion in initial polymer, mg/g	K ion in polymer, mg/g	
		soil	hydroponic		soil	hydroponic
73*	58.3	52.0	87.5	1.36	0.4	3.0
13*	2.4	3.5	11.5	6.2	0.6	2.2
13	5	3.25	8.0	1.76	1.2	2.4

Thus, the use of polymers 73*, 13*, and 13 for basil growing does not lead to the recovery/removal of "useful" potassium from soil or fertilizer.
Additional amount of nutrient solution is not required under the hydroponics conditions, if polymer sorbent is used. The hydroponic 1L nutrient solution contains 310 mg K, and during the whole vegetation period we used 1600 L hydroponic nutrient solution (496g K) and 30 g polymer for $2m^2$ hydroponic equipment. In fact, at the end of vegetation period 1 g polymer absorbed only 0.01% of the whole potassium.

3) Investigate the use of polymers and different inorganic materials in a Japanese rice paddy, and develop a method to remove the polymer after its use. For example, use a sheet of polymer and remove it after rice is harvested.

We have developed a way to use polymers that can be successfully applied to rice fields. Polymers wrapped in latticed cloth in shape of ribbons. After rice harvest the ribbons can be easily extracted from soil without any loss of polymer (Fig. 1).

Figure 1. Sample 13 polymer wrapped in latticed cloth

Experiments were carried out in 2 Groups (1st Group with and 2nd Group without rice plants) by the following variants:

1. Control variant - Cs-contaminated soil without polymer in it.
2. Test variant - Cs-contaminated soil with Sample 13 polymer in it.
3. Test variant - Cs-contaminated soil + Sample 13 polymer in water.
4. Test variant - Cs-contaminated soil with Sample 73* polymer in it.

Soil initial radioactivity ≈ 2870 Bq/kg.
Unfortunately, all our attempts of rice cultivation failed. After sprouting and reaching 10-15 cm height they dried.
After four months polymers of 2nd Group (without rice plants) were removed from Cs-contaminated soil and submitted for radiochemical analyses. The results showed that Sample 73* polymer distinguished by the maximum absorption of cumulative radioactive cesium, which was higher than Sample 13 and Sample 13* polymer by 2.1 and 1.3 times, accordingly (Table 2).

Table 2. ^{137}Cs and ^{134}Cs absorbance ability of different polymers after being in Cs-contaminated soil for 4 month, Bq/kg

N^{O} Test variant	Polymer types	^{137}Cs	^{134}Cs	$\Sigma^{137+134}$Cs	$\Sigma^{137+134}$Cs uptake from soil, %	$\Sigma^{137+134}$Cs uptake from water %
2.	Sample 13 polymer in soil	317 ± 25	203 ± 16	520	>18	>60
3.	Sample 13 polymer in the water	527 ± 42	316 ± 25	843	>29	>60
4.	Sample 73* polymer in soil	716 ± 57	385 ± 31	1101	>38	>85

Soybean cultivation. Studies of soybean cultivation were carried out in normal (Taronik land) and Cs-polluted soil.
1st Group.
1. Control (soil from v. Taronik without polymer).
2. Test variant - soil from v. Taronik + Sample 13 polymer, 1g/pot.
2nd Group.
1. Control: Cs-contaminated soil, without polymer.
2. Test variant: Cs-contaminated soil + Sample 13 polymer, 1g/pot.
As the plants were grown in winter and early spring period under indoor conditions in the small pots (there is no greenhouse), then the accumulated biomass of plants was too small.
In two variants of 1st Group radioactive cesium was not found in plants organs as well as in used polymer.
The results of 2nd Group tests showed that radioactive cesium concentration in soybean seeds in the control was: **^{137}Cs**=150Bq/kg and **^{134}Cs**=130Bq/kg while in the seeds of polymer variant radioactive cesium was not found. Used Sample 13 polymer accumulated 81Bq/kg **$^{137+134}$Cs.**
The results of our initial experiments showed that obtaining of ecologically sound biomaterials is possible in Cs-contaminated soils by the use of polymeric sorbents.
For performing more detailed research on soybean and obtainment of reliable results it is necessary to continue our investigations in a broader framework and outdoor conditions (under natural climatic conditions).

Technical approach, method, experiments, theory, etc

Technical Approach

The classical stage-by-stage scheme for selection of polymer materials was used in the project:

- synthesis; selection of polymers according to physical and chemical properties. The sorbent compositions were obtained using inorganic fillers.
- growing different plants varieties in field and hydroponic conditions with and without application of polymer in PRIZ in different radio-ecological tension zones (with the radius of 2-15 and 30km from the ANPP).
- determination of both quantitative and qualitative productivity of plants depending on the presence (control-without presence of polymer) and type of polymer material and water regimen.
- determination of radionuclides content in a system *water – soil – plant* in the vicinity of the ANPP (zone radius = 2-15km), as well as under hydroponic conditions of (zone radius = 30km) in a system *water – nutrient solution – plant*.
- determination of migration and accumulation of radionuclides in soil layers if different depth (0-5; 5-10; 10-20cm), below-ground and above-ground parts of plants depending on application of the polymer; revealing the dependence of redistribution of radionuclides in systems *water – soil – plant* and *water – nutrient solution – plant* on the type of a polymer material, variety of a plant and number irrigation.

In this project we offer technologies for radioactive Cs minimization in soil and water by polymer systems that can absorb cesium without any transfer to the plant.
In the system *water – soil – plant* the polymer might play two role:

- bind (or extract) the radionuclides;
- non-bind the radionuclides.

Selection of polymers was done on the basis of the following parameters:
1. Sorption (uptake) of radioactive cesium from contaminated water;
2. Desorption of radioactive cesium from the Cs-contaminated polymer sample into pure water.

Methods

Determination of ^{137}Cs and ^{134}Cs uptake

Low-background gamma spectrometer (Canberra, USA) with pure Ge detector and supporting "GENIE" software was used to measure the radioactivity of ^{137}Cs and ^{134}Cs. The uptake was evaluated at room temperature after 24 hours. For each experiment, 0.2 g adsorbent and 100 ml of radio-contaminated water from the ANPP were applied.

The uptake of radioactive Cs in per cent (%) was assessed as follows:

$$Uptake,\% = \frac{Ai - Af}{Ai} \times 100 \qquad (1)$$

where $\mathbf{A_i}$ is the initial water activity in Bq/L and $\mathbf{A_f}$ is the final water activity 24 h after polymer removal of, in Bq/L, respectively

Determination of radioactive cesium desorption

Following the absorption of the radionuclides, the polymer was dried to a constant weight, placed in pure water and preserved for 24 hours. Afterwards, the water was drained from beaker, and the content of radionuclides in water was determined.

For the investigations 2 plant spices were chosen:

1. for food: Japanese basil - Shi-so (*Perilla frutescens* v. *crispa*). Perilla (shiso [紫蘇, しそ, シソ]) is herb of Japanese extraction. It is commonly cultivated as a vegetable in Japan. The green, Japanese basil leaf which can be served as tempura, in salads and with sashimi.
2. Redroot pigweed (Amaranth*us retroflexus*). This plant is known as one of the Cs-hyperaccumuliator (up to 3000 Bq kg^{-1}) plants (with the corn, reed canary grass, tepary Beans, sunflower, *Brassicaeae).*

The experiments were carried out in 2 zones of radio-ecological tension:

1. Zone of low radio-ecological tension, zone radius = 30km far from the ANPP, outdoor hydroponic conditions (Yerevan, Institute of Hydroponics Problems).
2. Zone of high radio-ecological tension, zone radius = 0-7 km far from the ANPP:

3.1 Polymer sorbents selection

We report a newly developed radioactive cesium sorbents: 1) Polymers with inorganic cation; 2) Polymer compositions with inorganic fillers.

Water-swelling and water-retaining polymeric gels are effective moisture absorbers and retainers for growth of agricultural plants. The ability of polymer gels that absorb and then release water is widely used for plants cultivation in conditions of arid land and hydroponics. Earlier, in A-1671 project, we have shown the effect of water-retaining copolymers on the transport of cesium and strontium radionuclides in plants. The main objective of A-2072 project was development and selection of effective polymer sorbents for remediation of Cs-contaminated soils.

3.1.1 Polymers with inorganic cation

Methods of polymers obtaining

Water-retaining polymers for multiple application in water sorption and desorption processes were synthesized by radical polymerizations of vinyl monomers in aqueous medium.

-*Copolymers of VA with AAc as film materials*

The polyelectrolyte polymers containing Ca^{2+}, NH_4^+ and K^+ ions were formed from copolymerized VA and AAc cross-linked with a polyfunctional reactant. Acrylic acid was partially neutralized with alkali solutions. Allyl-glycidyl ether was used as a cross-linking agent. After the synthesis of copolymers dispersions those films were prepared. Their water swellings achieved 100g/g. Degree of water swelling of cross-linked VA/AAc copolymer reduced with the increase of electrolytes concentration.

-*Ammonium acrylate gel as solid particles (size 1-3 mm)*

Hydrogel Preparation: Different amounts of acrylic acid, deionized water, and redox reagents as initiator [AP + NH_3] were mixed at 80°C. After 1 h, hydrogels were cut to 15-20 mm size shards and were dried at room temperature. Water swellings of ammonium acrylate gels made 100-150 g/g.

Ammonium acrylate gels were prepared through a variety of processing conditions. The effects of initiator concentration and monomer were investigated. Ammonium acrylate gel Sample 11 (encoded notation) synthesized at molar ratio of redox reagents C_{AP}/C_{NH3}=0.012/0.1 was selected as high resistant/strong in aqueous medium. Sample 11 is stabile in several cycles of swelling in water and hydroponic nutrient medium.

Uptake of radioactive cesium from Cs isotopes-contaminated water

Water purification was used for indirect proof of adsorbing property of proposed materials. Experiments on sorption of radioactive cesium from water were performed under static condition. Polymer (0.2 g) was placed in 100 ml contaminated water and left for 24 hours contamination.

Purification coefficient =$A_{initial}$ /A_{final}, where

$A_{initial}$[Bq/L] is initial water activity,

A_{final}[Bq/L] is water activity after a day (after removal of polymer).

Measurements relative error was 4-6 %. T= 20±1.0°C

Radioactive cesium adsorption of ammonium acrylate gel was insignificant (Sample 11), and film VA/AAc (% ratio 56/44) copolymer (Sample 3) showed water purification coefficient at the level of 1.5 (Figure 2). The VA/AAc copolymer adsorbent takes up cesium ions from the aqueous solution based on ion-exchange. Figure 3 illustrates uptake of radioactive cesium from Cs-contaminated water by copolymers with all ions used. The uptake of cesium isotopes from water was approximately 30% of the starting concentration.

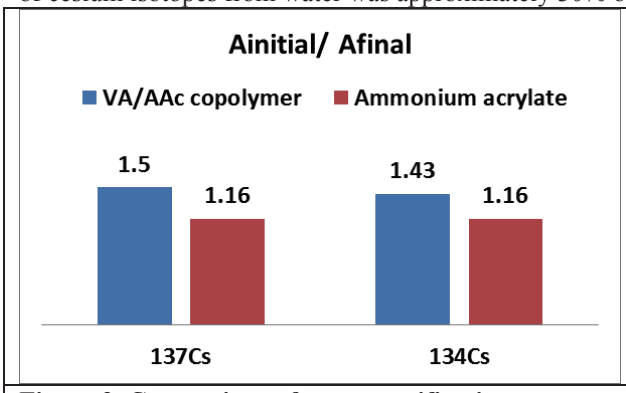


Figure 2. Comparison of water purification coefficient of VA/AAc copolymer film and ammonium acrylate gel

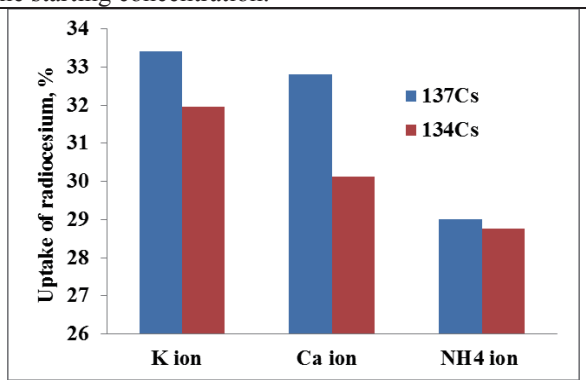


Figure 3. Uptake of radioactive cesium from water by VA/AAc copolymer films

Polymer = 0.2 g; ANPP water =100 ml; Time 24 h; T = 20±1.0°C

The initial water radioactivities were 18600 Bq/L for ^{137}Cs and 21750 Bq/L for ^{134}Cs, respectively

Thus water- retaining VA/AAc copolymers (% ratio 56/44, neutralization degree of AAc is 3.6 % for all ions) can absorb ^{137}Cs and ^{134}Cs from radio-contaminated water.

3.1.2 Polymer compositions with inorganic fillers

Composites consisting of synthetic polymers with inorganic fillers were prepared:

- Composites on the base of VA/AAc copolymer in the film form;
- Poly(acrylic)acid-based compositions in the form of solid particles and film form;
- Ammonium acrylate gel compositions in the form of solid particles.

To develop all compositions, we added the filler into reaction mixture during polymerization process. Fillers from Armenian deposits (bentonite, zeolite, and diatomite) and Silica gel (for comparison) were used. The properties of filled polymer materials depend on the content of polymer matrix, the type, and quantity of the filler.

Uptake of radioactive cesium from Cs isotopes contaminated water

Irrespective of the polymer matrix type, the filled polymers were characterized by high water purification coefficients for ^{137}Cs and ^{134}Cs (Figure 4).

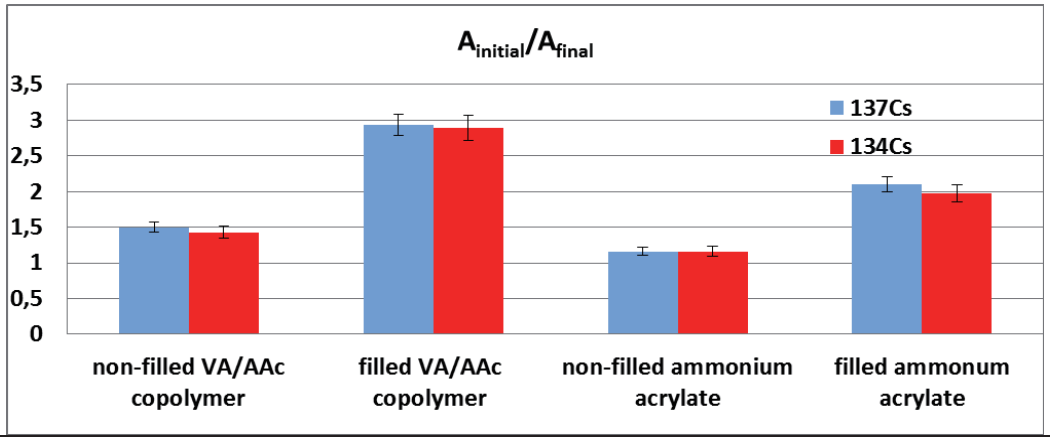


Figure 4. Comparison of water purification coefficients: non-filled and filled polymers
Composition = 0.2 g; ANPP water =100 ml; Time 24 h; T = 20±1.0°C
The initial water radio-activities were 18600 Bq/L for ^{137}Cs and 21750 Bq/L for ^{134}Cs, respectively

Cs-uptake by composites on the base of VA/AAc copolymer

Without any filler, the uptakes of radioactive cesium by VA/AAc copolymers (% ratio 56/44) were approximately 30 %. Filled samples of VA/AAc copolymer with Ca^{2+}, NH_4^+ and K^+ ions have high radioactive cesium adsorption in comparison with non-filled samples and achieved 65% (Figure 5).

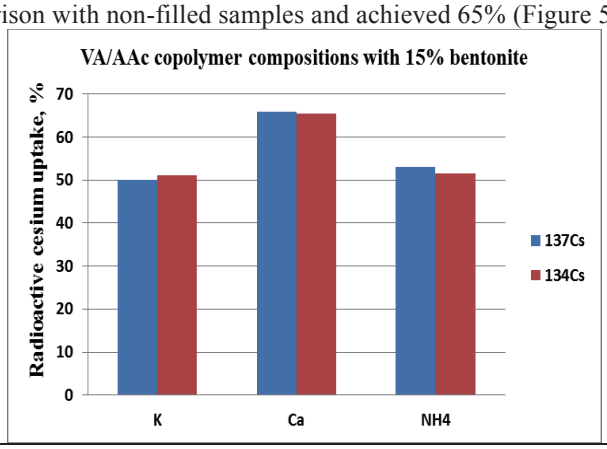


Figure 5. Radioactive cesium uptake by VA/AAc copolymer/Bentonite compositions
Composition = 0.2 g; ANPP water =100 ml; Time 24 h; T = 20±1.0°C
The initial water radio-activities were 37200 Bq/L for ^{137}Cs and 36500 Bq/L for ^{134}Cs, respectively

The water-stable composition of VA/AAc (% ratio 56/44) copolymer with 15 wt % filler was selected for further studies, as the water purification coefficient was the highest.

The addition of ferrocyanide increases adsorption of radioactive cesium from water by the VA/AAc copolymer/Bentonite compositions. The VA/AAc copolymer/Bentonite/ Ferrocyanide composition (mass ratio bentonite to ferrocyanide - $K_4Fe(CN)_6$ is 10 : 0.75) uptake achieves 90% (Figure 6).

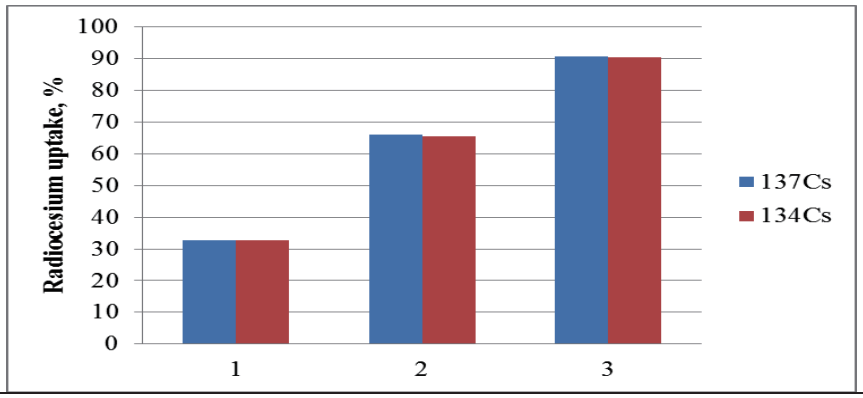


Figure 6. A comparsion between radioactive cesium uptake of 1-VA/AAc copolymer; 2-VA/AAc copolymer/Bentonite and 3-VA/AAc copolymer/Bentonite/ Ferrocyanide

Sorbent = 0.2 g; ANPP water =100 ml; Time 24 h; T = 20±1.0°C

The initial water radio-activities were 28070 Bq/L for ^{137}Cs and 24270 Bq/L for ^{134}Cs, respectively

Figure 5 and 6 illustrated that the adsorption of radioactive cesium from water increased as

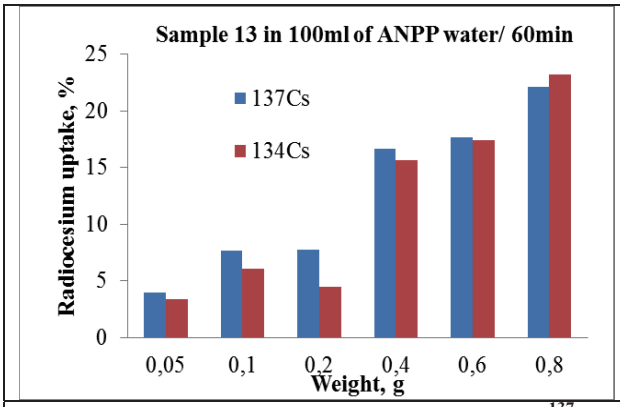


Figure 7. Effect of film Sample 13 amount on ^{137}Cs and ^{134}Cs adsorption during 1h

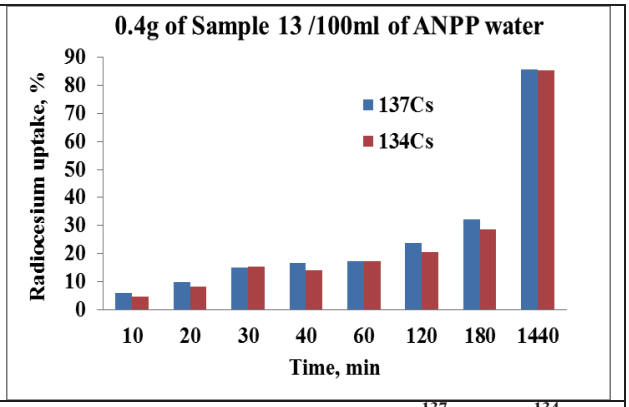


Figure 8. Effect of contact time on ^{137}Cs and ^{134}Cs adsorption by 0.4g of Sample 13

Films with 0.4-0.5 mm thickness; Water from ANPP=100 ml.; T= 18±1.0°C

The initial water radio-activities were 4068 Bq/L for ^{137}Cs and 2262 Bq/L for ^{134}Cs, respectively

follows:

Non-filled polymer < Filled polymer < Filled polymer+ ferrocyanides.

Cs-uptake by poly(acrylic) acid-based compositions

- IPN hydrogels were synthesized by polymerization of AAc in water solution of other polymer without the cross-linking agent. The process of polymerization was performed in the presence of fillers. Polymer compositions consisting of bentonite (20-30%) were shown efficient for Cs contaminated water purification. Adsorption of radioactive cesium from water achieved 60%.

Data obtained shows that film of composition Sample 13 (encoded notation) is specified by high sorption characteristics relatively to radionuclides ^{137}Cs and ^{134}Cs.

As evident from Figure 7, during 1 hour the significant radioactive cesium absorption (above 17%) was observed for 0.4g sorbent at room temperature. Therefore, the optimized solid to liquid ratio of 0.04g ml^{-1} for Sample 13 was selected to study dependence on contact time.

The effect of contact time is shown in Figure 8. The uptake of radioactive cesium increased with time, and in 3 hours it achieved 30%; in 24 hours the uptake made 85%. Based on obtained research findings, for further measurements we selected contact time of 24 h.

Radioactive Cs "sorption-desorption" by Sample 13

Sample 13 in film form takes up ~62% of radioactive cesium from ANPP water (Table 3). After radioactive cesium adsorption without drying Sample 13 was placed in clean water and preserved for 24 hours, and the content of radionuclides in water was determined. The averaged results of measurements demonstrated ~6 % desorption of radioactive cesium.

Table 3. Results of radioactive cesium *"sorption-desorption"* process by film Sample13

0.1 g of Sample 13 in 100 ml of medium; Time = 24 h

Sorption from contaminated water Contact time: 24 h				**Desorption into pure water** Contact time: 24 h			
Water from ANPP ^{137}Cs $A_{initial}$ =12950Bq/L and ^{134}Cs $A_{initial}$ =10300Bq/L				Deionized water			
A_{final}, Bq/L		Uptake,%		A_{final}, Bq/L		Desorption,%	
^{137}Cs	**^{134}Cs**	**^{137}Cs**	**^{134}Cs**	**^{137}Cs**	**^{134}Cs**	**^{137}Cs**	**^{134}Cs**
4900	3900	62.2	62.1	520	425	6.5	6.6

Thus, Sample 13 is an effective sorbent of radioactive cesium ions from aqueous solutions in static conditions. Polymer compositions consisting of Sample 13 and ferrocyanide were shown efficient for Cs-contaminated water purification. We experimentally determined mass ratio of of polymer to bentonite to ferrocyanide as 32.5 : 10 :

0.75. Sample 13* (encoded notation) was prepared by addition of ferrocyanide to Sample 13. The adsorption of radioactive cesium from water by Sample 13* achieved 80%.

In conclusion, despite the type of polymer matrix [VA/AAc copolymer or Poly (acrylic) acid)] the additions of ferrocyanide increased adsorption of radioactive cesium from water by 20 %.

- Some of polymer gels were synthesized by the method of room-temperature polymerization of AAc in the presence of the cross-linking agent and filler (bentonite). These compositions have high-efficiency uptake for ^{137}Cs and ^{134}Cs from water. The search for adsorbents obtained through this technique will be continued in future as a follow-up of our investigations.

Cs-uptake by composites on the base of ammonium acrylate gels
The ammonium acrylate compositions in the form solid particles in size 2-3 mm were used.
Without any filler, the uptake of radioactive Cs by ammonium acrylate hydrogel was low and made about 13%. Inorganic filler increased the uptake of radioactive Cs. The uptake by filled ammonium acrylate hydrogels depended on the filler type and quantity.
Figure 9 shows the effect of filler amount. The optimal quantity of filler was found to be 30 % for ammonium acrylate gels. Figure 10 shows the effect of filler type. Samples with silica gel and diatomite demonstrated low absorptive capacity. The addition of zeolite or bentonite to ammonium acrylate matrix demonstrated the maximal uptake of ^{137}Cs and ^{134}Cs ions reaching about 50%. Thus, both ammonium acrylate hydrogels filled with bentonite or zeolite showed good results of cesium adsorption from water.

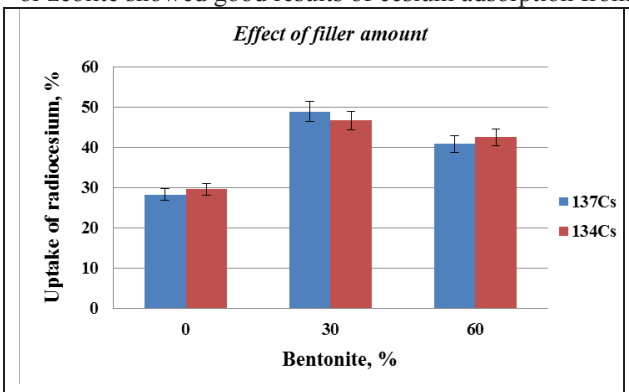


Figure 9. ***Effect of filler amount:*** **Radioactive cesium uptake by Ammonium acrylate/Bentonite composition**

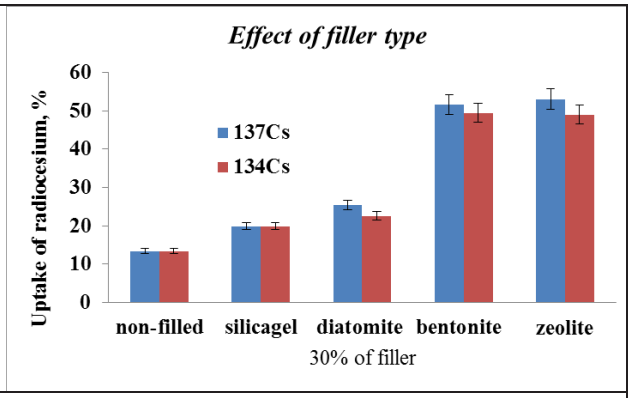


Figure 10. ***Effect of filler type:*** **Radioactive cesium uptake by compositions with ammonium acrylate and 30% fillers**

Composition = 0.2 g; ANPP water =100 ml; Time 24 h; T = 20±1.0°C

The initial radio-activities were 3530-6872 Bq/L for ^{137}Cs and 2620-6348 Bq/L for ^{134}Cs, respectively

Radioactive Cs desorption from ammonium acrylate compositions
An important feature of the radionuclides adsorbent is its stability to leaching by water. The cesium desorption in water was done for compositions of ammonium acrylate with both bentonite and zeolite by high ^{137}Cs and ^{134}Cs uptake (Table 4).
Cesium desorption occurred in both case. It reached 10-15% desorption, and thus the composite retained no larger than 90 % of the absorbed radioactive cesium. The results showed that polymer compositions with bentonite or zeolite effectively absorbed radioactive cesium and fixed the significant amount of radioactive cesium inside gels.

Table 4. The results of study on radionuclides desorption from filled ammonium acrylate

Filler	Activity of polymer, Bq		Activity of water, Bq		Desorption, %	
	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs
bentonite	1.08×10^3	1.23×10^3	0.16×10^3	0.19×10^3	14.81	15.44
zeolite	1.28×10^3	1.14×10^3	0.13×10^3	0.11×10^3	10.15	9.65

3.1.3 Selected polymer materials for plant growing

Table 5 . Summary characteristics of Samples for plant growing (1st and 2nd VSs)

Sample code	Chemical composition	Filler	Form of sorbent	^{137}Cs uptake from water, %	^{134}Cs uptake from water, %	Water swelling Q, g/g	Stability in cycles	Efficiency for plant growing in soil/hydroponics	Concentration ^{137}Cs in basil leaves or whole Redroot pigweed biomass
3	VA/AAc copolymer	none	film	32.8	30.0	95±5	7-8***	good for soil and hydroponics	Increases up to 5.3 times in hydroponics and has no effect in soil
4	Ammonium acrylate	zeolite	1-3 mm solid particles	53.0	49.0	40±5	5-7**	good for soil and hydroponics	has no effect in hydroponics and decreases 2.3 times in soil
11	Ammonium acrylate	none	1-3 mm solid particles	13.4	13.4	30±5	5-7	good for soil and hydroponics	Increases up to 15.1 times in hydroponics and has no effect in soil
12	Ammonium acrylate	bentonite[1]	1-3 mm solid particles	52	49.4	54±3	5-7	good for soil and hydroponics	Decreases 3.2 times in hydroponics and 2.2 times in soil
13	IPN gel	bentonite	film	63.0	62.0	6±1.5	more 100	good for soil and hydroponics	Decreases up to 6.7 times in hydroponics and up to 5.6 tims in soil for basil leaves, Increases up to 3.3 times for pigweed in hydroponics and has no effect in soil
13*	IPN gel	bentonite	film	81.2	81.2	3.0±0.5	more 20	good for soil and hydroponics	Decreases 8.7 times in hydroponics and 9.5 times in soil
73*	VA/AAc copolymer	bentonite	film	86.8	84.5	60±2	8-10	good for soil and hydroponics	Increases 1.7 times in hydroponics and decreases 6.8 times in soil

*+ ferrocyanide

**When stored in water up to 6 months Sample 4 almost dissolved, when removed from the soil or substrate after vegetative period sticks to the roots/substrat

[1] Particle size is 1-2 mm, specific surface area is 63.8m^2/g

3.2 Assessment of the effectiveness of polymers in plants hydroponic culture

Closed hydroponics system (regulated artificial medium) allows controlled observation (model tests) of the effects of polymers on growth and productivity, as well as radio-biochemical characteristics of plants. The physical-chemical properties of soil can't play any role in this situation and can't influence on behavior of investigated polymer. And we can evaluate the real properties of tested polymers.

The automatic hydroponics equipment with 2 m^2 nutrient surfaces was used for plant cultivation under hydroponic conditions. Gravel+volcanic slag substrate (relations of 1:1) was used. The plants were supplied with Davtyan's nutrient solution, pH=5.5-6.5. Different polymers (1 g/plant) were applied in PRIZ during planting of seedlings.

3.2.1 Japanese basil

The experiments were carried out in the following variants:
1st VS:
1. Control-without applying polymer, nourishment frequency (NF) twice a day.
2. PRIZ + Sample 4, NF once a day.
3. PRIZ + Sample 12, NF once a day.
4. PRIZ + Sample 13, NF once a day.
2nd VS:
1. Control-without applying polymer, NF twice a day;
2. PRIZ + Sample 13, NF once a day (repitation of 2013 tests).
3. PRIZ + Sample 13*, NF once a day.
4. PRIZ + Sample 73*, NF once a day.
(As a result of the first vegetation period for basil we have chosen Sample13 as the best polymer and as the Research group of Plastpolymer synthesized new polymers (Sample 13* and 73*), which are distinguished with the higher absorption of radioactive cesium from radioactive water, therefore we found it advisable to test them in the second vegetative period, as well as to repeat the experience with the Sample 13)

The test results showed that there was no significant difference among all variants with regard to leaves yield, though the nourishment of plants with polymers reduced twice. Only leaves yield of Sample 12 reached 1.3 times as the control plants and 1.4 – 1.6 times as the other polymers. In polymer variants essential oil, β-karotin and C vitamin content in leaves were the same or higher compared to the control variant.
In fact the use of polymers in Japanese basil hydroponic culture creates an opportunity to decrease the nourishment frequency twice and ensure the same and even high yield of leaves (for food) with high quality indices.
The lowest ^{137}Cs concentration in the leaves was observed in Sample 13 and 13* polymer variants, the highest ones in Sample 4 (1st VS) and 73* (2nd VS) polymer variants. It means that the use of Sample 4 and 73* polymers enhanced the transfer of ^{137}Cs from roots to leaves (Figure 11).

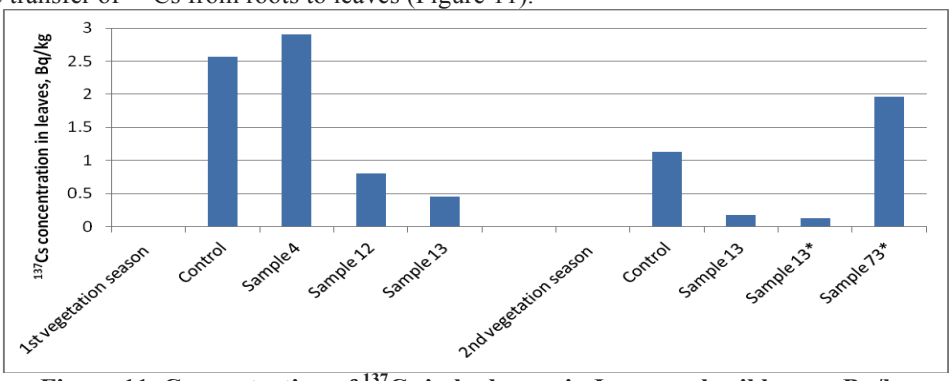


Figure 11. Concentration of ^{137}Cs in hydroponic Japanese basil leaves, Bq/kg

In the control variant ^{137}Cs concentration in the leaves was higher than those in Sample 13 and 13* polymer variants 5.7 (1st VS)-6.3 (2nd VS) and 8.7 times, respectively. The roots of control showed the lowest uptake of ^{137}Cs both in 1st and 2nd VSs compared to all polymer variants.
In 2nd VS ^{137}Cs uptake by basil per plant in the control was 3.1 times higher than in Sample 13 and 13* polymer variants. Sample 73* polymer variant was distinguished by the highest AC of ^{137}Cs in all organs of Japanese basil (leaf, stem and root).

Thus, the use of Sample 12 and 13 polymers in PRIZ prevented ^{137}Cs accumulation in basil above-ground part, moreover Sample 13 was more effective. It can be concluded that the use of Sample 12 and 13 polymers will support getting more ecologically sound biomass for food.

3.2.2 Redroot pigweed

The experiments were carried out in the following variants:

1st VS:

1. Control-without applying polymer, NF twice a day.
2. PRIZ + Sample 3, NF twice a day.
3. PRIZ + Sample 11, NF twice a day.
4. PRIZ + Sample 13, NF twice a day.

2nd VS (we reduced the number of testing variants by recommendation of Expert Group)

1. Control-without applying polymer, NF twice a day.
2. PRIZ + 0.5g Sample 13 + 0.5g Sample 11, NF twice a day

   (in 1st VS Sample 13 and Sample 11 showed the same level of ^{137}Cs accumulation in above ground part of Redwood pigweed in soil, but Sample 11 was very effective under hydroponics -^{137}Cs accumulation in above- ground part was 4.6 times higher compared to the Sample 13). It was very interesting from a scientific point of view to know how the result of their joint application will be).

The results showed that there was no significant difference among control and polymer variants both in 2 VSs.
It was found out that in 1st VS in polymer variants ^{137}Cs concentration in different organs of Redroot pigweed was higher than those in the control: in above-ground part by 3.3-15.1, in the roots by 1.3-1.5 times. That is, the polymers promoted the accumulation of ^{137}Cs in above-ground part, besides Sample 11 showed higher performance. It is cleared up that ^{137}Cs was mainly accumulated in the shoots of pigweed only in Sample 11 variant, where ^{137}Cs concentration exceeded roots 1.3 times.
The data of Fig. 12 showed that ^{137}Cs uptake by pigweed per plant in polymer variants was higher than in the control by 2.8 – 8.8 times. Also in polymer variants, ^{137}Cs uptake by above-ground part was higher than in the control 3.8 – 13.1 times. In Sample 11+13 variant ^{137}Cs uptake by Redroot pigweed per plant was 11.3 times lower compared to the control.

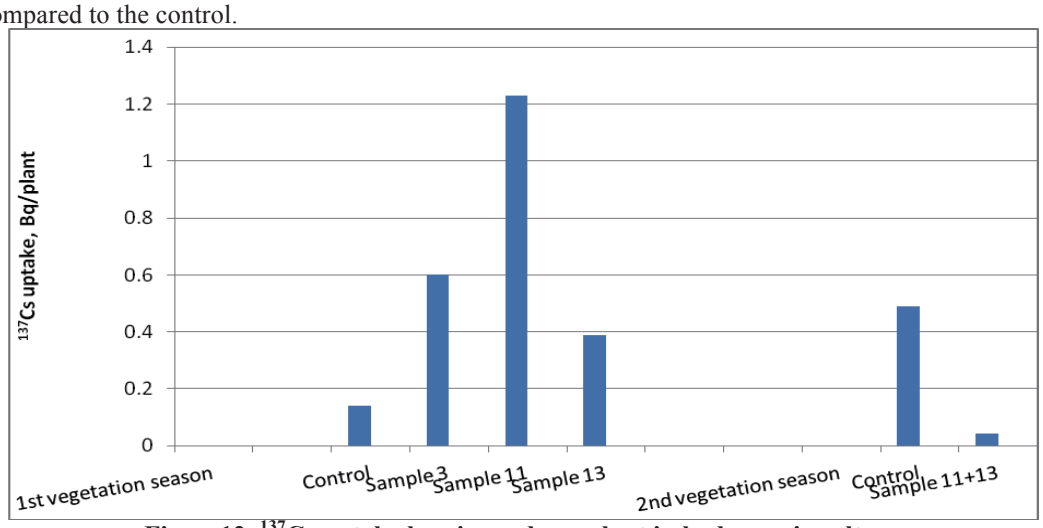


Figure12. ^{137}Cs uptake by pigweed per plant in hydroponic culture

Thus, the separate use of polymers in PRIZ promoted the accumulation of ^{137}Cs in Redroot pigweed biomass. Sample 11 polymer showed higher performance. Joint application of Sample 11 and 13 polymers in hydroponics prevented ^{137}Cs accumulation in biomass of Redroot pigweed, especially in the roots.

3.2.3 Co-growing system.

Co-growing system means to grow food-plants (Shi-so, basil) and Cs-hyperaccumulator plants (Redroot pigweed) together (row by row) applying corresponding compositions/polymeric sorbents and polymer types. We postulated that Redroot pigweed would accumulate radioactive cesium from the soil and it would give an opportunity to obtain ecologically sound basil for food. Co-growing system was used for the first time.

The experiment was carried out in the following variants:

1. Control - without applying polymer, NF twice a day.
2. Sample 11 polymer in PRIZ of pigweed + Sample 13 polymer in PRIZ of basil, NF once a day.

The results showed that yields of plants in the control and polymer variant (pigweed with Sample 11 and basil with Sample 13) were not significantly different. So we can conclude the use of polymers in co-growing system of Japanese basil and Redroot pigweed decreases the NF 2 times and gives an opportunity to obtain the same yield in hydroponic culture.
In the control ^{137}Cs concentration of basil leaves and stems was higher than those in Sample 13 polymer variant 2.6 and 2.1 times, respectively. The variants did not differ with regard to ^{137}Cs concentration in the roots. Thus, the translocation of ^{137}Cs was reduced in the biological chain "root-stem-leaf" in case of applying Sample 13 polymer.
Almost the same regularities were obtained for pigweed: ^{137}Cs concentration in pigweed shoots was higher than in the roots by 2.9 times in the control, by 3.0 times in Sample 11 polymer variant. In the control ^{137}Cs concentration in pigweed shoots and roots was 2.2 times higher than those in Sample 11 polymer variant.
In the control the uptake of ^{137}Cs by basil per plant was 2.7 times higher than in Sample 13 polymer variant, the uptake of ^{137}Cs by pigweed per plant was 2.6 times higher than in Sample 11 polymer variant.
So, co-growing system was effective for basil growing, but did not meet our expectations for pigweed (we postulated that Redroot pigweed would accumulate more radioactive cesium).

3.3Assessment of the effectiveness of polymers in field
The tests were carried out in natural field conditions (v. Taronik, with the radius of 7km from ANPP**)** with and without application of polymers in PRIZ.
The same polymers (1 g/plant) were applied in PRIZ during planting of seedlings.
3.3.1Japanese basil
The experiments were carried out in the following variants:
1st VS:
1. Control-without applying polymer, irrigating frequency (IF) once a three days.
2. PRIZ + Sample 13, IF once a four days.
3. PRIZ + Sample 12, IF once a four days.
4. PRIZ + Sample 4, IF once a four days.
2nd VS:
1. Control-without applying polymer, IF once a three day.
2. PRIZ + Sample 13, IF once a four day (repitation of 2013 tests)..
3. PRIZ + Sample 13*, IF once a four day.
4. PRIZ + Sample 73*, IF once a four day.

The test results showed that in 1st VS dry yield leaves in all polymer variants reached 1.7-2.0 times as the control plants, but in 2nd VS there were no significant differences among all variants, though the irrigating frequency of plants with polymers was reduced by 25-30 %. Only leaves yield of Sample 13* reached 1.3 times as the control plants in 2nd VS.
In 1st VS control plants differed by high content of essential oil in dry leaves (1.3-1.6 times). Meanwhile the plants in polymers variants distinguished with high content of β-karotin (by 20-40%). In 2nd VS application of Sample 73* polymer decreased the intensity of essential oil biosynthesis 1.5 times compared to the control and other polymer variants. In fact the use of polymers in Japanese basil field experiments gives an opportunity to decrease the irrigating frequency by 25% and ensure the same or high yield of leaves for food.
The results showed that in the control ^{137}Cs concentration in the leaves was higher than all polymer variants both in 1st (by 2.1-2.3 times) and 2nd (by 5.6-9.5 times) VSs (Figure 13). Polymer variants did not differ with regard to ^{137}Cs concentration in the leaves both two VSs.

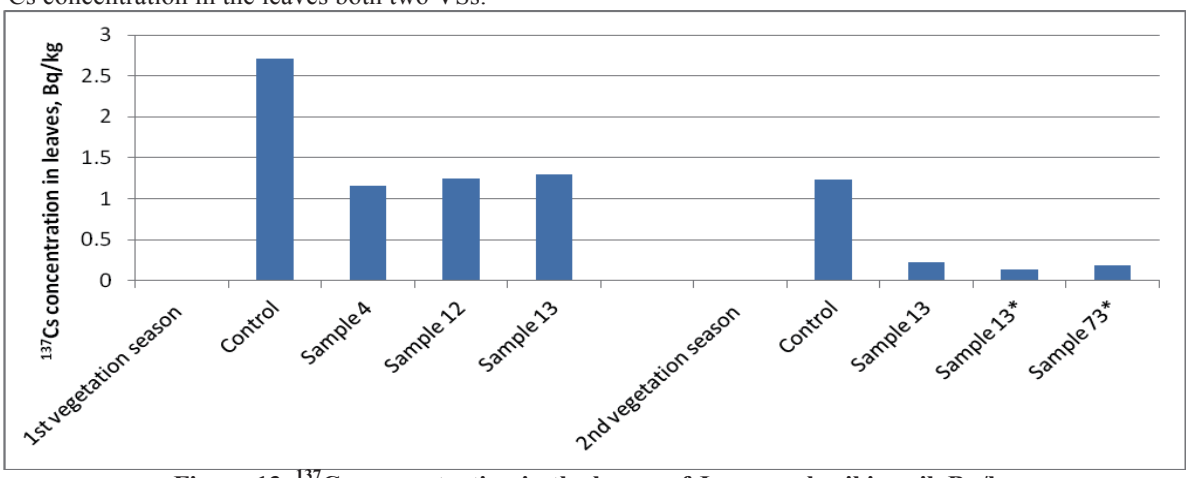


Figure 13. ^{137}Cs concentration in the leaves of Japanese basil in soil, Bq/kg

In Sample 4 and 12 polymer variants ^{137}Cs concentration in the roots was higher 3.0-3.9 times compared with the control. It suggests that polymers prevented ^{137}Cs migration from the roots to stems and leaves, Sample 4 and 12 showed the higher performance.
In 2nd VS the maximum value of ^{137}Cs concentration in the roots was observed in Sample 73* which exceeded other variants 4.2-6.5 times. Sample 13 and 13* polymers prevented the accumulation of ^{137}Cs in the stems and leaves.
In soil as well as in hydroponics the roots of control variant showed the lowest uptake of ^{137}Cs both in 1st and 2nd VSs compared to all polymer variants.
In 2nd VS ^{137}Cs uptake by basil per plant in the control was 3.0 - 3.2 times higher than in Sample 13 and 13* polymers variants (Figure 14).

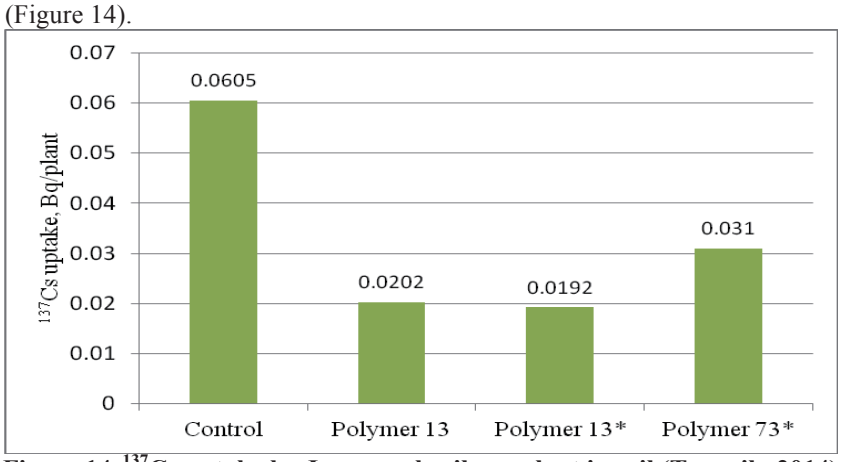


Figure 14. ^{137}Cs uptake by Japanese basil per plant in soil (Taronik, 2014)

The lowest AC of ^{137}Cs in the roots was observed in the control variant in both VSs.
In 1st VS there was no significant difference in terms of ^{137}Cs concentration in soil different layers both in the control and polymer variants, but in 2nd VS Sample 13 polymer provided the decrease of ^{137}Cs concentration in 0-5 cm layer by 1.3 times compared to the control.
It also turned out that at the end of vegetation period, compared with the beginning, the concentration of ^{137}Cs in 0-20cm layer decreased by 1.6, 1.9, 1.8 and 1.5 times for the control, Sample 13, 13* and 73* polymer variants, respectively (Figure 15).

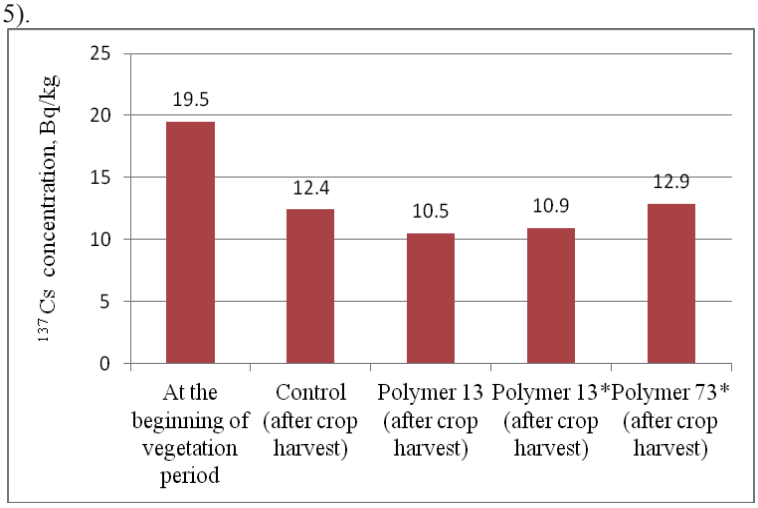


Figure 15. ^{137}Cs concentration in soil 0-20cm layer at the beginning and at the end of vegetation period, Bq/kg (cover crop-Japanese basil, 2nd VS)

3.3.2 Redroot pigweed

The experiments were carried out in the following variants:
1st VS:
1. Control-without applying polymer, IF once a 3-4 days.
2. PRIZ + Sample 3, IF once a 3-4 days.
3. PRIZ + Sample 11, once a 3-4 days.
4. PRIZ + Sample 13, once a 3-4 days.
2nd VS (we reduced the number of testing variants by recommendation of Expert Group)
1. Control-without applying polymer, IF once a 3-4 days.
2. PRIZ + 0.5g Sample 13 + 0.5g Sample 11, IF once a 3-4 days.

The results of Redroot pigweed productivity grown in field in 1st VS showed that dry yield biomass of Sample 13 in PRIZ reached 1.8 times as the control plants (above-ground part 1.9, roots 1.5 times), but there were no differences among control, and Samples 3 and 11 polymer variants. Dry yield biomass of Sample 11+13 variant reached 1.9 times as the control plants (above-ground part 1.9, roots 1.6 times).
It turned out that in case of separate application of polymers, the control and polymer variants with the ^{137}Cs concentration in the above-ground parts of Redroot pigweed didn't significantly differ (1st VS). While the use of Sample 11+13 polymer reduced ^{137}Cs concentration in biomass of Redroot pigweed compared to the control variant (2nd VS).
It was cleared up (Figure 16) that in 1st VS in Sample 13 polymer variant ^{137}Cs uptake by per plant of pigweed was 2 times more compared to the control: by the above-ground part 1.9, by the roots 3 times. This resulted in decrease of ^{137}Cs concentration in soil (0-20cm layer) 1.7 times at the end of vegetation period compared to the control. It suggests that the use of Sample 13 polymer promoted the ^{137}Cs concentration decrease in the soil (Figure 17).
On the contrary, in 2nd VS in Sample 11+13 polymer variant ^{137}Cs uptake by per plant, above-ground part and roots of Redroot pigweed was lower 1.8, 1.9 and 1.3 times, respectively, compared to the control. Sample 11+13 polymer didn't influence on ^{137}Cs concentration in the soil layers.

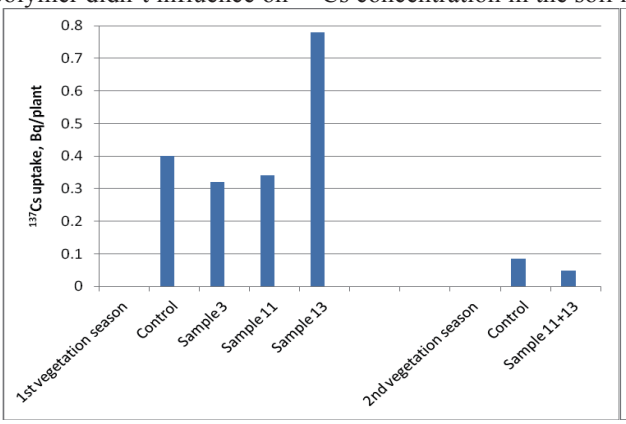


Figure 16. ^{137}Cs uptake by pigweed per plant in field (1st and 2nd VSs).

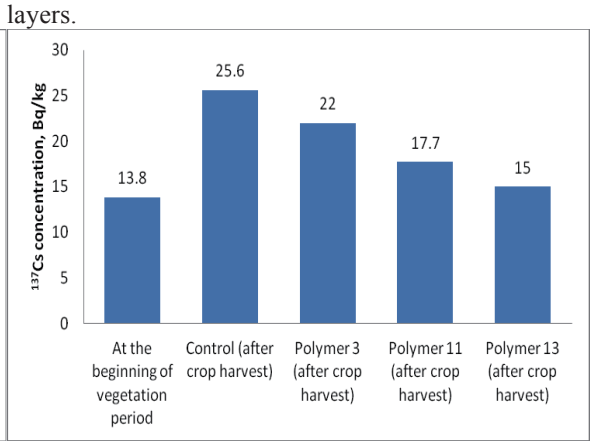


Figure 17. ^{137}Cs concentration in soil 0-20cm layer at the beginning and at the end of vegetation period, Bq/kg (cover crop – Redroot pigweed, 1st VS)

3.3.3 Co-growing system

The experiment was carried out in the following variants:
1. Control - without applying polymers;
2. Sample 13 polymer in PRIZ of basil + Sample 11 polymer in PRIZ of pigweed.

The variants didn't differ with regard to yields as in hydroponics. We can conclude that applying of polymers in soil co-growing system has no influence on the yield of Japanese basil and Redroot pigweed.
In the control the concentration of ^{137}Cs in basil leaves was higher 1.7 and in basil roots was lower 1.8 times than in polymer variant. This suggests that Sample 13 polymer promotes the ^{137}Cs accumulation in the roots and at the same time prevents the ^{137}Cs movement from the roots to leaves.
The concentration of ^{137}Cs in pigweed roots was higher than in the shoots 1.3 times for control, and 4.2 times for Sample 11. That is to say the movement of ^{137}Cs in the biological chain "root-shoot" is more intensive in the control variant. In the control ^{137}Cs concentration in pigweed shoots was higher than in Sample 11 polymer variant 1.6 times. On the contrary, in the control ^{137}Cs concentration in pigweed roots was lower than in polymer variant 2.1 times. This suggests that polymer promotes ^{137}Cs accumulation in roots and at the same time prevents ^{137}Cs movement from roots to leaves.
As in hydroponics, co-growing system was effective for basil growing, but did not meet our expectations for pigweed (we postulated that Redroot pigweed would accumulate more radioactive cesium).

4 Conclusions

A novel composite adsorbent containing inorganic filler and binding synthetic polymer was prepared and characterized.

Radioactive cesium uptake from water

-The polyelectrolyte polymers having different counter-ions and acting as cation exchangers can influence the transport of radionuclides in *water – plant* systems.

Water-retaining VA/AAc copolymer can absorb approximately 30% ^{137}Cs and ^{134}Cs from radio-contaminated water.

-Inorganic fillers in polymer compositions significantly enhance the uptake and retention of radioactive cesium from water. The increase of Cs uptake was 2-2.5 times for VA/AAc copolymer, and 2.5-3 times for ammonium acrylate.

- Despite the type of polymer matrix the addition of ferrocyanide in compositions increased uptake of radioactive cesium from water more than 20%.

After testing some of potentially effective polymers for plants growing we made some conclusions:

- Applying of polymers in Japanese basil culture creates an opportunity to decrease the nourishment frequency 2 times under hydroponic conditions culture and the irrigating frequency by 25% in field, and ensure the same and even high yield of leaves (for food) with high quality indices.
- Applying of polymers in Redroot pigweed in hydroponic culture had no influence on the yield; only Sample 13 polymer and joint application of Sample 13 and Sample 11 polymers in field increased it 1.8-1.9 times compared to the control.
- The use of Sample 13 and 13* polymers in PRIZ prevented the accumulation of ^{137}Cs in basil leaves both in hydroponics and soil.
- Sample 13 and 13* polymers decreased ^{137}Cs concentration in the soil layer by 10-20% compared to the control.
- The use of Sample 13 and 13* in soil and hydroponics will support to get more ecologically sound biomass for food.
- Separate application of Sample 11 and Sample 13 polymers in hydroponics promoted the accumulation of ^{137}Cs in Redroot pigweed biomass and decrease ^{137}Cs concentration in the soil layers, but their joint application, on the contrary, prevented ^{137}Cs accumulation in biomass of Redroot pigweed both in hydroponic culture and in soil, as well as didn't influence on the ^{137}Cs concentration in the soil layers.
- Co-growing system wasn't effective: polymers decreased ^{137}Cs accumulation both in basil leaves and in pigweed whole biomass (we expected ^{137}Cs concentration decreasing in basil leaves and increasing in pigweed biomass).
- Based on our initial experiments there is an opportunity to obtain ecologically sound soybean in Cs-contaminated soils by using polymeric sorbents (additional investigations are necessary).
- In Cs-contaminated soil by radioactive cesium absorbance capacity polymers have the following trend: 73* > 13* > 13.

5 Applicability and Recommendations for Japan

We propose new countermeasures to Fukushima in this project: to bind radioactive cesium using polymeric sorbents with subsequent possibility of removal of the polymer (accumulated radioactive cesium) from the soil.
Laboratory samples of polymeric sorbents which are the most effective can be provided for the tests in the contaminated area of Fukushima.
High water purification level (up to 90%) of our sorbents allows us to offer them as well for water purification at Fukushima region.
We recommend to use Sample 13 and 13* polymers for growing plants in areas with low level of contamination which will give an opportunity to obtain harmless and radioecologicaly sound biomass for food.
Also we suggest using hydroponic cultivation of food plants in the areas with low and medium level of Cs-contamination.
It is important to note that another advantage of Sample 13 is that after adsorbing radioactive cesium, it can be condensed into small volumes with carbonization.

Experiments of Cesium Uptake by Livestock and Crops

ISTC-K-2085

Development of a set of measures for production of assured quality agricultural goods under radioactive contamination conditions

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National Nuclear Center of Kazakhstan (Project Manager) (Kazakhstan)

ISTC/STCU Fukushima Expert Meeting

Tokyo, Japan
November 5-6, 2015

Summary Report – project #K-2085

on work performed from *September 01, 2013* to *August 31, 2015*

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Duration:	24 months
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1. Brief description of work: objectives, and expected results

The Fukushima accident caused extensive pollution with artificial radionuclides in vast areas of Japan. Most of these areas were used for agriculture. Prohibition or restrictions of agriculture in the affected areas negatively affect the food security because of rapid urbanization in Japan. To produce agricultural goods that are safe in terms radiation parameters, it is must to characterize radionuclides distribution in tissues and organs of crops and animals. For animal feeding it is important to collect information about the dynamics of radionuclides in the tissues and organs of animals. Given the shortage of agricultural land it is essential for Japan to conduct activities that make it possible to reduce the transfer of radionuclides in crop products, which is the main in the human food basket and the diet of farm animals.

Such data can be found on the territory of the former Semipalatinsk Test Site (STS), where there are ecosystems with various radioactive contamination caused by different types of tests (ground, atmospheric and underground nuclear tests, excavational nuclear explosions, tests warfare radioactive substances, etc.).

Thus, the main objective of the project was to develop a set of measures for production of agricultural goods with assured quality in conditions of radioactive contamination.

The data analyses presented by the Ministry of Agriculture, Forestry and Fisheries (Statistical Handbook of Japan, 2010), showed that the most developed in Japan is swine breeding, beef and dairy cattle and poultry. Analysis of the international data showed that transfer parameters of ^{137}Cs into livestock products have well been studied, whereas data on pork and poultry products are scarce. Therefore, in this project it is advisable to investigate the transfer of radionuclides into pig and poultry products at various forms of uptake (with soil, water and plant food). The data obtained will allow predicting the possible content of radionuclides in the products, and the data on the dynamics of the radionuclides concentration in the products at uptake and excretion of radionuclides will regulate the intake of radionuclides with diet for safe products.

According to the Ministry of Agricultural, forestry and Fisheries (Statistical handbook of Japan, 2010) in Japan out of crop products the dominating are rice, potatoes, cabbage, onions, sugar beets. Analysis of global data shows quite well-studied parameters of radionuclides transfer in these types of products. Therefore, the project focus on the experimental evaluation of the effects of physical and chemical properties of soils on the radionuclides transfer in some types of crop products.

Thus, given the characteristics of agriculture in Japan we've identified three main objectives to achieve the Project Goals:

1. Study of features of animal products contamination with radionuclides.
   1.1. Study of features of pork products contamination with radionuclides at various forms of uptake.
      1.1.1. Study of features of pork products contamination with radionuclides at uptake with soil and water.
      1.1.2. Study of features of pork products contamination with radionuclides at uptake with vegetable foods.
   1.2. Study of features of poultry products contamination with radionuclides.
      1.2.1. Study of features of poultry products contamination with radionuclides at uptake with soil and water.
      1.2.2. Study of features of poultry products contamination with radionuclides at uptake with vegetable foods.
2. Study of features of radionuclide contamination of crop products used as feed for farm animals.
   2.1. Investigation of effects of soil's physical and chemical parameters on radionuclide contamination of food components of the plants studied.
   2.2. Study of the distribution of radionuclides in the vegetative organs of the agricultural crops.
3. Development of recommendations for production of assured quality agricultural goods to be produced in the conditions of radioactive contamination.

Experimental data on the transfer of radionuclides in agricultural products obtained through field scale experiments with farm animals and plants in the conditions of radioactive contamination of STS. The experiments were carried out in ecosystems with concentrations of artificial radionuclides in the environment. Different forms of radionuclides uptake in experimental animals with components of the diet (with soil, water, plants) will be considered in experiments with farm animals. In experiments with plants that make up the food supply for the test animals the distribution of radionuclides in individual organs of crops and effects of some physical and chemical properties of soil on their availability the studied species pabular part were studied.

Conducting research under the selected tasks of the project allowed to for:
– obtain data on the distribution of radionuclides in pigs and poultry (chicken);
– determine transfer factors of radionuclides in pig and poultry products;
– determine the dynamics of radionuclide concentrations in pork and crop products at different forms of radionuclides uptake with components of the diet (soil, water and plant food);

– determine dynamics of radionuclides excretion from the body of pigs and chickens and set the half-periods of radionuclide concentrations in pig and poultry products when shifting to a "clean" diet;
– determine transfer factors of radionuclides in crops, being the main food basket of people and diet of farm animals (pigs and chickens);
– experimentally to determine dependencies of radionuclide transfer into the studied species of wild and agricultural plants on the physical and chemical parameters of the soil.

2. Modifications of work after April 2014 meeting

After the April 2014 Meeting, the experts gave the following recommendations:

Develop detailed data of Cs uptake in animals-different organs based on soil concentration similar to the Cs level found in Japan. Feed and water for animals are Cs free in Japan. Investigate time-dependent relationships between feedstuffs contamination and livestock contamination to build a scientific basis to helps husbandry farmers build confidence for their future business.

To fulfil these recommendations, the experts proposed the following changes to the project:

Task 1: Study only cesium uptake with soil contaminated at the level of Fukushima, and on subsequent cesium excretion from pigs and chickens with clean soil and feed. The use of the Fukushima level concentration is for Japanese to accept study results of this project, rather than to question the validity of scientific data of measured transfer parameters. In this regard, the physical-chemical form of ^{137}Cs should be similar to Japanese conditions.

The productivity and feeding ration of animals studied should be similar to those typical for Japanese animals.

Task 2: Study only distributions of cesium in agricultural crops.

Since the experimental studies of radionuclides transfer into agricultural products at intake with plants and water were completed before the meeting in April 2014, and taking into account the experts' recommendations, in the remaining period of the project we continued researching only Cs transfer and only with the soil. In addition, Japanese experts wondered whether it is possible to use data on the transfer parameter obtained for domestic pigs, which live within the territory of the impact of the Fukushima 1 accident. Given the interest of the Japanese colleagues we conducted additional experiments with wild boars. Parameters of Cs transfer into meat of wild pigs at prolonged intake with soil.

The experiment used soil, with the values of the Cs activity concentrations that are within range of radionuclides activity concentrations in soil impacted by Fukushima.

Agricultural experiments with plants also continued only for ^{137}Cs.

3. Technical approach, method, experiments, theory, etc

The full-scale experiments with farm animals and plants, as well as with wild vegetation were carried out on radioactively contaminated areas of the Semipalatinsk test site. These are "Degelen" site, representing a mountain range within which underground nuclear test were done in horizontal workings - adits and "P-2" site on the "Experimental field" area were ground nuclear tests were carried out.

Test procedure

Study of features of pork products contamination with radionuclides at various forms of uptake

Three groups of animals were used for the studies. Each group consisted of 16 piglets at 2 months of age. The first group of animals was daily fed to the soil with radionuclides from a prepared thoroughly homogenized mass. The second group was daily watered to radionuclide contaminated water with constant level of artificial radionuclides.

The third group was daily fed to forage with grass meal containing ^{137}Cs.

The maximal duration of the animals on the "dirty" diet was 56 days, after which the animals of both groups were shifted to a "clean" diet. From each group after 7, 14, 28, 56, 63, 70, 84, 112 days from the start of radionuclides uptake 2 pigs were slaughtered per each term. This scheme of the experiment allows to determine the dynamics of radionuclides tansfer into the products, as well as the dynamics of the radionuclides excretion from the body.

The element to be studied was ^{137}Cs. The animals were kept in stalls.

Liver, muscle tissue were taken for analyses at each term. From the animals contained in the experiment for 56 days we selected more tissues and organs – muscle, bone, skin tissue, heart, kidneys, lungs, brain, blood, spleen and liver. During the experiment we monitored radionuclides uptake with soil and water by periodic monitoring the radionuclides activity concentration in the prepared mass of soil and water and accounting the amount of their consumption.

Additional studies of parameters' variability of radionuclides transfer into meat of wild and domestic pigs

This experiment was carried out with 2 types of soils contaminated with ^{137}Cs – chestnut and meadow-chestnut soils. These soils are characterized by different physico-chemical properties (mechanical composition, humus content, etc.).

According to previous data obtained in this project, the equilibrium between the uptake and excretion of radionuclides occurs after 30 days. Therefore, the animals daily for 45 days were fed to radioactive contaminated soil at the rate 260 g/d/head. After 45 days the animas were slaughtered. In total, for each type of soil we used 3 domestic pigs and 5 wild boars.

Features of poultry products contamination with radionuclides

The studies used three groups of chickens. In each group had 27 chickens. The first group of birds were fed daily to soil containing radionuclides. The second group was daily watered to radionuclide contaminated water, the third group was daily fed to a diet with grass meal containing ^{137}Cs.

The maximal duration of the animals on the "dirty" diet was 56 days, after which the birds were shifted to a "clean" diet. From each group after 7, 14, 28, 56, 63, 70, 84, 112 days from the start of radionuclides uptake by the birds, 3 birds were slaughtered per each term. This scheme of the experiment allows to determine the dynamics of radionuclides transfer into the products, as well as the dynamics of the radionuclides excretion from the body.

The element to be studied was ^{137}Cs.

Muscle tissue and by-products were taken for analyses at each term. From the birds contained in the experiment for 56 days we selected more tissues and organs – muscle, bone, heart, liver and ventricles.

Features of crop products' radionuclide contamination

In order to study the characteristics of crop products' radionuclide contamination we selected agricultural crops: potatoes, beet, sunflower, wheat, barley, corn, radish, buckwheat and oilseed rape, produced under the full-scale experiment on the "Experimental field" site.

Effect of physical-chemical parameters of soil on radionuclide contamination of the experimental plants.

To assess the effect of some properties of the soil on the formation of radionuclide contamination of the feed component of the test plants we laid research areas (experimental plots) to change certain physical and chemical parameters of soil by administering test solutions:

1. HCl and NaOH – to change soil's pH,
2. KCl – to multiple increase K in soil (an element similar to ^{137}Cs),
3. $CaCl_2$ – to multiple increase Ca in soil,
4. NaCl – to change ion composition.

In order to investigate the effect of soil's physical and chemical parameters on radionuclide contamination of crops' feed component (wheat, potatoes, beets) we laid research plots: plots for administering KCl, for administering $CaCl_2$, for administering NaCl, for administering HCl, for administering NaOH, and control plot) for each of the listed crops.

From each plot we sampled plants studied and mixed soil samples.

Distribution of radionuclides through vegetative organs of the investigated crop

Distribution of radionuclides through the vegetative organs of the investigated crops were studied for all of these crops.

4. Conclusions

Features of pork products contamination with radionuclides at various forms of uptake
Distribution of ^{137}Cs in organs and tissues of the animals

^{137}Cs distribution in organs and tissues of animals is presented as relative concentrations, which have been calculated in respect of ^{137}Cs activity concentration in a particular organ against the radionuclide activity in heart. The relative activity of ^{137}Cs in the organs and tissues of animals is shown in the table (Table 2).

Table 1 –^{137}Cs relative concentrations in the organs and tissues of animals at prolonged uptake (^{137}Cs activity in the heart is taken as one)

Parameters	*AM±SD	Min.–Max.	Q-ty
kidneys	1,6±0,4	2,4 – 1,2	8
muscle	1,3±0,3	1,9 – 1,0	7
tongue	1,2±0,3	1,8 – 0,8	8
heart	1,0	1,0	8
liver	0,9±0,1	1,1 – 0,7	8
bone tissue	0,9±0,4	1,4 – 0,5	7
spleen	0,8±0,1	1,1 – 0,6	8
lungs	0,7±0,1	0,8 – 0,6	4
brain	0,5±0,2	0,7 – 0,3	7
blood	0,2±0,08	0,3 – 0,2	7

*Note: AM – arithmetic mean; SD – standard deviation

The table shows that, overall, ^{137}Cs unlike tropic radionuclides is distributed in the body evenly. However, the difference between the highest and lowest ^{137}Cs activity in the body can be up to 8 times. At prolonged uptake the receipt highest ^{137}Cs concentrations are found in muscle tissue and kidney, and the lowest in brain and blood.

At prolonged uptake ^{137}Cs activity in the body decreases in the order: kidney> muscle> tongue> heart> liver> bone tissue> spleen> lung> brain> blood.

^{137}Cs distribution in organs and tissues is not dependent on the species and source of the radionuclide in the body.

In general, one can say that after prolonged entry of ^{137}Cs into body its distribution has a specific pattern that allows the prediction of this radionuclide concentration in one organ by its concentration in another.

^{137}Cs transfer under various uptake conditions and timing

Figure 1 shows the dynamics of accumulation and excretion of ^{137}Cs from animal organs and tissues. Data are presented as the relative concentrations of ^{137}Cs (maximum activity of the radionuclide is taken as 100%).

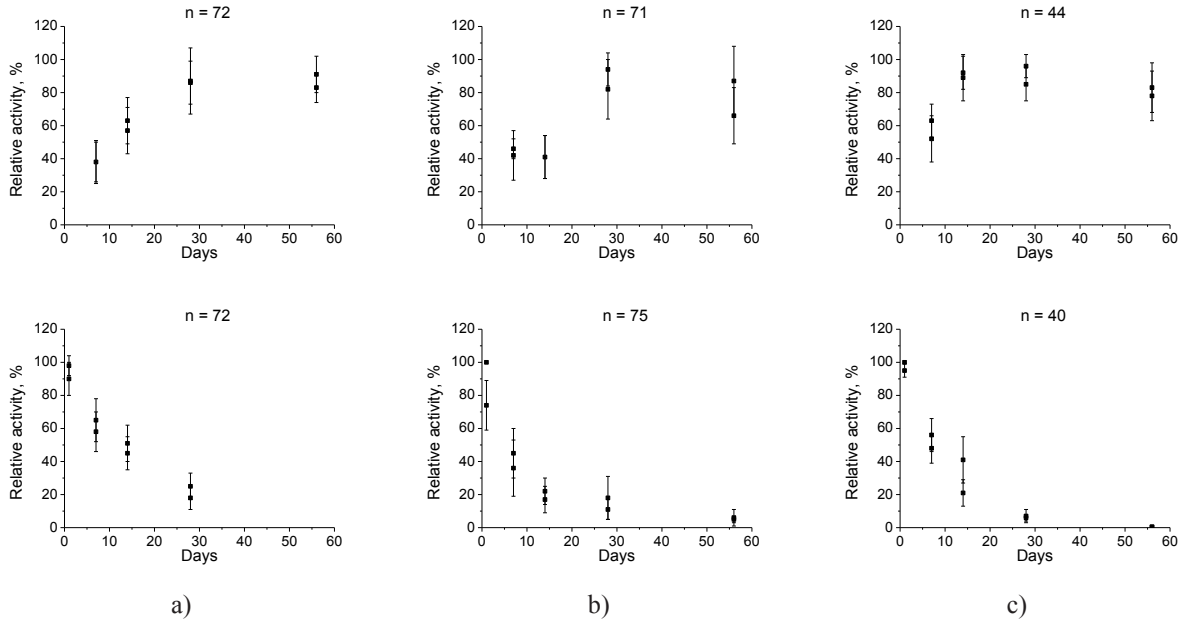


Figure 1. Dynamics of accumulation in and excretion of ^{137}Cs from animal organs and tissues at uptake with water (a), with soil (b) and forage (s)

The results showed that under conditions of daily uptake of ^{137}Cs for a long period with various components of the environment, ^{137}Cs concentration in organs increases to a certain value, followed by a dynamic balance between accumulation and excretion. No further increase occurs in ^{137}Cs activity in organs. Thus, the equilibrium state for a variety of organs and tissues occurs on 28th day.

The chosen scheme of the experiment allowed obtaining data characterizing the dynamics of ^{137}Cs excretion from pig organ tissues.

It is clearly seen that the half-decrease of ^{137}Cs activity concentration with water and forage is no longer than 14 days and on 56th day ^{137}Cs is excreted almost entirely. With soil the activity concentration half-decrease does not exceed 7-10 days, and on 56th day it is excreted almost completely.

Factor of ^{137}Cs transfer into organs and tissues of animals are presented in table (Table 3).

Table 2 – Factor of ^{137}Cs transfer into organs and tissues of animals at different forms of uptake (at equilibrium state).

Organs and tissues	AM±SD	Min.–Max.	Q-ty
Uptake source – water			
Kidney, muscle, tongue	0,74±0,1	0,98 – 0,54	12
Heart, liver, bone tissue, spleen	0,50±0,1	0,71 – 0,21	16
Brain, blood	0,21±0,08	0,34 – 0,11	8
Uptake source – soil			
Kidney, muscle, tongue	0,12±0,03	0,18 – 0,08	12
Heart, liver, bone tissue, spleen, lungs	0,083±0,03	0,14 – 0,046	19
Brain, blood	0,056±0,02	0,076 – 0,037	4
Uptake source – forage			
Kidney, muscle, tongue	0,81±0,1	0,6 – 1,12	18
Heart, liver, bone tissue, spleen, lungs	0,45±0,1	0,21 – 0,68	30
Brain, blood	0,36±0,1	0,26 – 0,43	5

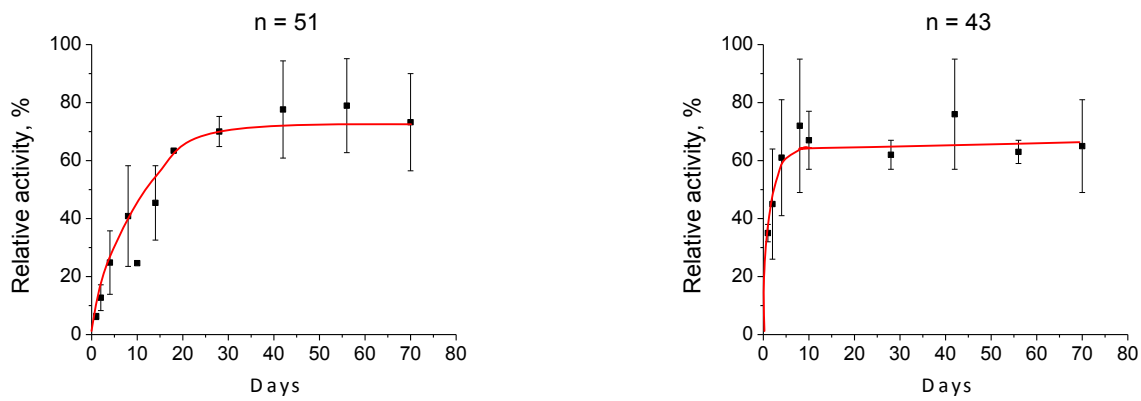
^{137}Cs concentrations in kidneys, muscle and tongue are identical, as in the heart, liver, bone tissue, spleen and lungs. Therefore, for these groups of organs and tissues we calculated separate transfer factors.

The table shows that the parameters of ^{137}Cs transfer into the organs and tissues of animals of different groups differ, and depend on the source of the radionuclide uptake. It was found that ^{137}Cs entered with food, transfers better than if entered with soil and water. Upon that the least amount ^{137}Cs transfers with soil.

In view of the information available in the IAEA database on the factors of ^{137}Cs transfer into pork, which vary from 1.2×10^{-1} to 4.0×10^{-1} (average 2.2×10^{-1}, n = 22) it was assumed that in arid climate of Kazakhstan the transfer factor of this radionuclide will also vary within this range. However, the transfer factors turned out much higher than the average transfer factors represented in the IAEA database. The factor of 137Cs transfer into pork amounted to 0,81. This difference explained by the fact that in this experiments used 2-month pigs, but IAEA database includes data obtained on mature specimens.

Features of poultry products contamination with radionuclides

The figures (Figure 2) shows the dynamics of ^{137}Cs accumulation in muscle tissue and liver at long-term daily uptake. The data are presented as relative activity of ^{137}Cs (maximum concentration in organs and tissues are taken as 100%). The presented values are average for each period.

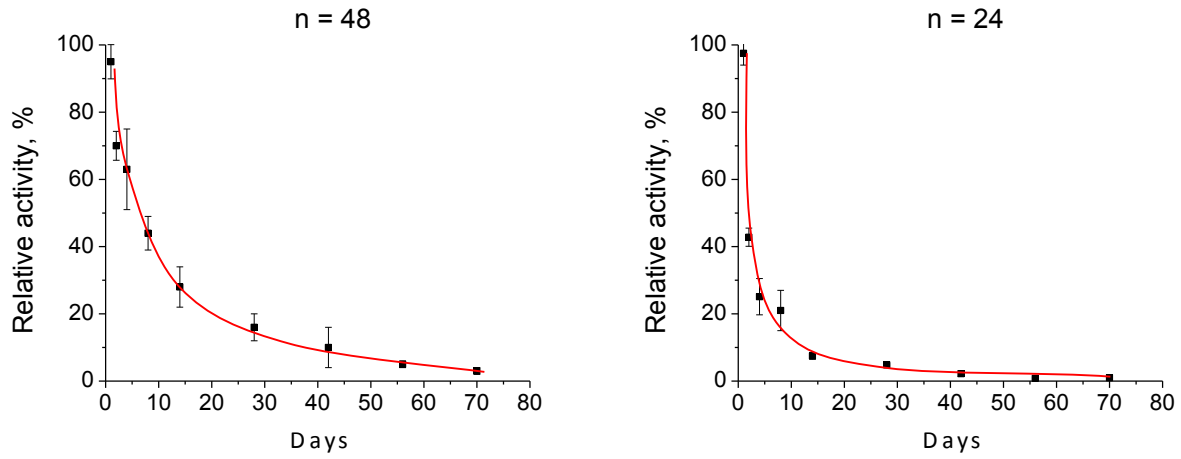


a) b)

Figure 2– Dynamics of ^{137}Cs accumulation in muscle tissue (a) and liver (b)

The charts have combined the data from the various sources of uptake. These data show that regardless of the uptake source (soil, water, food) the dynamics of accumulation does not change. Accumulation for both muscle and liver is identical. The difference is observed in the intensity of its transfer to the organs and tissues. Where it is clear that as the radionuclide gets into the body along with the redistribution and excretion from the body its concentration increases slowly, but the intensity is damped, tending to a relative constancy – constant level. The state of equilibrium for muscle tissue is characterized by 28 days, for the liver - 8 days.

Dynamics of radionuclide excretion from the liver and muscle tissue shows a similar manner in the relative concentration (Figure 3). However, the excretion dynamics for various organs is different, as evidenced by the curves shown. From the data, it became known that the half-life for the muscle tissue is 8 days. Whereas in the liver on day 2 after the birds shifted to the “clean” diet the radionuclide concentration does not exceed 50% of the maximum concentration.



a) b)

Figure 3 – Dynamics of ^{137}Cs excretion from the muscle tissue (a) and liver (b)

The resulting curves are approximated by the following equations for muscle tissue $A = 73{,}302e^{-0{,}05x}$, for liver $A = 36{,}67e^{-0{,}064x}$.

An intensive accumulation (Figure 4, Figure 5) is observed at the beginning of the radionuclide accumulation in the egg. Further, the accumulation tends to relative constancy. Dynamics of accumulation depending on the uptake sources does not differ. The state of equilibrium in the egg at different uptake sources occurs on the 21st day and no further increase in in the activity concentration is observed.

^{137}Cs concentration in the egg (Figure 4, Figure 5) does not exceed 50% of the maximum content on the 6th day.

The transfer factor is one of the main parameters. Factors of cesium transfer into organs and tissues of chickens with different uptake sources are presented in the table (Table 4).

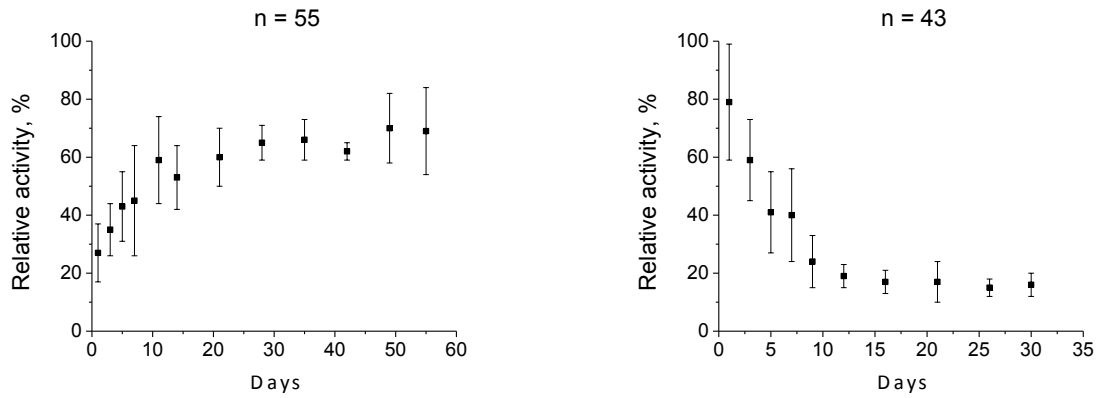


Figure 4 – Dynamics of ^{137}Cs accumulation in and excretion from the eggs at uptake with soil

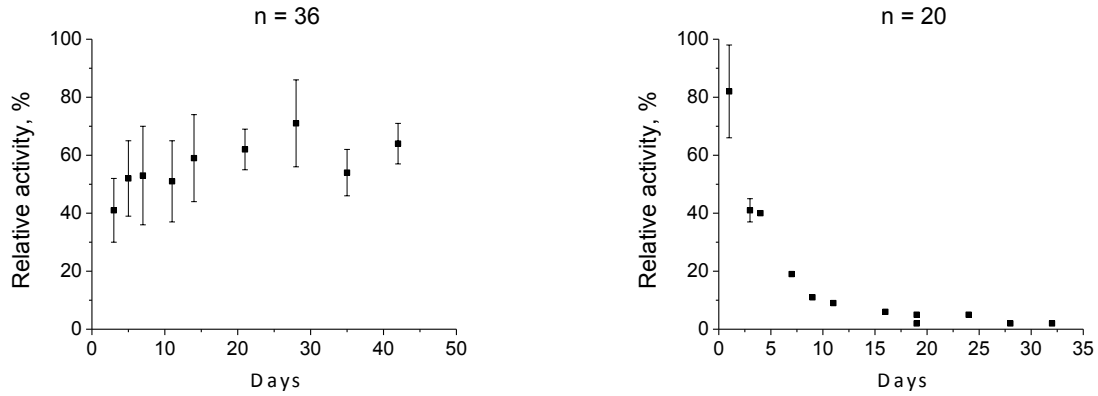


Figure 5 – Dynamics of ^{137}Cs accumulation in and excretion from the eggs at uptake with water

Table 3 – Factor of ^{137}Cs transfer into the muscle tissue and the liver under different conditions of uptake

Organs and tissues	Transfer factor with various uptake sources		
	Soil (n=13)	Forage (n=11)	Water (n=5)
Muscular tissue	0,18±0,05	2,0±0,3	4,2±0,8
Liver	0,078±0,03	0,62±0,1	1,2±0,5
Eggs	0,05±0,01	-	0,1±0,02

The table shows that the parameters of the radionuclides transfer, depending on the uptake source, are different. The radionuclides uptaken with water transfers better than when uptaken with soil and food. Upon that, the smallest value falls for soil uptake. Comparing the factors shows that the transfer factor for muscle tissue is 2-3.5 times greater than for liver regardless of the source of uptake. Availability of cesium that got in with water into the meat and liver are higher than when uptaken with food by two times, and with the soil by ~ 15 times.

Features of crop products' radionuclide contamination

To estimate the radionuclide transfer from soil to plants we used "Transfer factor" (TF) - the ratio of radionuclide per unit mass of plants and soil. The international publications, in accordance with the recommendations of the International Union of Radioecology, use the same indicator - Transfer Factor (TF or Fv) (similar to accumulation factor) [i]. Transfer factor (Fv, TF) - the ratio of radionuclide concentration per unit dry weight of plants (Bq/kg dry weight) to the concentration in soil (Bq/kg dry weight).

The ^{137}Cs transfer factor (TF) was calculated as follows:

$$TF = \frac{C_{plant}}{C_{soil}} \quad (1)$$

TF – radionuclide accumulation factor;
C_{plant} – radionuclide concentration in dry plant sample (Bq/kg);
C_{soil} – radionuclide concentration in dry soil sample (Bq/kg).

The studies achieved accumulation parameters (TF) of ^{137}Cs for 6 crops. The ^{137}Cs accumulation factors obtained are presented in table (Table 5).

Table 4 – ^{137}Cs accumulation factor

Crop	Sample type	TF*	
		Experimental	IAEA [ii]

beet	aerial part	9,6E-03	2,6E-02
	underground part	7,0E-04**	7,5E-02
potatoes	leaves	2,7E-03	-
	stems	8,8E-04	-
	roots	5,3E-03	-
	tubers	2,4E-04	4,6E-02
sunflower	seed	<***	4,3E-02
	inflorescence	<	-
	leaves	1,4E-03	2,7E-01
	stems	<	2,7E-01
	roots	2,6E-02	-
corn	grain	7,1E-04	2,3E-02
	stems	<	2,1E-02
	leaves	1,5E-03	2,1E-02
	roots	1,6E-02	-
barley	head	2,5E-04	4,3E-02
	stems	1,6E-03	2,7E-01
	roots	1,8E-02	-
wheat	head	4,1E-04	4,3E-02
	stems	4,8E-04	2,7E-01
	roots	3,1E-02	-

Note: TF- arithmetic mean; 7,0E-04**- single value; <***- radionuclide concentration in plant sample was below the detection limit of the method used; IAEA data are primarily for clay soil types*

As the results show, the parameters of ^{137}Cs accumulation for crops were 1-2 orders of magnitude below the values listed in the IAEA reference database.

Certain effect on the accumulation of radionuclides by plants can be caused by both physico-chemical properties of the soil and radionuclide species in them. In this regard researched the physicochemical properties of the soil (acidity, humus and carbonates, amount of salt in the aqueous extract and grain-size distribution (content of alphitite)) and ^{137}Cs species (Table 6, Table 7).

Table 5 – Physico-chemical properties of the soil studied (0-15 cm layer)

Sapling point	**pH**	**Humus, %**	**Amount of salt in water extract, %**	**Exchange** (mg-eq per 100g of soil)		**Amount of exchange Ca^{2+} and Mg^{2+} mg-eq per 100g of soil**	**Carbonates, %**	**Grain-size distribution, %**	**water-soluble** (mg-eq per 100g of soil)		
				Ca^{2+}	**Mg^{2+}**				**Ca^{2+}**	**Mg^{2+}**	**SO_4^{2-}**
1	6,7	3,4	0,34	13,2	2	15,2	no	40,37	0,4	0,5	2,6
2	6,8	2,3	0,36	13,2	3,2	16,4	no	40,54	0,6	1,25	2,9

In general, the studied soil on "Experimental field" site, is light-chestnut, normal (thickness of loose deposits is more than 80 cm), loam (alphitite content is 40.37%). The soils of the experimental plots have a neutral environment - pH from 6.7 to 6.8. The content of humus in the soil is within the range of 2.3 - 3.4%. In the surface soil horizons amount of soluble salts is 0.34-0.36%, which allows them to be attributed to the slightly saline soils. Among the absorbed bases, there is an absolute predominance of Ca content over Mg, which is for the majority of the different types of soil is the norm. Thus, it tells us that the physical and chemical properties of the soil do not affect ^{137}Cs transfer from soil to plants.

Table 6 – ^{137}Cs species concentration in soil on the research platform of the “Experimental Field” site

Sampling point	^{137} Cs concentration, Bq/kg (% of total concentration)									
	Water-soluble		Exchange		Organic		Mobile		Tightly bound	
	(H_2O dist.)		(1M CH_3COONH_4)		(0,1н NaOH)		(1M HCl)			
	Bq/kg	%*	Bq/kg	%*	Bq/kg	%*	Bq/kg	%*	Bq/kg	%*
1	< 3,5	<2,8	< 4,5	<3,6	< 6	<4,7	< 2,5	<1,97	110±22	86,9
2	< 1,5	<0,19	< 5,0	<0,62	< 7,5	<0,94	< 6	<0,75	780±160	97,5
3	< 1,5	<0,41	< 5,5	<1,5	< 4,5	<1,2	< 5	<1,4	350±69	95,5
4	< 8,5	<0,88	< 7	<0,72	< 3,5	<0,36	< 8,5	<0,88	940±190	97,15
Range, Bq/kg	<1,5- <8,5		<4,5- 7,0		<3,5- <7,5		<2,5- <8,5		110- 940	
Range, %	<0,19- <2,8		< 0,6- 3,6		< 0,36- <4,7		<0,88- <1,9		86,9- 97,5	
*- estimated data										

It has been established that in the soils of the "Experimental field" area the predominant concentration ^{137}Cs is observed as tightly bound species. In most cases, the proportion of tightly bound species is at least 86.9% of all species. This fact may be the main reason for the low TFs compared to international data.

Radionuclide distribution through different organs of plants

In general, the difference in ^{137}Cs TF for individual organs of plants (fruits, leaves, stems, roots) reaches 1-2 orders of magnitude. However, the distribution of ^{137}Cs in the organs of the test plants were similar: minimal accumulation is observed for generative organs (seeds, root crop and tubers), medium accumulation for the leaves and stems, and the maximal for the roots of plants (root crop in this respect is an exception). For a clearer presentation and comparison of the radionuclide TFs in the vegetative and generative organs of different crops, all obtained data on TF for plants of different species have been normalized to the ^{137}Cs TF for the leaves, which were taken as a one (Table 8).

Table 7 – Distribution of ^{137}Cs through different organs of plants

^{137}Cs TF for a particular organ / ^{137}Cs TF for leaves				
Cop	Plant organs			
potatoes	roots	Leaves	stems	клубни
	0,96	1	0,28	0,26
sunflower	roots	Leaves	seed, inflorescence, stem	-
	148,9	1	<0,5	-
corn	roots	Leaves	ear, stem, grain	-
	39,9	1	<0,38	-
barley	roots	stems	head	-
	15,76	1	0,21	-
wheat	roots	stems	head	-
	46,97	1	0,87	-

Effect of physical-chemical parameters of soil on radionuclide contamination of feed component of the plants studied

Assessing the effect of physical and chemical parameters of the soil when using chemicals that alter the original properties of the soil, enabled to obtain the following TFs (Table 9).

Table 8 –^{137}Cs TF for experimental plant species

№	crop	Sample type	Accumulation Factor	Note	Amount administered, g/m ^{2} or ml/m^2
1	wheat	stems	7,1E-03	2 × KCl	493,9
2		**roots**	**2,1E-02**		
3	wheat	head	9,7E-03	4×KCl	987,5
4		stems	1,1E-02		
5		**roots**	**1,6E-02**		
6	wheat	head	5,6E-03	2 × $CaCl_2$	367,9
7		stems	5,4E-03		
8		**roots**	**3,4E-02**		
9	wheat	head	1,4E-03	4 × $CaCl_2$	735,9
10		**roots**	**4,5E-03**		
11	wheat	head	<2,7E-03	2×NaOH	330
12		roots	3,7E-02		
13	wheat	head	<1,9E-03	4×NaOH	710
14		**stems**	**2,1E-02**		
15		roots	3,4E-03		
16	wheat	head	1,1E-03	2×Na_2SO_4	470,7
17		**stems**	**1,3E-02**		
18		roots	9,2E-03		
19	wheat	head	3,1E-03	4×Na_2SO_4	941,5
20		stems	1,1E-02		
21		**roots**	**1,2E-02**		
22	potatoes	leaves	4,8E-03	2×$CaCl_2$	367,9
23		**stems**	**5,8E-03**		
24		roots	2,4E-03		
25	potatoes	leaves	1,5E-02	4×$CaCl_2$	735,9
26		**stems**	**2,1E-02**		
27		roots	5,1E-03		

The results showed that the administering various kinds of chemicals and different dose of these substances in the soil affect the ^{137}Cs accumulation in plants. At high doses of KCl (4× KCl- quadruple dose) there is slightly greater accumulation of ^{137}Cs in the stems of wheat, but on the contrary in the roots there is slightly lower compared to the lower dose (2×KCl- double dose). In both cases, the difference in the accumulation of the radionuclide is no more than 1.5 times. Comparison of parameters of ^{137}Cs accumulation at different dosage of NaOH (double or quadruple dose) showed the impossibility to set the difference for the aerial part of wheat (head and stem), but for the roots this difference is significant and reaches about 1 order of magnitude. Using $CaCl_2$ showed that at dosage of 4×$CaCl_2$ (quadruple dose) there is a decrease in ^{137}Cs accumulation in the roots of wheat up to 10 times, and in the head up to 4 times. When using Na_2SO_4 in various dosage there is only a slight difference in the accumulation of ^{137}Cs, which does not exceed 1.3-2.8 times. In case of different doses of $CaCl_2$ we found that potato with 4×CaCl2 (quadruple dose) have higher TF compared to 2×$CaCl_2$ (double dose). This difference in the accumulation of ^{137}Cs in potatoes reaches 2-4 times.

If to compare ^{137}Cs TF, which we obtained by administering various substances for the generative organs of plants among themselves, strange to relate, the greatest accumulation is observed for the head in the case with 4×KCl. As it is known Cs is an element-analogue to K and gets into the plant the more intense the lower the concentration of potassium in the soil. However, in our case (higher doses of KCl) it turned all the way around, the reason for this may be K species in the soil, the general condition and development of plants and biological characteristics of individual plants. At the moment, this question is carefully analyzed by determination of K and its species in the soil, as well as a connection is established with the results obtained and the general state and development of the plant at the time of sampling.

Effect of different kinds of fertilizer on the parameters of radionuclide accumulation by crops

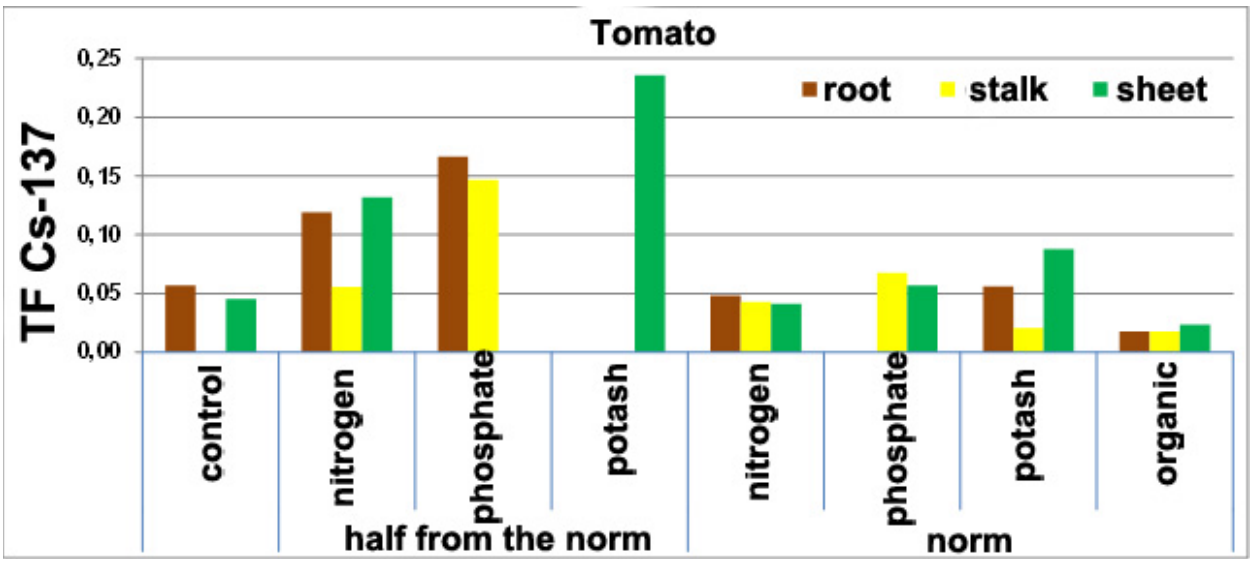
Past studies on the effect of fertilizers on the ^{137}Cs accumulation features showed that administering different types of fertilizers and the amount of fertilization leads to a change in the degree of ^{137}Cs accumulation in plants (Table 10).

Table 9 – Types of fertilizers on the experimental plot in 2011

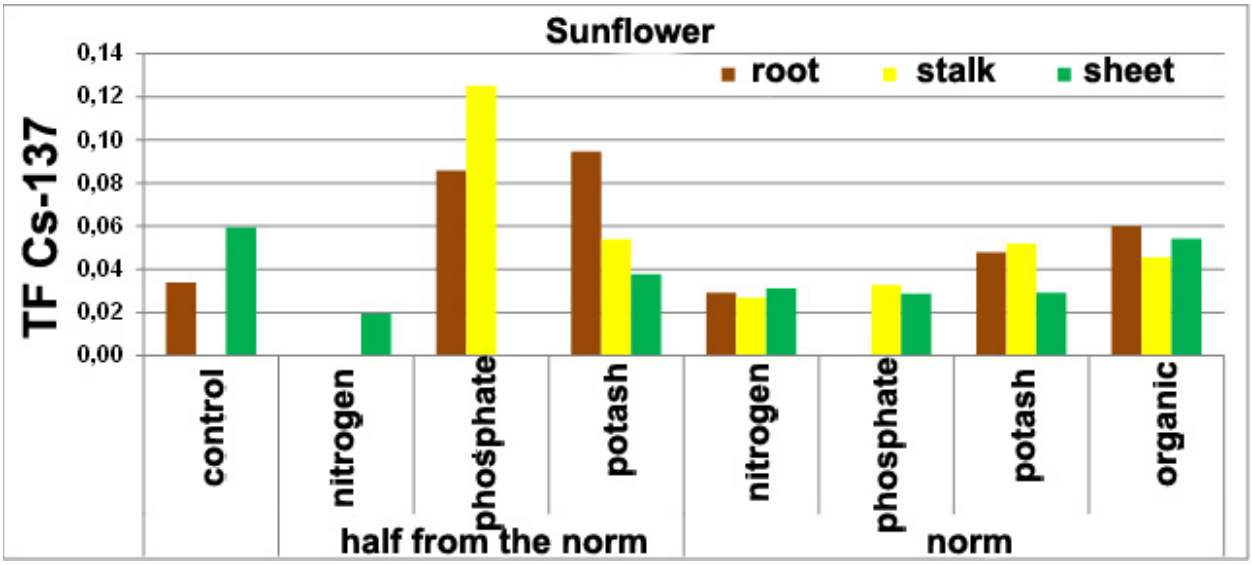
Farm crop	Mineral fertilizers, g/m^2						Organic fertilizers, kg/m^2	
	Nitrogenous (urea) $CO(NH_2)_2$		Phosphorus (superphosphate) $(CaH_2PO_4)_2 \times H_2O + 2CaSO_4 \times 2H_2O$		Калийные (Potash (potassium sulfate)) K_2SO_4		Cow manure	
	Administered	Agricultural norm**	Administered	Agricultural norm	Administered	Agricultural norm	Administered	Agricultural norm
watermelon	13,0 (6,5)*	13-19	30,0 (15,0)	30-45	15,0 (7,5)	10-13,3	6	1,5-2
tomato	18,6 (9,3)	19-26	45,2 (22,6)	30-45	15,2 (7,6)	13,3-20	5	4
sunflower	7,5 (3,75)	13-19	18,5 (9,25)	30-45	7,5 (3,75)	13,3-20	6	1,5-2

Note: * - 50% of the administered dosage, ** - summary data and recommendations of the Pavlodar Agriculture (Kazakhstan), of Professor V.I. Edelstein and Professor of Agricultural Sciences Academician P.P. Vavilov.

Thus, for example, when administering agricultural norms of nitrogen and phosphate fertilizers in the aeral part of the tomato, watermelon and sunflower there no increase or decrease in the accumulation of ^{137}Cs, compared to the control point (without fertilizer application), but when using potassium fertilizer ^{137}Cs accumulation factor exceeds ^{137}Cs accumulation factor in the control up to 2 times (Figure 6).



a)



b)

Figure 6 – Features of ^{137}Cs accumulation at different dosage of different types of fertilizers: a) for tomatoes, b) for sunflower

Administering organic fertilizers (manure) in soil also leads to the ambiguous change in accumulation of radionuclides versus control, and ranges from 0.5 to 1.8 times.

One should note that when administering 50% of the agricultural rate we obtained ^{137}Cs TF versus ^{137}Cs TF in control on average of by 2-7 times in tomato in the aerial parts and by 2.8 times in sunflower roots.

5. Applicability and Recommendations for Japan

The data on the radionuclide distribution in the body, Cs activity concentration dynamics in the products at long-term uptake by the body and, after shifting to a "clean" diet, factors of radionuclide transfer into organs and tissues of animals studied can be used to evaluate possible uptake of Cs by a human body. In addition, the data obtained can be used for regulation of radionuclide concentrations in the soil or diet that allows safe products in terms of radiation.

For example, in the Kazakhstani regulations the ^{137}Cs activity concentration permissible level for chicken meat is 180 Bq/kg. Knowing the transfer factor for muscle tissue, given the permitted levels, and diet of birds we are able to calculate the boundary parameters of ^{137}Cs. So, a chicken eats 50g of soil per day, transfer factor is 0.18, and the permissible level of ^{137}Cs activity concentration for chicken meat in Kazakhstan is 180 Bq/kg, then the boundary parameter will be ~ 25000-30000 Bq/kg (Table 11). In the case with food, the chicken eats 150 grams of feed per day, transfer factor is 2.0, the permissible level is 180 Bq/kg, then the boundary parameter will be ~ 600 Bq/kg. At uptake with water, given the daily intake of 150 ml of water, then the boundary parameter will be 215 Bq/kg.

Table 10 – Calculation of activity concentrations' boundary parameters

Diet component	Perm.Lev., Bq/kg	TF	Daily intake, kg/day.	Activity concentration, Bq/kg
Soil	180	0,18	0,050	20000
Forage	180	2,0	0,15	600
Water	180	4,2	0,15	215

In case activity concentration levels exceed, presented in the table (Table 4), permissible levels may be elevated as prescribed in the regulations. Similarly, we can calculate the boundary parameters for pork products.

Likewise the data may apply obtained for the crop plants. Using these TFs for crops and allowed radionuclide activity concentration in plant products, we can calculate the permissible concentration of radionuclides in the soil. The permissible concentration of radionuclides in the soil was calculated as follows:

$$\mathbf{ПДС_{soil} = ДУА_{dry}/Кн}, \text{ where} \quad (3)$$

$ПДС_{soil}$ – maximum permissible concentration of radionuclides under study,
$ДУА_{dry}$ – radionuclide activity concentration in the plant products, on conversion to dry weight
Кн – accumulation factor of the radionuclide studied.

For example, in agricultural plants the maximum permissible concentration of ^{137}Cs in the soil, at which the established norms in Kazakhstan for plant products are not exceeded, are shown in the table (Table 12).

Table 11 – Estimated maximum allowable concentration of ^{137}Cs in the soil for crops (dry weight of the plant) (based on norms of the Republic of Kazakhstan)

Farm crop	Products	Maximal allowable concentration of ^{137}Cs in soil, Bq/kg
Potatoes	tubers	90900
beets	root crops	92300
Corn	grain	120000
corn	grain	195000
Barley	grain	320000
sunflowers	seed	13500
Tomato	fruit	26000

The experimental data indicate the possibility of growing acceptable quality crop products in areas with elevated levels of soil contamination with ^{137}Cs. In order for the plants to conform allowable levels for ^{137}Cs in the soil similar to the STS soil, the radionuclide concentration must not exceed $n\times10^3$ Bq/kg for ^{137}Cs.

One should note that sunflower and tomato, the products of which accumulate ^{137}Cs greater than in the rest of the studied crops. This fact must be taken into consideration when growing the farm crops and receiving crop products in the STS study areas, to use the crops with the lowest radionuclide accumulation: wheat, barley, corn, and potatoes and beets.

In order to carry out an earlier evaluation of the crop products' quality (before the fruits mature, etc.) one may use leaves and stems of plants (aerial part), as organs with the highest AF. With the data in only one organ, and knowing the difference in the accumulation of radionuclides between different plant organs one can find estimative radionuclide concentration in the desired organ.

Currently analytical works are in progress. The final conclusions will be made after γ-spectrometric analyzes and presented at a meeting in Tokyo, 05-06 November 2015

6. References

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Countermeasures of Cesium Uptake by Farm Crops and Livestock

STCU-5953

Methodology for long-term radiation monitoring to dose assessment using radiological zoning and modeling of radionuclides migration in environmental and food chains

Dr. Mykola Talerko
Institute for Safety Problems of Nuclear Power Plants (Ukraine)

ISTC/STCU Fukushima Expert Meeting

Tokyo, Japan
November 5-6, 2015

Summary Report – project #5953

on work performed from Sep 01, 2013 to Aug 31, 2015

Institute for Safety Problems of Nuclear Power Plants NASU

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Doctor of technical sciences

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Contents

1. Brief description of work: objectives, and expected results

A common feature of severe radiation accidents is the contamination of large areas with intensive agriculture. Assessment of radiation situation in the agricultural sector which is the main factor of internal radiation exposure of population is not highlighted as one of the main tasks of post-emergency monitoring. The agrosphere monitoring was substituted by radiation control of product quality in fact. During these accidents information necessary to assess the radiation situation was obtained after forming the main part of population exposure. The assessment of radioactive contamination of individual parts of the environment was planned and implemented without matching and coordination. As a result, grids, their scales and integrated results are difficult to adjust in space and time, thereby uncertainties of both assessment results and planning countermeasures in contaminated areas are significantly increasing.

The aim of the project is to develop principles of creation of the comprehensive system of radiation situation forecasting, radiation monitoring of agricultural sector, radiation control of produce quality, planning and conducting counter measures to significantly increase the level of preparedness for severe radiation accidents response.

The fundamental difference of the developed monitoring methodology is *preventive before an accident* study (from the point of view of agricultural radioecology) of area that may be subjected to radioactive contamination after a large radiation accident at the nuclear power plant. The stage of preventive preparation finishes with the creation of radioecological model of territory using GIS technologies, map layers and parameters database necessary for quantitative modeling processes of the radiation condition formation that could help to prevent the formation of a large part of the human exposure dose.

Testing of the methodology was carried out using the example of two test polygons on the territory of Ukraine and Japan. The sources of input data are determined for digital maps creation and the GIS technologies are designed to process and analyze of spatial information, the same as approaches and requirements are formulated for databases and data collection system to assess the radiation situation in accordance with the recommendations of the IAEA and radiation control regulations of nuclear power plants. The methodology universality is ensured by the development of key parameters classifiers. The algorithm of creating cartographic documents for decision support is developed using remote sensing data and results of numerical forecast of territory contamination.

The decisive principle of agrosphere monitoring optimization and ensuring of countermeasures address application are the preventive comprehensive basin-landscape zoning the nuclear power plants surveillance zones and their influence zones in the case of severe accidents. The structural units of territory zoning (basin, elemental landscape or its components) are considered as environmentally homogeneous areas, and the radionuclides deposition density or other radio-ecological characteristics (types of underlying surface, soil and land use) within it are assumed to be averaged. *This structural unit is considered as a unified object of the prediction, monitoring and countermeasures planning subsystems, which makes the methodology zoning to be universal and allows for superposition of thematic map layers. The database combines information on the object at all levels of spatial scale and at all trophic levels of the food chain.*

Selected areas and their structural elements are ranked by potential criticality - the ability to form larger dose of internal exposure at equal density of soil contamination. The objects criticality is estimated through expert assessment by the integral index, which shows priority when planning of monitoring, control and countermeasures. It is defined more exactly on the base of forecast results and the radiation situation monitoring data.

The set of numerical models for prediction of radiation situation due the emergency (preventively or immediately after it) has been created, which includes the models developed using data of the Chelyabinsk (at “Mayak” plant) and Chornobyl accidents. Assessment of radiation situation is conducted comparing forecast results with the international and national standards.

The approach used in the methodology enables to execute monitoring countermeasures purposefully with compliance with priorities. The integration of the forecasting, monitoring, control and planning countermeasures systems enables to optimize the volume of monitoring and produce control and to increase their effectiveness and information capability of accepted decisions.

2. Modifications of work after April 2014 meeting

Participants of the project are based on the thesis that the developed methodology of radio-ecological zoning and organization of radiation monitoring in the contaminated area of agricultural production is universal and can be adapted and applied to any territory. Therefore, according to the initial Work Plan of the project 5953 the work was planned on the development of this methodology on the example of the test area selected in the north-western part of Ukraine (the area between the Chornobyl NPP and the operational Rivne NPP). The choice of such territory is determined by the fact that it clearly shows the impact of ecological features of the terrain on

the formation of agricultural products contamination as a result of the formation of the western trace of radioactive fallout after the accident at the Chornobyl nuclear power plant. In addition, participants in the project have all necessary radiological and cartographic information for this area.

The ISTC / STCU Technical Review Committee at its meeting in April 2014 in Tokyo recommended the project participants to adapt as much as possible the project results to the needs of work on radiological monitoring and decontamination of agricultural lands in the contaminated area around the Fukushima-1 NPP. Following these recommendations, in addition to the Work Plan the project team adapted the developed methodology to the territory of Japan. It should be noted that during the project implementation Japanese side didn't provide the project team with specific radiological and cartographic information about environmental and radioecological conditions in Japan.

As a test polygon for working-off of the methodology of the area radio-ecological zoning the territory was selected, which is characterized by a significant gradient of agrochemical properties and density of soil contamination by ^{137}Cs from 1 to 1000 kBq · m^{-2} (Fig. 1) and belongs to Date district, Fukushima prefecture. The test site includes areas that belong to Date, Soma, Miyagi, Fukushima city and administrative subordinate territory. The geographical coordinates of angles: N - 37.934º, S- 37.686º, W - 140.465º, E - 140.766º. Area test site 728.7 km^2, it is located in the north-west of the Fukushima Daiichi NPP. Distance from the NPP site to the polygon center is 57 km. Most of the test site is the territory covered by forest. There is developed agricultural production within it. The typical crops for the Date district is rice, vegetables, soybeans, potatoes, canola, fruit trees.

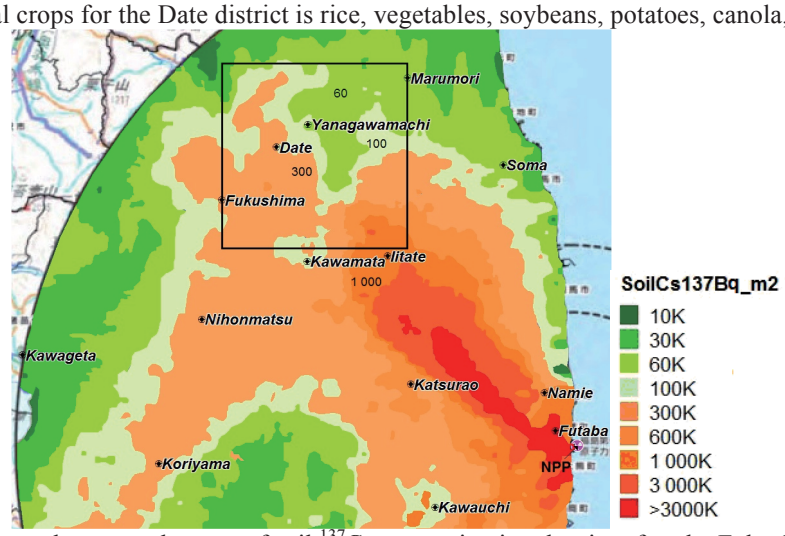


Fig. 1. The test polygon on the map of soil ^{137}Cs contamination density after the Fukushima accident.

In addition to the project Work Plan a number of issues of concern to the Japanese side in work on overcoming the consequences of the accident at the Fukushima-1 NPP were discussed:

1. Analysis of the possibility of using the value Radiocesium Interception Potential (RIP) for predicting the migration of cesium in the system "soil - plant" in Japan.
2. Analytical review of the practice of countermeasures application in order to reduce ^{137}Cs contamination of agricultural products in the region of the Chornobyl accident, which can be used in the area of the accident at the Fukushima-1 nuclear power plant.
3. Prediction and assessment of aerial contamination of crops due to ^{137}Cs resuspension in the contaminated zone around the Fukushima-1 NPP.
4. Evaluation of the ^{137}Cs contribution into contamination of the chickens meat and eggs, which is ingested at outdoor keeping.

Project members are deeply grateful to Foreign Collaborator of the project Prof. Onishi, recommended us to analyze these issues in the framework of the project.

3. Technical approach, method, experiments, theory, etc

3.1 The general structure of the comprehensive system of predicting the radiation situation, agrosphere monitoring and produce radiation quality control

The technology of creating the comprehensive system of predicting the radiation situation, agrosphere monitoring and produce radiation quality control is to create a consistent set of models, databases and technological procedures, which unites and forms the flows of information, and carries out the tasks of forecasting

and monitoring the radiation situation by means of geographic information systems and databases. The structure of system of predicting the radiation situation, agrosphere monitoring and produce radiation quality control is shown in Fig. 2.

The comprehensive system consists of the following subsystems:

- ***Subsystem of preventive radioecological assessment and zoning of the territory*** in which on the base of the territory features analysis the radio-ecological zoning is carried out with assessment of the area criticality at three spatial levels (national, regional and local) using the basin-landscape principle;

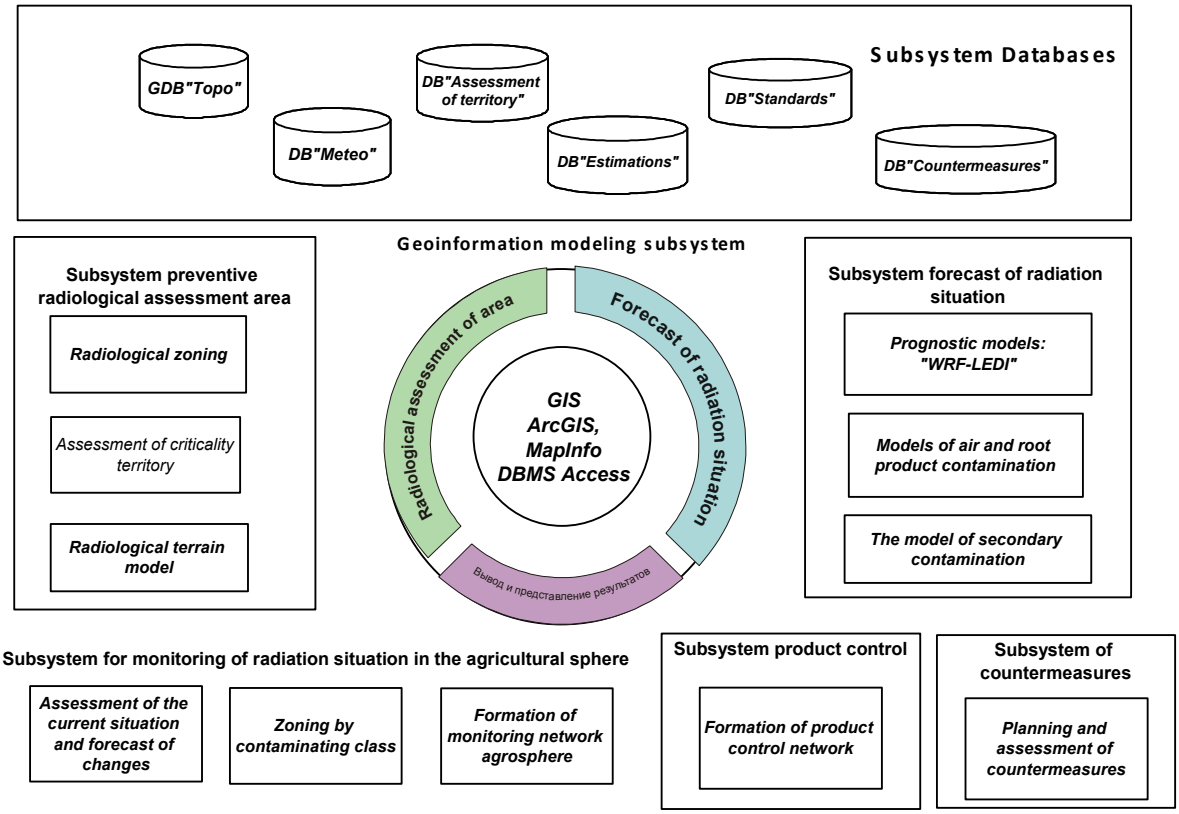


Fig. 2. The structure of the system of predicting the radiation situation, agrosphere monitoring and radiation quality control.

- ***Subsystem of the radiation situation forecast and modeling*** is intended to calculate the consequences of accidental releases in the acute and late phases of a radiation accident.
- ***Subsystem of geoinformation modeling*** provides:
  - consistent interaction and successive execution of the models included in the subsystem “Radiation situation forecast and modeling",
  - space analysis and evaluation of the radiation situation, formation of a monitoring network at regional and local levels,
  - visualization of the results of analysis and prediction of the products contamination in fields and home gardens, construction of situational maps using GIS and DBMS technological procedures.
- ***Subsystem of radiation situation monitoring*** performs tasks of the territory zoning according modeling results by classes of product s contamination and the formation of a monitoring network, processing and visualization of the monitoring results.
- ***Subsystem of production quality control*** provides processing and visualization of the products control results on the territory of accidental contamination, planning and optimization of the control network with the aim of not exceeding the national standards.
- ***Subsystem of planning and evaluating the countermeasures effectiveness*** solve management problems to optimize and to prioritize protective measures to prevent exceeding the population radiation dose limit in the contaminated areas in the event of large-scale accidents at nuclear power plants.
- ***Subsystem of information databases*** provides the system with actual input information: cartographic (DB "Topo"), aerosynoptic (DB "Meteo"), calculated (DB Calculations"- the data of radionuclides transport and deposition modeling for different types of synoptic situations), radio-ecological (results of a comprehensive

analysis and preliminary radioecological assessment of the territory), as well as the standardized interaction of the numerical models through the agreed input and output data.

Let's consider the structure and functions of the subsystems in more detail.

3.2 The subsystem of information databases

The subsystem provides a unified technology of analysis and processing of radio-ecological information with natural and geographical (topography, underlying surface, the angle of slope) and soil data using the developed and modified classifiers for soil, products, crops, landscapes, basins, etc. The relation between the cartographic and thematic databases at the three spatial levels is established through the elements of the basin, landscape, soil and administrative division with the use of GIS.

The database "Topo" includes information about climatic and landscape features of the territory. The main characteristics of the terrain (soil, river basin, vegetation, terrain elevation, slope angle, the integrated ecological code) are collected in the base table-grid obtained by the intersection of the 2x2 km grid with all map layers. The integrated information in the base table enables analyzing and zoning the territory by individual environmental indicators.

The aerosynoptic DB "Meteo" consists of operational synoptic data of the numerical weather prediction obtained using the USA model WRF and the operational data from the public server (http://nomads.ncdc.noaa.gov). The original data are objectively analyzed and interpolated into the nodes of a regular grid with different spatial step (27 and 9 km) horizontally and vertically (27 levels). These prediction data are processed, stored in a database and sent as input information into the radionuclides atmospheric transport model LEDI.

The radioecological DB "Assessment of the territory" is designed to conduct a comprehensive analysis and preliminary evaluation of radioecological situation in the territory in case of a radiation accident at the nuclear power plant. It includes the scenario maps of vegetation; tables of relationship between the radionuclide transfer factors from soil to vegetation, depending on the soil and vegetation types for foliar and root contamination; tables for calculations of the time dependence of the radionuclide transfer factors from soil to vegetation; maps of modeling results of radionuclides (^{131}I, ^{137}Cs, ^{90}Sr) concentration in agricultural plants and basic foods (milk, meat, roots) in case of emergency; **the territory zoning maps** in accordance with the IAEA criteria (OIL1-OIL3 for external dose, OIL4 for density of radionuclide contamination and OIL5-OIL6 for radionuclides concentration in food) and the types of emergency.

The database of calculated (modeling) data "Calculations" includes sets of modeling (calculated) data of atmospheric transport and deposition of radionuclides (11 items) by types of synoptic situations and models of the atmospheric boundary layer parameterization. Tables are generated by data on the density of deposition of radionuclides on the underlying surface (vegetation), on the volume activity of radionuclides in the air, and the external dose rate. Model data of transport and deposition are processed in accordance with the requirements for input information for models of calculating the products radionuclide contamination by aerial and root ways and are transmitted to the model unit.

The DB "Standards" includes national and international regulations concerning the values of control levels of radionuclides in the environment, the criteria for response to a nuclear or radiological emergency, countermeasures and protective measures with indication their effectiveness at different stages of the accident, etc. The database "Standards" includes the classifiers of cartographic and thematic information to unify, organize and analyze spatial data.

3.3 The subsystem of preventive radioecological assessment of the territory

The subsystem enables the assessment of the territory by ecological and radio-ecological factors and to identify the most critical areas, formed dangerous levels of radiation exposure to the population in emergency situations at nuclear power plants. On the basis of the basin-landscape principle and the analysis of the territory features with the use of maps of relief, basins, the underlying surface, land use the natural factors are selected that determine the ecological characteristics of the territory: type of elemental landscape, soil type, the type of the underlying surface and the type of vegetation. For the test site in Date district the sets of maps have been created (Fig. 3).

To evaluate the radioecological properties of the basins territories the soil types for each basin are grouped according to the characteristics of radionuclide migration in the system "soil-plant". Assessment of the territory criticality is carried out using the complex integral index, which is the sum of radioecological and ecological factors estimated expertly using rating and weight of each factor. The integral index as the sum of the weighted ecological parameters is calculated for the selected typologies of objects (of the same type of soil within each basin) of the basin map at state, regional and local levels. The output of the subsystem are thematic maps with the assessment of the degree of criticality of the territory and the tables of the most critical combinations of objects with the integral index calculated for three spatial levels.

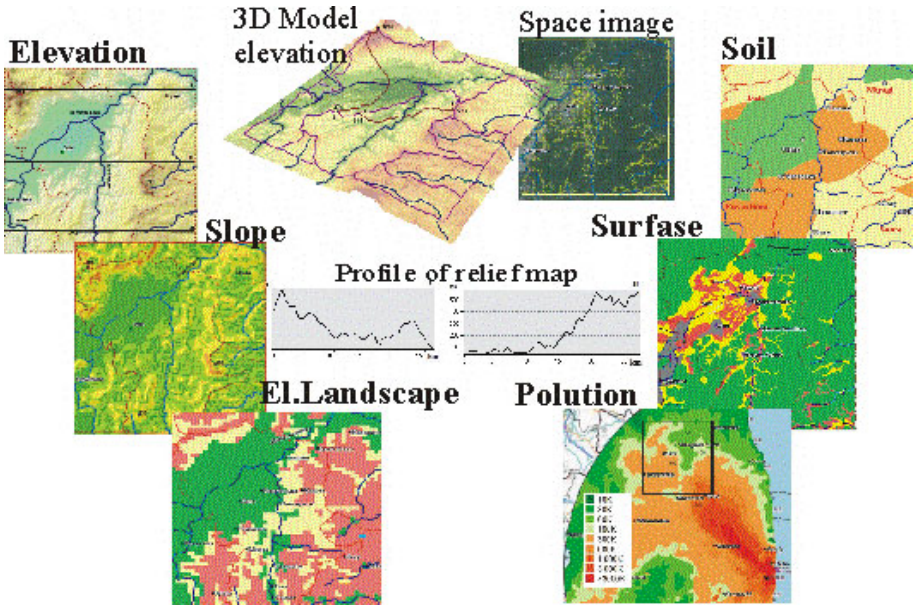


Fig. 3. Basic cartographic layers for radioecological assessment of the test site.

Fig. 4 presents the results of radio-ecological zoning with assessment of the degree of territory criticality.

The result of radioecological zoning is a selection of homogeneous typological objects with assigning a uniform code "**basin – soil – landscape – land use**", which determines the location of a homogeneous soil (plant) unit in a certain class of landscape in the basin of an appropriate spatial scale. The uniform code is used as a geo-referencing of thematic information to digital maps, and enables to move on to different spatial scales and to complex analyze heterogeneous data.

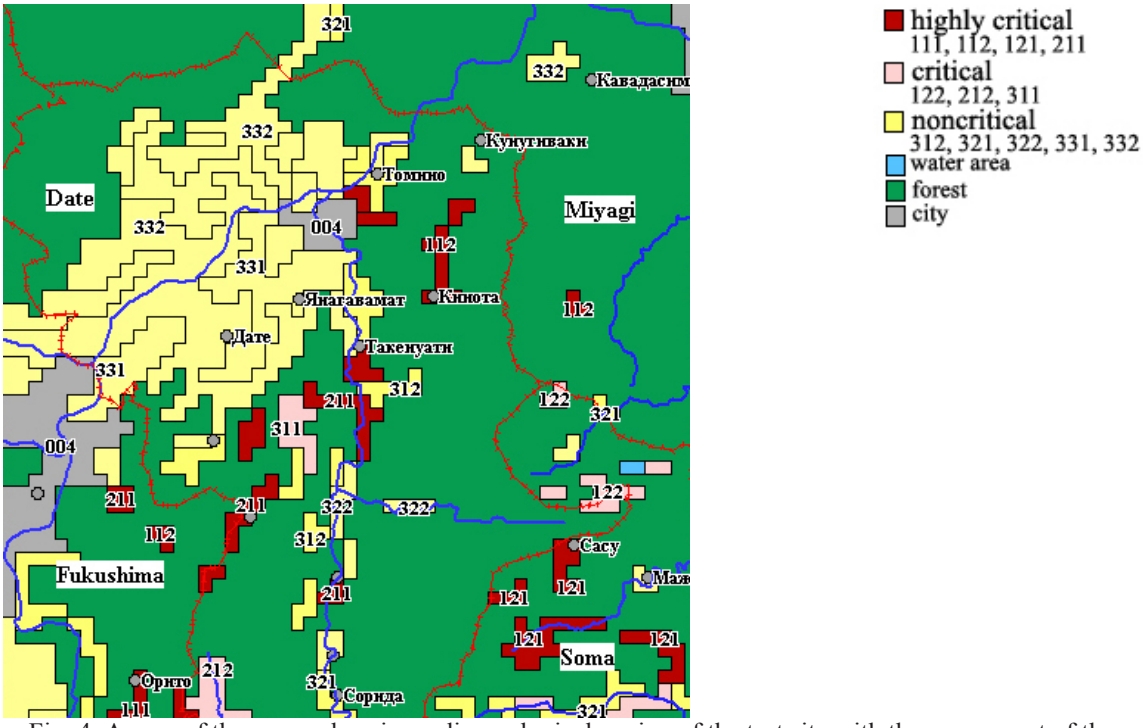


Fig. 4. A map of the comprehensive radio-ecological zoning of the test site with the assessment of the potential criticality of selected areas.

Results of the subsystem of preventive area radioecological assessment are integrated into the database "Assessment of the territory" for creation of the radioecological territory model and using in numerical simulation for predicting the development of radio-ecological situation and planning the monitoring network in an emergency. The radioecological territory model is a table or map layer in a form of regular grid where each grid cell is characterized by natural and geographic (latitude, longitude, terrain elevation, slope angle, the type of

elemental landscape, the type of the underlying surface, the basin code, type of soil and vegetation and etc.) and ecological parameters (assessment numbers and the total integral index based on a weighted contribution of characteristics used, radionuclides transfer factors from soil to plants, the soil agrochemical characteristics, etc.).

3.4 The subsystem of the radiation situation forecast and modeling

The subsystem is a consistent set of models for following tasks:

1. Calculations of radioactive substances atmospheric transport and deposition in a case of accidental releases from nuclear power plants with the use of the regional model LEDI. Meteorological information for the model LEDI comes from the numerical weather prediction model WRF.
2. Estimation of aerial product contamination using the model AeralPlant in the acute phase of the accident.
3. Assessment of the root product contamination using the model SoilPlant in the system "soil-plant-products."
4. Assessment of contamination of livestock production using the model of radionuclide transfer from feed to animals and animal products.
5. The secondary pollution of products - a model of resuspension and deposition on vegetation.

The models are experimentally grounded and parameterized in a large series of field experiments and verified according to data after the accidents at Chornobyl and Fukushima-1 NPPs.

All used models are matched at input and output data. All thematic spatial information are linked to digital maps using the comprehensive "ecological code" (the code of basin, the code of the soil, the code of landscape and the code of vegetation) and a unique cell number with geographic coordinates. Input data are presented in the cells of a regular grid with different grid steps depending on the scope of tasks: at the state level the grid step is 2 km, at the regional one is 500 m, and on the local one is 250 m. For the models interaction the technological procedures are developed to convert data into different formats and the procedures for information geocoding - converting into map layers. Unified coding of information, created classifiers and use of grids (grid - data) enables to ensure the interaction of the thematic and cartographic databases and to perform GIS modeling with building situational maps for a decision on the protective measures to reduce the population dose in the event of severe accidents.

3.5 The subsystem of geoinformation modeling

The subsystem provides a consistent operation of all subsystems and integrates the obtained results for assessment, analysis and modeling of radioecological situation. The subsystem performs the following tasks:

- analysis and evaluation of the radiation situation at all three spatial levels,
- the territory zoning using the IAEA regulations and national standards,
- classification of the territory in accordance with the task of a monitoring network forming.

Comprehensive analysis of various information obtained by the different programs (monitoring, modeling and forecasting) with different steps both in time and in space is possible under the organization of information on a unified methodological and programming basis. The established subsystem of geoinformation modeling using GIS ArcGIS, MapInfo, Surfer and DBMS Microsoft Office Access enables in combination to process, to analyze model data and to prepare situational maps and tabular material for decision-making.

Assessment of the territory from the point of view of the population living possibility belongs to the field of radiation monitoring. The agrosphere monitoring is made only in those areas where living or farming is allowed according to decisions of public bodies. If necessary, the monitoring in the exclusion zone must be carried out by special regulations.

Assessment of radiation situation at the three spatial levels is conducted by comparing the density of soil contamination and pollution of products with international and national standards.

The territory zoning is based on the results of numerical modeling at the beginning, and then using monitoring data it carried out in accordance with the criteria of the IAEA OIL1 - OIL3 using the data on external radiation dose and data of deposition on the underlying surface ^{131}I, ^{137}Cs, ^{90}Sr.

Calculation and evaluation of vegetation aerial contamination and radionuclide content in plant products (natural and seeded grasses, vegetables and root vegetables) and in main foods (milk, meat, bread) in the acute phase of an accident (OIL6) is performed by means of GIS MapInfo.

To predict the contamination of agricultural products in the acute phase of the accident the model of plant aerial contamination AeralPlant is used (Prister, 1975, 2008). The principal difference between this model and existing ones is that it was experimentally grounded in a large series of field experiments. The model main parameters were verified on data obtained after the accidents at the Chornobyl and Fukushima-1 NPPs.

The concentration of radionuclides in plant biomass at time *t* after deposition was calculated using the formula:

$$C(t) = \sigma \cdot PF \cdot (a_1 \cdot e^{-0..693 \cdot \frac{t}{T_1}} + (1 - a_1) \cdot e^{-0.693 \cdot \frac{t}{T_2}}) \quad (1)$$

where $C(t)$ is radionuclide concentration in plant at the time t after the deposition, Bq kg^{-1}, σ is the nuclide contamination density, kBq· m^{-2}; PF is the proportionality factor between radionuclide concentration in the plants and radionuclide soil contamination; t is time after deposition, days; T_1 and T_2 are the periods of nuclide half-losses of plants due to easily and difficult removed forms of radionuclide, accordingly, day; a_1– part of easily removed form.

Using the model AeralPlant calculations of ^{137}Cs contamination of leafy vegetables and grass caused by the accident at the Fukushima- 1 nuclear power plant have been made (Fig. 5). To this aim the ^{137}Cs soil contamination density maps and the scenarios of crops placement for the acute phase of the accident with assigned to them coefficients of proportionality between the deposition density and the radionuclide specific activity in the product PF were used.

According to the modeling results (Fig. 5) in the first days after the accident the ^{137}Cs concentration in leafy vegetables within the selected polygon was high and reached 120 - 6 000 kBq • kg^{-1}. The operation intervention level OIL6 (IAEA, GSG-2) under the emergency is 2 kBq • kg^{-1}. That is, according to the modeling in the first days of the accident the contamination of leafy vegetables was 2-3 orders of magnitude higher than OIL6 within the test polygon area.

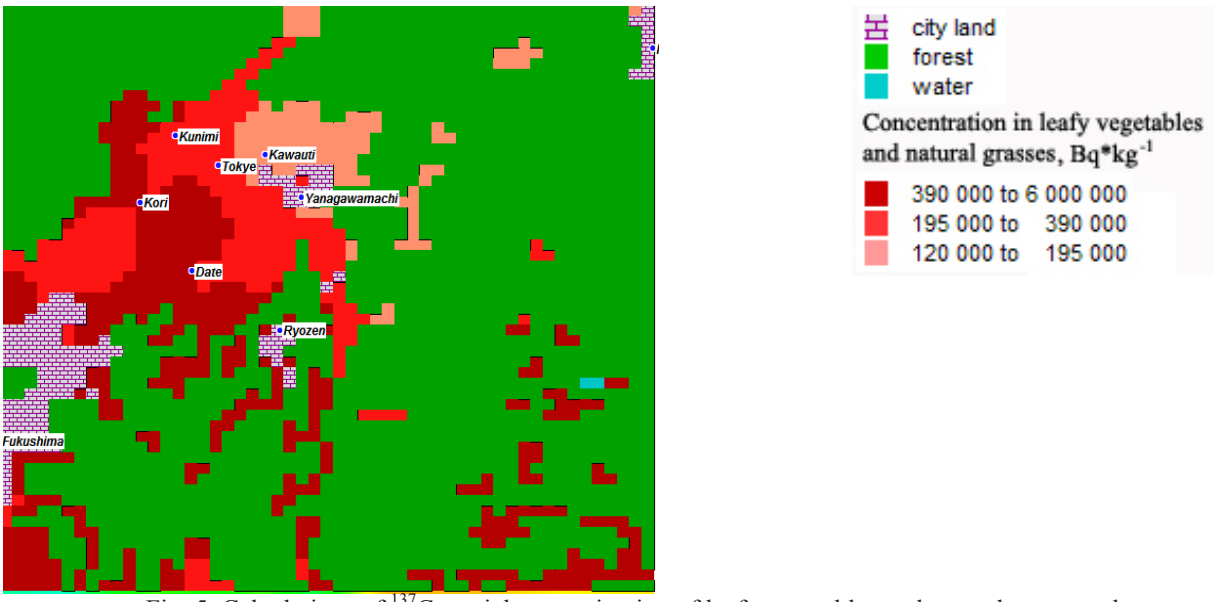


Fig. 5. Calculations of ^{137}Cs aerial contamination of leafy vegetables and natural grass on the test polygon due to the radioactive fallout after the accident at the Fukushima-1 NPP.

The concentration of ^{137}Cs in natural grass in the territory selected for analysis also reached hundreds of thousands and millions of Becquerel. Even if the diet of cows in Japan includes only 30% of the demand for roughage, grass feeding or cows grazing in this area would lead to significant contamination of milk. Provided that the daily use of grass is 10 - 15 kg, about 1 200 - 60 000 kBq entered the body of cattle, of which approximately 1% of ^{137}Cs passed into 1 liter of milk. Thus, nuclide concentration in milk could reach 12 - 600 kBq • l^{-1}, which is also significantly higher than the OIL6. It eliminates the possibility of using milk for consumption in a fresh kind and requires processing it into products of long holding time for clearance of ^{137}Cs. These calculations make it possible to recommend for Japanese specialists to reconstruct the concentration of ^{137}Cs and ^{131}I in milk in the first days of the acute period and support correctness of the recommendations to ban milk drinking and leafy vegetables use in the first 10 - 14 days after the accident without monitoring and control data to prevent people overexposure.

To verify the modeling results of aerial vegetation contamination we used the official data of radiological monitoring of the territory, contaminated by the accident at the Fukushima-1 NPP (MEXT, Japan) FMD database, which was provided for the IAEA. There are data of iodine and cesium radionuclides measuring in leaves in a few space points situated at different distances from the NPP, with different levels of soil contamination (Fig. 6).

One of the monitoring points en2_8 presented in the database with soil radiocaesium contamination of 100 - 300 kBq m^{-2} is located on the test polygon territory. To test the modeling results in the territories of the other contamination levels the monitoring points are used near the test polygon: for territory between isolines of ^{137}Cs soil contamination density 60 - 100 kBq m^{-2} (point en2_4) and 600 - 1000 kBq m^{-2} (point en2_1). Fig. 7 shows the dynamics of the ^{137}Cs contamination of leafy vegetables after the accident at Fukushima-1 NPP in these

monitoring points, as well as the dynamics of ^{131}I contamination of leafy vegetables for 3 other monitoring points (where the measurements of radioactive iodine were available).

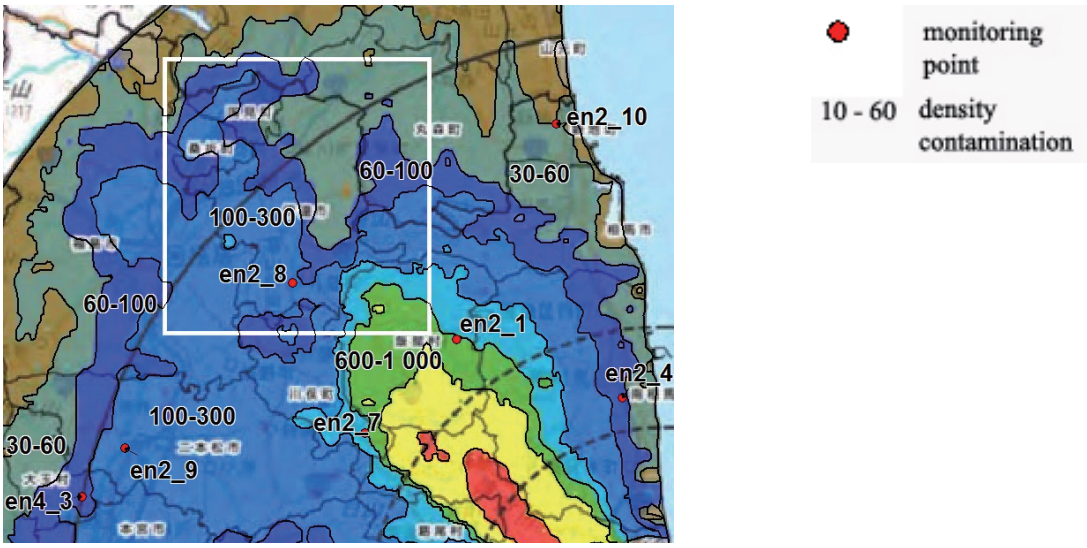
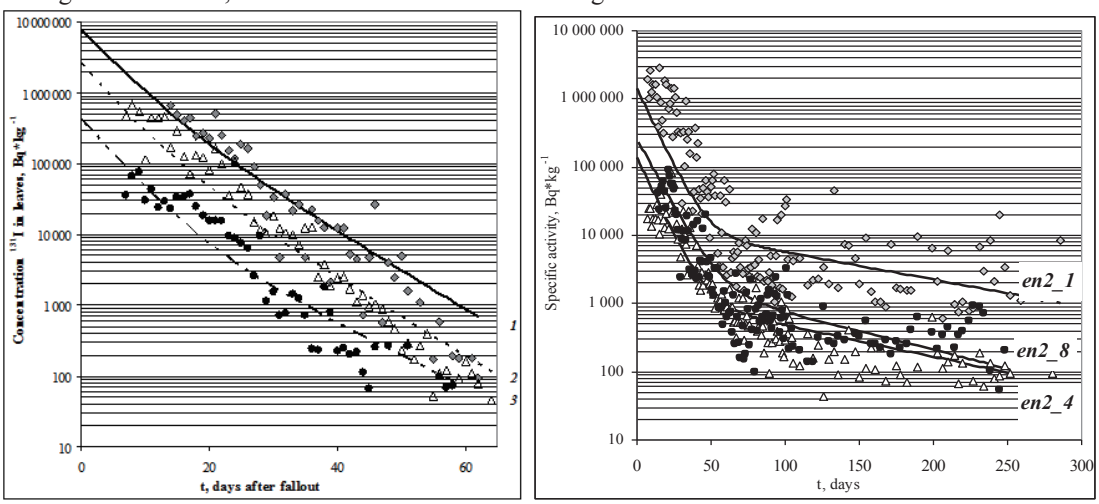


Fig. 6. Points of soil and leafy vegetables contamination monitoring after accident at the Fukushima-1 nuclear power plant located within or near the test polygon.

Monitoring of vegetation contamination was launched only on the seventh day after the accident. Fig. 7 shows that the radionuclide concentration in leafy vegetables this day reached values of 15 - 300 kBq m^{-2} similar to those obtained with using the model AeralPlant. Thus the results of the model and the actual radiation situation in the acute phase of the accident agree satisfactorily.

Subsequently concentration of radionuclides in agricultural products decreases rapidly in time - about 10 times during the first week, but even this time it was much higher standards.



a *b*

Fig. 7. Dynamics of leafy vegetables contamination after the accident at Fukushima-1 NPP according monitoring data (points) and the model of vegetation aerial contamination (Prister, 2008) (curves):
a - ^{131}I for monitoring points en_2.7 (*1*), en_2.6 (*2*) and en_2.3 (*3*);
b - ^{137}Cs for monitoring points en_2.1, en_2.8 and en_2.4.

As one could be seen in Fig. 7, the contamination dynamics are well approximated by two-exponent curve, indicating the possibility of using the AeralPlant model for assessments and forecasting the radiation situation due to aerial contamination of plants. Dynamics of radionuclides concentration in agricultural products must be taken into consideration when organizing and carrying out monitoring in the acute phase of the accident. It should be noted again that these high concentrations of ^{131}I in vegetables and grass appeared according this modeling and actual control data give raise to examine the issue of population thyroid doses reconstruction after the accident at the Fukushima-1 NPP.

In the intermediate and late phases of the radiation accident the plant contamination is caused by the root path of radionuclides intake. To simulate the root contamination of crop production the model SoilPlant was used,

developed by the authors on the basis of summarizing a database of scientific monitoring agrosphere after the Chornobyl accident, which has more than 3,000 factors of ^{137}Cs transfer from the soil of 4 types in harvest of 16 crops (Prister *et al*., 2003).

The ^{137}Cs concentration in agricultural crops is calculated using the formula:

$$SA(t) = \sigma \cdot TF(0) \cdot \left\{ \frac{k_1 - (2 \cdot k_2 + \frac{k_3}{2})}{k_1 - k_2} \cdot e^{-(k_1 + \frac{k_3}{2}) \cdot t} + \frac{k_2 + \frac{k_3}{2}}{k_1 - k_2} \cdot e^{-(k_2 + \frac{k_3}{2}) \cdot t} \right\} \quad (2)$$

where *SA* is specific activity in the plant, Bq • kg^{-1}; TF (0) is the radionuclide transfer factor from soil to plants extrapolated to the time of deposition; k_1, k_2, k_3 are the constants of the radionuclide sorption - desorption rate by soil - absorbing complex for each type of soil.

The model is fundamentally different from the known ones due to it is based on the analytically described TF dependence on the properties of the soil, which are quantified with the value of the complex parameter of agrochemical soil properties *Sef* (Prister, 2002; Prister *et al*., 2003). This parameter is calculated on the basic soil properties that reflect the state of the soil-forming process: the pH of the salt, content of organic matter or humus, absorption capacity or the amount of absorbed bases. *Sef* is defined as the cross-sectional area of three-dimensional space whose dimensions are mutually perpendicular vectors - soil properties.

The dependence of the transfer factor on the parameter *Sef* is described by the formula:

$$TF_{Sef} = TF_0(0) \cdot e^{-\lambda \cdot Sef} \left\{ (1 + 0.031 \cdot \ln(Sef)) \cdot e^{-0.31 \cdot (1 + Sef) \cdot t} + (-0.031 \cdot \ln(Sef)) \cdot e^{-0.055 \cdot (1 - Sef) \cdot t} \right\} \quad (3)$$

The calculation of the ^{137}Cs concentration in agricultural products at the Fukushima test site was made for the worst case scenario - a combination of maximum soil contamination density (according to the soil contamination map) and the minimum value of the parameter *Sef* for each soil type. Without real information on land use, we chose a scenario in which the cabbage grows in areas of residential land village (villages with gardens), placed on the landscape with the angle of slope of less than 3 degrees, and cucumbers - on slopes greater than 3 degrees. Grass, buckwheat and soybeans were placed sequentially on cultivated land. The calculation results of ^{137}Cs contamination of agricultural products in 1 year after the accident at the test polygon for the proposed cropping scenarios is shown in Fig. 8 and 9.

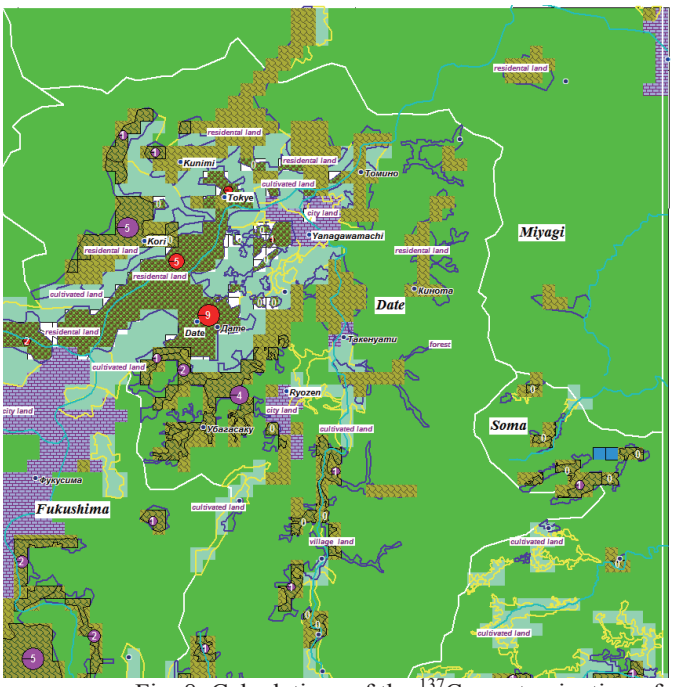


Fig. 8. Calculations of the ^{137}Cs contamination of milk on the cultivated land and vegetables on the residential land village within the test polygon in 1 year after the accident.

Fig. 8 shows that the predicted values of ^{137}Cs concentration in milk in 1 year after the accident, when the grass contamination caused by root path, do not exceed 35 Bq · kg^{-1}, which is less the limit level LL = 50 Bq · kg^{-1}. Consequently, according to the calculations milk obtained from cows that grazed in contaminated pastures of the test site a year after the accident, meet the national standards of Japan concerning the contamination with radioactive cesium. This means that in the long term the concentration of milk contamination will not exceed the LL.

The figures 8 and 9 show the concentration ranges of crops contamination, compared with the national norm LL. For all crops, except for milk, LL of ^{137}Cs content in the product is 100 Bq · kg^{-1}. The figures present the ranges up to 0.7 LL, from 0.7 LL to 3 LL, 3 LL and more. This approach is chosen with a view to further organize monitoring and protective measures implementation.

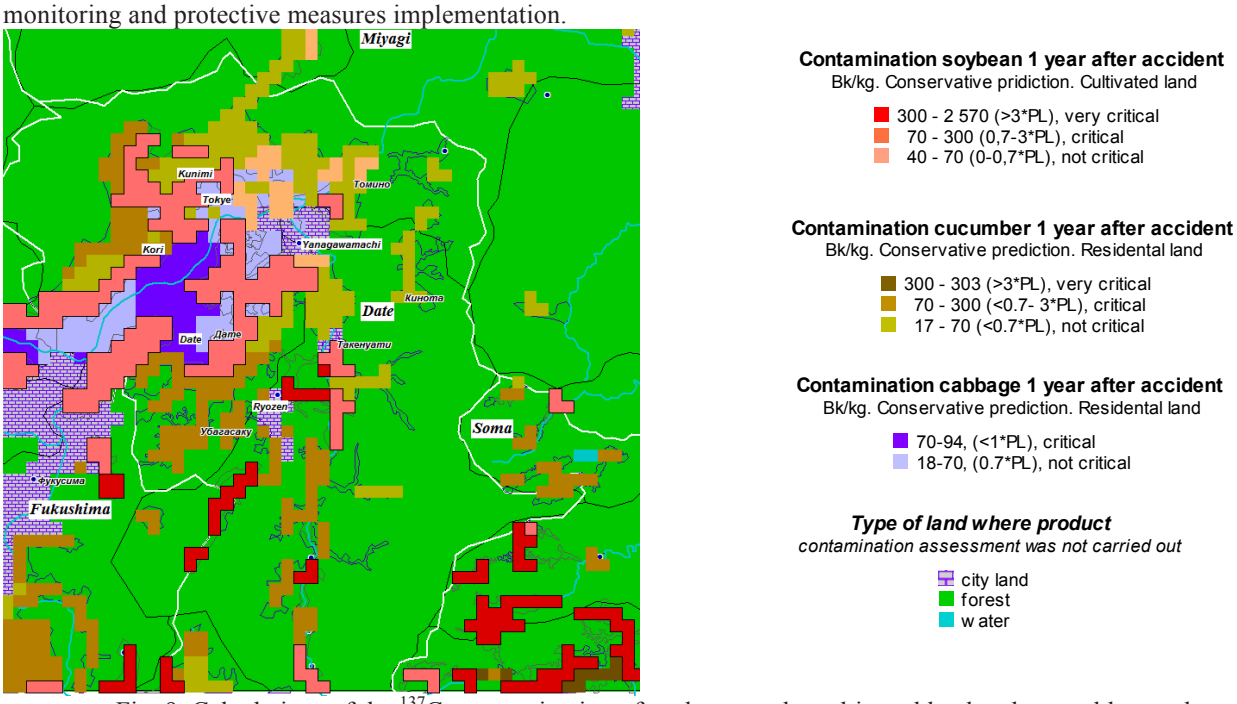


Fig. 9. Calculations of the ^{137}Cs contamination of soybean on the cultivated land and vegetables on the residential land village within the test polygon in 1 year after the accident.

Soybean and buckwheat, which are characterized by high content of a chemical analogue of cesium – potassium, have the highest ^{137}Cs contamination. In large areas the radionuclide concentration is in the range 70 - 300 Bq · kg^{-1} or even more. For cabbage a small number of excess of 70 Bq · kg^{-1} has been obtained.

As for the test site in Ukraine, the concentration of ^{137}Cs in plants, in addition to the density of soil contamination, is determined by the area place in the landscape, the soil type and the crop species. The largest ^{137}Cs concentration in these products taking into account the density of soil contamination is observed on the Brown forest soil, the lowest one is on Fluvisols. In areas with a range of the ^{137}Cs concentration values of from 0,7 LL to 3 LL it's necessary to carry out the primary monitoring (conjugated assessment of the soil and plants contamination levels) and the control (assessment of radionuclide concentrations in the crop within the corresponding element of zoning).

To verify the simulation results shown in Fig. 8 and 9, data were used from the IAEA database of the control of the radionuclide content in products for the test area. Fig. 10 shows the schematic maps of soil contamination and soil types in Date district, Fukushima prefecture used for the calculation. Fig. 11 and 12 present the control data of soybeans for two settlements in Date district (Motomura and Tomino) and of buckwheat for settlements Naganuma and Sirakava Oja compared with the modeling results.

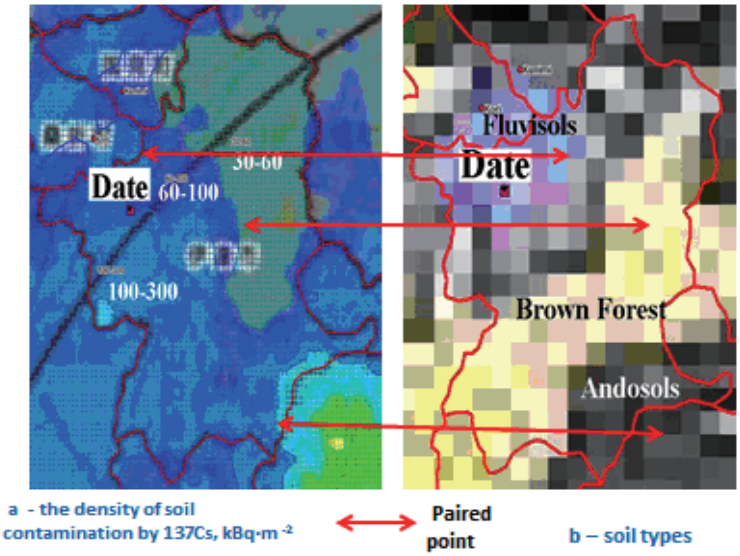


Fig. 10. The schematic maps of ^{137}Cs soil contamination and soil types in Date district, Fukushima prefecture.

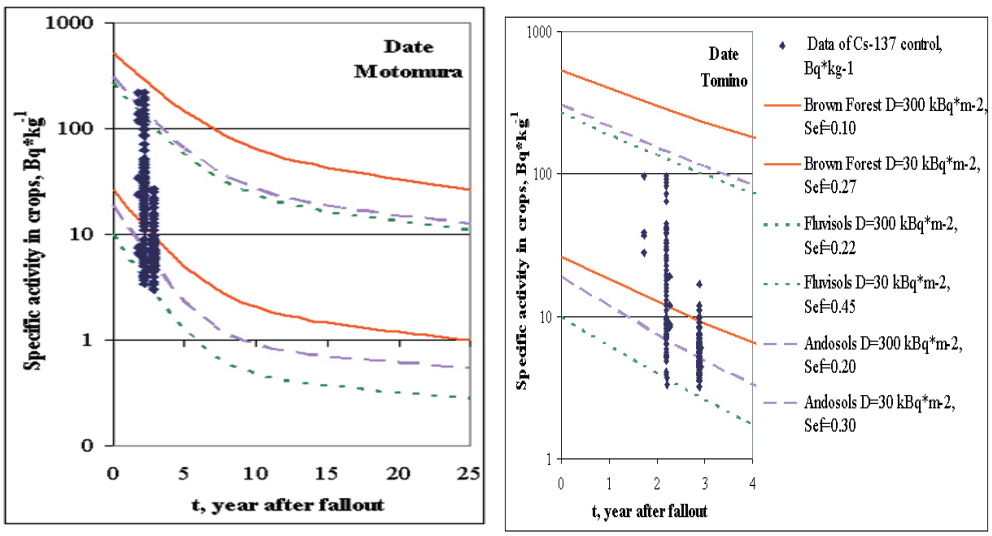


Fig. 11. The forecast values of ^{137}Cs contamination dynamics of soybeans (curves) for combinations of different soil types and the soil contamination density levels typical for the Date county, Fukushima prefecture, and the control data (points) obtained after the accident at the Fukushima-1 NPP.

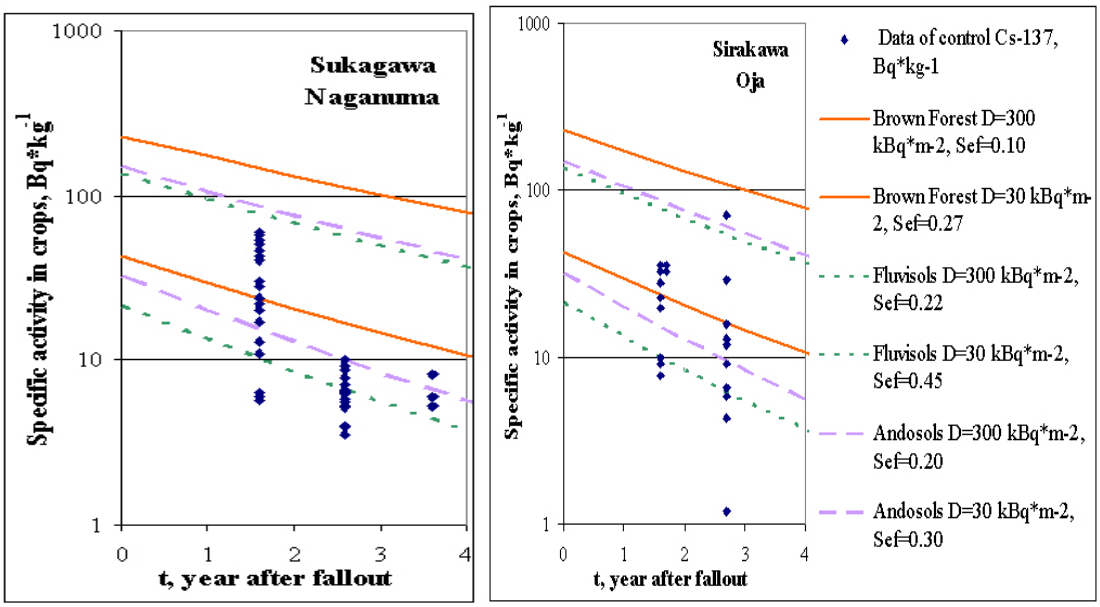


Fig. 12. The forecast values of ^{137}Cs contamination dynamics of buckwheat (curves) for combinations of different soil types and the soil contamination density levels typical for the Date county, Fukushima prefecture, and the control data (points) obtained after the accident at the Fukushima-1 NPP.

The variation of the control data is very significant (more than 25 times), and due to the influence of both the ecological characteristics of the territory and the ^{137}Cs soil contamination density. The data of control held in Tomino in 1 year after the accident show that the ^{137}Cs concentration in soybeans did not exceed 100 Bq · kg^{-1}. The considerable variability in the properties of density and soil contamination was found, which are confirmed by the control data: ^{137}Cs concentration in product varies in the range of an order of magnitude, but in the most cases it is much lower than the standard. However, around some settlements in Date district it exceeded the norm in the soybeans and buckwheat crop in a few times: the maximum values of 300 or more Bq · kg^{-1} agree with the forecast and the most reliable they were observed in the places where the highest soil contamination coincides with the least favorable for plant growth properties (Brown forest soil). On the border of the test site the higher concentration values are possible. The lowest concentration of ^{137}Cs in products corresponds to the combination of low density of soil contamination with the highest agrochemical indicators. To improve its efficiency the scheme of products control should be targeted – it should take into account the spatial distribution of the soil contamination density and its properties the same as it should foresee the representative sampling for control in the same locations at different times in order to assess the dynamics of the radionuclides transfer factors.

The forecast results at a distance of 1-2 km in the south - west of Tomino village (Fig. 9) demonstrate the possibility to produce these crops with a significantly larger content of radionuclides, and to the north the forecast values confirm the data control. Therefore, it's important to clarify - from which exactly territories the products are controlled in Tomino village the same as the question of the control of products which are grown in the territory where there is an excess of the standard for the content of radioactive cesium according to the forecast.

Around Motomura village (near Date) according to the control data the concentration of ^{137}Cs in soybeans in almost half of the samples exceeded the norm. The forecast calculations agree to the control data in this area. According to Fig. 11 the radionuclide concentration in production decreases with time after the deposition and already in the 7th year the probability of soybeans obtaining with the ^{137}Cs standard excess is small. Therefore it is necessary to take into account the forecast data and the dynamics of cesium contamination of agricultural products in the planning of the volume of monitoring and control.

It should be emphasized that the forecast of plant contamination has been carried using the worst-case scenario of a combination of the area ecological features and the levels of soil contamination and provides the widest range of concentrations. If you have actual information about the site the calculated range of contamination levels of production will be much narrower. Analysis of the figures shows that almost all possible scenarios came true when crops are grown on territories with different soils and contamination density. Some exceeding the norm take place, which can be avoided, since it is possible to select such a combination of soil type with the density of contamination (see dashed lines in Fig. 11 and Fig. 12) in which the concentration of ^{137}Cs will be below the standard. It is necessary to manage this process by placing a particular culture crops in areas where the combination "type of soil - contamination density" allows receiving the crop with the content of ^{137}Cs below 100 Bq · kg^{-1}. Another way is conducting agrochemical countermeasures based on soil properties and biological features of crops.

In this study it was suggested to select the zone by the radiation hazard depending on the radionuclide concentration in production due to the root path of radionuclide intake. During the project, we applied the principle of zoning by the ratio of excess concentration to the standards (*criticality*) and have identified the following zoned:

- less than 0.7 standard (products could be used under the quality radiation control),
- the concentration is in the range from 0.7 to 3.0 of the standard (the zone of compulsory use of countermeasures with the efficiency of the radionuclide accumulation reducing up to 3 times under strict radiation control),
- the concentration exceeds the norm by more than three times and can not be reduced to the standard by means of single countermeasures implementation - consumption of products is prohibited without processing,
- the concentration exceeds the norm by from 3 to 5 - 10 times depending on the possibility to apply the complex sequential countermeasures or measures with high efficiency under production (carrying out a radical improvement of pastures, processing of milk into butter, etc.).

The number of zones and intervals for their selection may vary according to specific conditions, the requirements of national regulations and possibilities of countermeasures application, possible monitoring volume etc. We consider the general approach and the algorithm for optimization of the control. The accidents experience shows that the most difficult time in terms of organization of monitoring and production control is an acute emergency period which is the most significant for the formation of the absorbed dose. It makes the problem of optimizing the monitoring and control of agricultural production as very important, especially in small farms. The Fukushima-1 NPP accident is distant from the Chernobyl nuclear disaster for 25 years, during which the level of technical equipment of radiation monitoring services increased considerably. Japan, one of the leading economies of the world, has fundamentally higher potential of emergency response, but the element of suddenness is very difficult to overcome. So monitoring in the area of the Fukushima-1 NPP was started with some delay.

To avoid the human thyroid overexposure it is proposed to restrict or prohibit the consumption of fresh leafy vegetables, whole milk and dairy products from it in an area where, according to the forecast, the ^{131}I concentration exceeds the norm. The ban is to be lifted on the basis of control or operational after-accidental monitoring. The first steps of the agrosphere monitoring program using even the most insufficient means of the instrument should be aimed at clarifying the level of contamination of these products. Therefore, preventive preparations for the organization of monitoring and control, especially working out the principles and guidelines of forming a network that provides representative assessment of the radiation situation in the contaminated zone on the base of the small amount of information, play a significant role in reducing the negative consequences of the accident.

Reducing the number of samples must be accompanied by an increase in the representativeness of the average sample by increasing the number of individual samples, included in its composition. The priority, especially in the initial period, should be given to those areas of the radioactive contaminated territory, which are assessed as critical in the process of preventive preparation according to the degree of criticality. As far as getting the actual information from the monitoring and control subsystems the corrections in the criticality assessment and in the formation of the network should be made.

3.6 The subsystems of the radiation situation monitoring in the agricultural sphere, produce quality control and countermeasures

The subsystem of the radiation situation monitoring carries out following tasks:

- formation of a network of monitoring and products control for the two phases of the accident on the basis of preventive assessment of the territory (potential criticality) and available monitoring data on the contamination of products (implemented criticality),
- the territory zoning by class of specific activity of radionuclides in products in order to optimize - the main attention should be paid to critical areas of the territory,
- visualization of solution with output of the formed monitoring network in the form of the thematic map.

Formation of monitoring network (calculation of the monitoring samples number and their spatial distribution) is carried out using the algorithm described in the project 5953 technical report T04. The total available monitoring volume (number of samples that can be selected and measured in a given period of time) is distributed between objects. It uses the method of weighting coefficients that characterize a particular object of monitoring: the area, the number of resident population, the possibility of further use in agricultural production, and others. Optimization of monitoring is the key to timely information obtainment for decision-making on countermeasures implementation.

The principal difference between the monitoring system and the control system is that it involves the assessment of technologies and production conditions - place location, soil type, landscape type, the density of soil contamination, the phase of plant development at the time of contamination and sampling and others. It enables not only to draw a conclusion about what factors determine the level of contamination of the sample but to justify

the choice of the necessary countermeasure. Understanding the causes that led to a certain level of contamination allows to determine purposefully the sampling points under monitoring continuation.

The subsystem of produce quality control sets the task to evaluate the actual content of radionuclides in products without respect to the conditions of production in order to determine its compliance with national standards and possibility uses it as food. The control uses standard international or national procedures. Product sampling for quality control can be carried out both at the place of production and in the process of receiving, selling and processing. It is important when sampling for the control to register a place of product farming that allows to place it in the database of appropriate zoning element and to use for evaluation of the radiation situation within the boundaries of this element. Rules of sampling are governed by the respective regulations, enacted at the time of the accident by interested agencies.

The countermeasures subsystem includes programs of planning and evaluation of the countermeasures effectiveness after their implementation (monitoring and products contamination control before and after the countermeasure) with mandatory monitoring of their implementation according to developed programs. Countermeasures are chosen so that their effectiveness is corresponds to ratio of excess of nuclide concentration in the production to the standards. If the effectiveness of individual countermeasures is not sufficient a set of measures should be applied consecutively.

Methodological aspects of monitoring are essential. It is very important to ensure conjugation of soil and plant samples in the space – the average samples of plants and soil should be representative for the same zoning element regardless of its size. In the system developed in the project a conjugation of information in space at different trophic levels of the food chain "soil - plants - animals – man" is achieved by the area zoning network the same for all information layers, divided by the ecological coordinates - basins, elementary landscapes and their elements, land use objects. Information on each layer in the database is collected in the cell of each zoning object. Analysis of each layer horizontally allows to identify patterns of spatial distribution of the water and wind radionuclides migration intensity between the elements of the landscape, and vertically – laws of radionuclides transfer from one level of the food chain to the other.

Thus, the principles of integrated data processing, analysis, forecasting the radiation situation with the formation of the monitoring and agrosphere products control network using the set of models and modern information products (GIS, databases) subject to natural and geographical features of the contaminated areas were developed and implemented.

3.7 Functional diagram of the comprehensive system of the radiation situation forecasting, agrosphere monitoring and production radiation quality control

Schematically, the system structure is shown in Fig. 13.

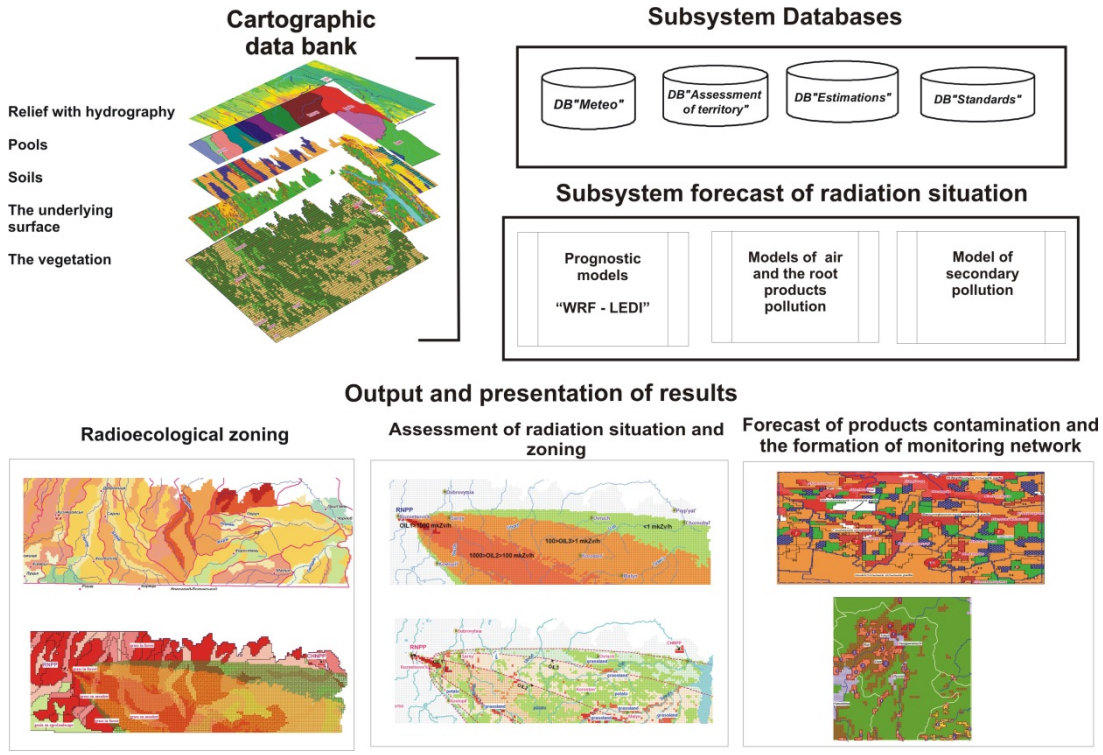


Fig. 13. Functional diagram of the comprehensive system of the radiation situation forecasting, agrosphere monitoring and production radiation quality control.

4. Conclusions

1. The analysis of the problem shows that in a result of all severe emergencies occurred in the nuclear power industry and power engineering - Chelyabinsk (USSR), Windscale (Great Britain), Chornobyl (USSR), Fukushima (Japan) large areas of intensive agriculture land, where significant numbers of the population lived, were subjected to radioactive contamination. These accidents were recognized as agricultural emergencies. At the same time the after-emergency regulations of agrosphere monitoring do not currently exist up to now. In the event of a severe accident at the nuclear power plant information of existing systems of environment radiation control is not sufficient to manage the process of doses forming and timely implementation of adequate countermeasures in the agricultural sphere.

As the main factors of radiation situation formation the external radiation dose rate and density of soil contamination are still remained without consideration of soil properties and landscape features, though the internal exposure dose at equal deposition density for soils of different quality can range up to two orders of magnitude.

2. The dose of internal exposure due to the use of contaminated agricultural products during long time (30 years after the Chornobyl accident) may cause the significant, and in some circumstances a decisive contribution into the total radiation dose of the population. However, evaluation of radiation situation in the agricultural sphere is not highlighted as one of the main tasks of monitoring.

In the course of the project implementation it was grounded and proposed to supplement the system of emergency radiation monitoring of the territory of the NPP possible impact with the section "Radioecological monitoring of agrosphere", aimed at clarifying the purely agricultural aspects of the problem of environment radioactive contamination. It enables for governing bodies to get information about - Where? When? Which? - countermeasures are necessary to carry out for preventing population overexposure.

3. The methodology of long-term preventive and after-emergency monitoring of agrosphere, developed during the project 5953 implementation, to account as a scientific and methodological basis for developing methods of agrosphere monitoring after severe accidents at nuclear power plants.

4. It was developed the general scheme of radioecological monitoring, which includes: the preventive territory preparation, including the radio-ecological zoning; assessment of the territory potential criticality and assigning radio-ecological parameters to be entered in the database; assessment of the radiation situation using a numerical simulation of the accident consequences, including the calculation of the contamination of agricultural products and comparison with standards; forming a monitoring network and planning countermeasures taking into account critical areas of the radioactive contaminated territory and some natural objects.

5. Experience of severe accidents minimization shows that the information necessary to predict radiation situation within the contaminated territory has been collected and classified after a considerable period of time after the accident, when large part of the human exposure dose has been formed. That is why the principal difference of the developed monitoring methodology is preventive (before an accident) targeted radioecological study of the agricultural area that may be subjected to radioactive contamination after a serious accident at the plant for all possible weather scenarios. The stage of preventive preparation finishes with the creation of the radioecological model of territory using GIS technologies, cartographic layers and databases of the parameters necessary for quantitative modeling of the radiation situation formation, which allows already in the acute emergency stage to implement monitoring and countermeasures preventing the formation of a large part of the human exposure dose.

6. The feature of the developed methodology of agrosphere radioecological monitoring is the preventive comprehensive basin - landscape zoning of nuclear power plants surveillance zones and zones of their impact in the case of severe accident, as a crucial principle of monitoring optimizing and ensuring the countermeasures targeted implementation. The basin is considered as the primary objective existing unit of territory, radiological characteristics of which - the landscape, the type of underlying surface, the types of soil and plants – enable to take account of the impact of landscape and geochemical conditions of territory on radionuclides deposition, to model their migration in the "soil-plant" system and formation of the population exposure caused by radionuclides intake into animal and human body.

7. The elementary landscape or its structural elements are considered as ecologically homogeneous area, where the radionuclides deposition density or other radio-ecological characteristics within it are allowed of averaging. This structural unit may be considered as a unified object of prediction, monitoring, planning and countermeasures implementation. The methodology of the territory zoning allows to made the superposition of thematic map layers (ecological characteristics, soil properties, etc.) at different scales (national, regional and local), to optimize and combine the spatial and temporal distribution of results of agricultural products radionuclide contamination modeling, monitoring, control and countermeasures implementation. The proposed approach enables targeted monitoring and countermeasures on specific areas with compliance of priorities.

8. The process of zoning is difficult to be fully formalized. You can define the basic rules and principles, but each area has its own characteristic features which should be reflected in the classifiers and taken into account in the analysis and synthesis of objects characteristics at different scales. The report provides examples that can be used as patterns. Zoning requires broad knowledge, experience and intuition of user.

9. Selected areas and their structural elements are ranked on potential criticality - the ability to form larger internal dose at equal density of soil contamination. The territories criticality is assessed by the integral index, which is determined by expert evaluation of the most influential indicators - types of elementary terrain, soil, underlying surface and vegetation. The contribution of the individual parameters criticality into the integral index is calculated by the weighting coefficients method. The integral index reflects the priority for elements of territory or object in the planning of monitoring, control and countermeasures. The positive correlation between the integral index of criticality for the territories of Ukraine contaminated by the Chornobyl accident and the average dose of inhabitants of settlements in these territories evaluated using the data of the special program of dosimetry passportization in Ukraine, which confirms objectivity this approach to criticality assessing.

10. In all severe accidents the importance of radioactive contamination of grass was noted, which greatly exceeds contamination of other agricultural products. A key role in grass contamination plays a set of factors: a place in the landscape, soil type, vegetation type. Grass makes a significant contribution to the formation of radiation situation in the acute and late phases of the accident. Floodplain, grazing land and meadow landscapes, contaminated grass, hay and livestock products from which at equal density contamination make a larger contribution into the dose formation in comparison with other landscape elements are attributed to potentially critical objects. This determines their priority under organizing agrosphere monitoring and implementation of countermeasures.

It was established that the density of surface contamination with radionuclides is not the only parameter that determines the radiation hazard and criticality of landscapes main elements - primarily they are determined by territory ecological features. Attention of monitoring should be focused on the main most critical objects.

11. One of the important tasks of the after-emergency response is to prevent the production of agricultural products exceeding the contamination standards. Analysis of the accidents shows that the efficiency of quality control system, despite its obvious redundancy is low, as it does not provide identifying the sources and the causes of products contamination. It is necessary to foresee preventively including the system of products radiation control as a subsystem of monitoring provided coordination of their operation. This conclusion is confirmed by the analysis of control systems in the area of the Fukushima-1 NPP accident, which registers cases of excess of the pollution standards for soybeans, buckwheat, rice, but doesn’t determine their reasons, doesn’t enable to find places of the most critical areas and to prevent the production of such contaminated products.

12. The methodology of the radiation situation forecasting has been developed after the accidental release using the methodology of radioecological zoning and comprehensive modeling based on the set of models: the model of the probable contamination territory; radionuclides atmospheric transport; radionuclides deposition on soil and vegetation; radionuclides resuspension and following atmospheric transport; foliar contamination of plants; radionuclides behavior in systems “soil – plant”, “food - the body of animals - livestock products”, dose models. The models input and output data are coordinated in space and time, which ensures their versatility and ability to use together.

13. Forecast of the radiation situation formation using models preventively adapted to specific synoptic and landscape conditions of the region enables in the acute phase of the accident to reduce essentially the time and the area of restrictions in the mode of population life. The algorithm of radiation situation assessment is perfected by comparing forecast or measured values with national and international standards, their ratio is used as basis for countermeasures planning.

14. The methodology of creating a network of radioecological monitoring and assessment of the radiation situation in the NPP influence zone in the emergency event has been developed. The monitoring methodology allows to make a representative estimation of radiation situation parameters for each zoning element of appropriate scale, that is achieved by the formation of monitoring network in space and the adequate methodology of conjugate "soil - plant" sampling representative for evaluated territory. The distribution of the number of sampling points between the zoning elements was calculated using the weights considering the main factors influencing the formation of the population exposure: by the object area, by its criticality class and the contribution of selected objects in the radionuclides intake into the human body with diet, etc. As an example, the distribution of the sampling points number for the basin was calculated and a monitoring system network was formed for the scenario of contamination of the test areas.

15. Approaches and requirements for databases and the system of collecting meteorological, radioecological and other information are formulated to assess the radiation situation in the area of emergency response in accordance with the guidelines of the IAEA and regulations for radiation monitoring of the NPP environment. Requirements include a method and frequency of data input into information bases, a list of their formats, classifiers, methods of statistical analysis, quality control parameters and methods of data storage.

16. Sources of input data are defined to create digital maps and the GIS technologies are designed to process and analyze spatial information for radiation zoning.

The algorithm for creating cartographic documents using remote sensing data and numerical forecast of territory contamination is proposed that can be used for decision support in case of accidents at nuclear power plants.

Conclusions for the Fukushima contaminated zone

1. Analysis of the agricultural products radiological control database in the Fukushima province showed that overall in 2011 - 2013 the radiation situation in crop production was satisfactory and there was no cause for concern. The concentration of ^{137}Cs in products on the whole was below the norm. However, in soybeans and buckwheat harvest it varies within an order of magnitude, and in the Date district it exceeded the norm by several times around several settlements.

2. It was concluded that up to now the control scheme is unaddressed, that is it does not account for the spatial distribution of the soil contamination density and soil properties that affect the accumulation of ^{137}Cs by plants, which reduces the effectiveness of the control. In addition, sampling for control in the same locations at different times to assess the dynamics of the radionuclides transfer factors is not provided. Scheme of control needs to be adjusted, and the control system is expedient to coordinate and harmonize with the monitoring system.

3. Using the cartographic data from the Internet made it possible to carry out zoning of Date district at the regional level, which showed that there is considerable variability in soil properties and density of pollution - more than an order of magnitude. The standards excess in yield contamination or its maximum values are observed most likely in places where the maximum levels of soil contamination coincide with the least favorable soil properties for plant growth - brown forest etc. The lowest concentrations of ^{137}Cs in the product correspond to the combination of low density of soil contamination with the highest agrochemical indicators of soil.

4. Using published data on soil properties of Japan the value of integrated assessment of soil properties *Sef* was calculated. Despite the significant differences in morphology and agricultural chemistry of soils in Japan and Ukraine, the interval of *Sef* values for main soil types in Fukushima prefecture - 0.10 – 0.45 - was very close to the range of this value for main soils in Ukraine - 0.11 - 0.41. This suggests the possibility of applying the Ukrainian "soil - plant" model used in the project for the Japan conditions.

5. The results of ^{137}Cs measurements in soybeans and buckwheat in Date district, Fukushima prefecture in 2012- 2013 from the monitoring database confirm the forecast of the Ukrainian model of radionuclide migration in the system "soil - plant" using published data on agrochemical properties of soils in Japan. The forecast of the dynamics of soybean contamination levels over time was made using the Ukrainian kinetic model, according to which in 4 years after the accident ^{137}Cs concentration in soybean will decrease about 3 times due to natural processes of nuclide fixation in the soil only.

6. As followed from the comparison of the model results with the available data of the control database, the Ukrainian model of radionuclide migration in the system "soil - plant" generally correctly reproduces the dynamics of the concentration and changes in transfer factor within 4 years after the accident, but for more reasonable conclusions the activity values in soybean from the database obtained in the same sampling points for 4 years after the accident should be compared (unfortunately, their coordinate location is absent in the existing database). Based on the results, it is hoped that with using the models one can explain in more detail the variability of control results and to predict crop contamination.

7. To obtain accurate and reliable forecasts of the ^{137}Cs concentration in agricultural production using the model of radionuclide migration in the "soil - plant" system the values of agrochemical properties of soils (pH_{KCl}, soil organic matter, the amount of absorbed bases) should be obtained for typical (reference) control points, in which conjugated samples of soil and plants should be selected by all means with their coordinate location to the territory analyzed.

8. The analysis of the possibility of using the Radiocesium Interception Potential (RIP) value was made, which was suggested in Japan to predict migration of cesium in the system "soil - plant". It is concluded that the use of RIP to predict crop contamination is not yet sufficiently justified. The correlation between the RIP and the ^{137}Cs accumulation coefficient in vegetation is virtually absent. The RIP does not clearly reflect the diversity of soil properties. In addition, RIP is independent of time, that it does not consider the kinetics of sorption processes, which determines the TF dependence on time since the fallout. Currently using RIP as a prognostic indicator is very problematically and requires a pilot study and additional experimental justification.

9. The evaluation and forecast of the aerial contamination of crops due to the ^{137}Cs resuspension in the area around the Fukushima-1 NPP was made. For this purpose the model of the radionuclides resuspension and following atmospheric transport from the surface planar source under natural resuspension and technological impacts has been developed and the model of aerial plant contamination has been used.

For 5 monitoring posts (Minamisoma, Ishigami; Minamisoma, Ota; Kawamata, Tomita; Nihonmatsu, Shibukawa; Nihonmatsu, Harimichi) in Fukushima prefecture, where in 2013-1014 measurements of radioactive contamination of soybeans were carried out, the model estimations have been made. It was obtained that in 2-4 years after the formation of the deposition field for all 5 monitoring points the value of ^{137}Cs aerial contamination of vegetation due to resuspension is from 2.5 to 6.3% of the cesium total content in soybean during this period and

plays no significant role in the contamination of the crop. These estimations are conservative and represent the maximum expected value of vegetative organs contamination.

10. It was made the analytical review of the practice of countermeasures application in order to reduce ^{137}Cs contamination of agricultural products in the region of the Chornobyl accident, which can be used in the area of the accident at the Fukushima-1 nuclear power plant. It was shown that now (4 years after the accident) in Japan the effective countermeasures that reduce the ^{137}Cs transfer in harvest of farm crops are agrochemical reclamation (liming, application of organic and mineral fertilizers, potassiun especially). The effectiveness of these measures depends on many factors: the type of measure, type of soil and its properties, plant species and other. In soils containing exchangeable potassium large than 10 mg / 100 g an effectiveness of potassium fertilizer as a means of reducing yield contamination diminishes sharply. Japan's experience confirms these conclusions. Increasing doses of potassium fertilizers above 120 - 180 kg / ha is inexpedient.

11. Generalization of the Chornobyl experience of Ukraine and other countries shows that the most effective means of obtaining high yields with low ^{137}Cs contamination is the complex fertilizer introducing. The dose of nitrogen fertilizer is calculated under planned yield of this crop. The doses of phosphate and potash fertilizers to minimize the radionuclides intake should be increased, respectively, 1.5 and 2.0 times relative to nitrogen doses calculated for the planned harvest.

12. The results of long-term observation of the ^{131}I and ^{137}Cs concentration dynamics in the plant leaves from the database in 2011 were used for verification of the aerial plant contamination model developed by the project team. For all 9 stations the dynamics of ^{131}I and ^{137}Cs concentration in leaves could be adequately described by the two-exponent model. The periods of leaves semi-cleaning for ^{131}I (2.8 and 6.0 days) and ^{137}Cs (4.4 and 47 days) on average in Fukushima are satisfactorily agree with the average for all data after the Chornobyl accident and experiments 3.2 and 5.4 days for ^{131}I and 6.6 and 59 days for ^{137}Cs.

The large absolute values of ^{131}I (0.29 – 12.3 MBq · kg^{-1}) and ^{137}Cs concentration (0.045 – 2.90 MBq · kg^{-1}) in leaves on the 7th day after the accident and at a considerable distance from the reactor could lead to the intake of biologically significant quantities of ^{131}I into humans and animals body and demonstrate the need for monitoring of aerial pollution of plants immediately from the first day after the accident. Obviously it is advisable to analyze retrospectively at such points the radionuclide content in cow milk (if it was produced there) and in the thyroid gland of people at monitoring points with high concentrations of ^{131}I and ^{137}Cs in leafy vegetables.

13. The estimation of the ^{137}Cs contribution into contamination of the chickens meat and eggs at outdoor keeping has been made. For chickens fattening in Japan clean feed are using, so contaminated soil is considered as a single radionuclide input source. The allowable nuclide concentration in the soil is limited to the value of 1500 Bq • kg^{-1}. Conservative estimates made using the maximum values of the ^{137}Cs metabolism parameters in the chickens body show that the ^{137}Cs concentration in chickens meat and eggs at outdoor keeping will be below the norm in Japan 100 Bq • kg^{-1}.

5. Applicability and Recommendations for Japan

The general methodology developed as a result of the project for organizing radiation monitoring of agricultural production areas, contaminated as a result of radiation accident at the nuclear power plant, and quality control on them can be used as part of the emergency preparedness system (for creating a network of radioecological monitoring taking into account the availability of critical areas of the radioactive pollution territory, the same as for planning and optimizing countermeasures) and the emergency response system (for operational forecasting the consequences of a radiological emergency at its different stages).

The national authorities responsible for emergency preparedness may be customers of work on the adaptation of the developed methodology to geographic and environmental conditions of the territory of the NPP potential impact, which includes preventive preparing information on the territory (including its radio-ecological zoning).

Formalization of processes of preventive preparation of data about agricultural production territories in the area of existing nuclear power plants potential impact, complex radioecological zoning on the basis of the developed methodology, improvement of systems of radiation situation assessment and prediction taking into account the radioecological features of the territory can be performed in case of development the document "Guide for the long-term monitoring of the agricultural sphere in the case of emergency" based on the methodology developed (potential customers of this work are IAEA, FAO, DOE USA).

Proposals for the Fukushima zone

1. To arrange a seminar with Japanese experts, deciding the feasibility of using the methodology developed for the Japan conditions under the project team support - implementation of the methodology in the agrosphere monitoring system in the area of the accident at the Fukushima-1 NPP.

2. To carry out mapping and radioecological zoning the territory using data about the deposition density and agrochemical properties of soils (pH_{KCl}, soil organic matter, the amount of absorbed bases) at regional and local levels with the specification of the potential critical areas at local level in accordance with the methodology developed in the project, so it enables to focus the control on the most critical areas and to conduct targeted countermeasures in the places where ^{137}Cs accumulation in produce is the most likely. The appropriate recommendations have been provided.

3. To form an optimal carefully scientific reasoned network of radioecological monitoring. The principles of its creation were described in the document "Recommendations for the long-term monitoring of the agricultural sphere in the zone of the accident at Fukushima-1 nuclear power plant" (the annex to the Q4 report). To conduct a scientific monitoring for establishing the dependence of the TF main crops on the properties of soil. During a scientific monitoring to determine RIP. Every RIP evaluation should be accompanied by information on the agrochemical properties of soil, the value of the integrated parameter of soil properties Sef and the transfer factor for main crops.

4. To conduct zoning and scientific monitoring in the part of the evacuated zone where evacuees likely to return or the agriculture recovery is possible for reliable preventive (before a population return) predicting the levels of radioactive contamination of the products.

5. To check efficiency of the ratio of fertilizer elements N: P: K = 1.0: 1.5: 2.0 in applied fertilizers to reduce the accumulation of ^{137}Cs in crops for the conditions of Japan.

An important veterinary lesson of the Chelyabinsk and Chornobyl accidents is that both before accidents and after them the formal radiation standards for veterinary services and the regulations for corresponding section of radiation monitoring were not been introduced. In Ukraine, Belarus and Russia it has result in an unjustified slaughter of more than 150,000 head of cattle. It was recommended to include into the program of long-term radioecological monitoring of agricultural areas the veterinary examination and the forecast of possible doses to the farm animals' body, their possible radiation damage estimation, and developing the recommendations for the treatment of contaminated and exposed animals. It should be recommend to the IAEA and countries concerned to formally put in place the corresponding guidance in case of accidents.

Forest Ecosystem – Mushrooms

STCU-5954

Monitoring of radioactive pollution of forest ecosystems after accident on Chernobyl NPP

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ISTC/STCU Fukushima Expert Meeting

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Summary Report – project # 5954

on work performed from *September 01, 2013* to *September 01, 2015*

Institute for Nuclear Research of National Academy of Science of Ukraine

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1 Brief description of work: objectives, and expected results

Following the accident at the Fukushima Daiichi NPP, radionuclides including long-lived biologically active ^{137}Cs got into environment. According to literature data (Yoshida S., Muramatsu Y. - Contaminated Forests. Recent Developments in Risk Identification and Future Perspectives.- 1999; Kaneko S. - ISTC/STCU Technical Working Group Meeting on the environmental assessment for long term monitoring and remediation in and around Fukushima, - 2012), around 67% of Japan and 71% of Fukushima Prefecture are covered with forests. As a result of the accident, the Fukushima Prefecture forests are contaminated with ^{137}Cs. The forests remediation problem could not be solved without preliminary thorough study of ^{137}Cs accumulation in various forest and soil objects. In addition, ^{137}Cs behavior prognosis estimations have to be developed for forest ecosystems at different stages of the after-accident situation development.

On the first stage, the project #5954 planned to develop a radiation monitoring program, adapted to the climatic and geographical conditions of Fukushima Prefecture. The results of a customized radiation monitoring program (data of the content of ^{137}Cs in different objects of forest ecosystems) expected to be used for further studies and forecast.

Secondly, within the project framework we expected to estimate the redistribution of ^{137}Cs in forest ecosystems to create a model of long-term behavior of ^{137}Cs in forest ecosystems of Japan. The basis for the model were almost 30 years of radionuclide data accumulation in different objects of forest ecosystems collected in the Chernobyl exclusion zone. Verification of the model is to be performed using results of measurements of ^{137}Cs contents in different objects of forest ecosystems from Fukushima Prefecture.

The project main objective was to forecast the behavior and redistribution of ^{137}Cs in the contaminated areas, using mathematical and statistical analysis of the data and the model (stage 3). This forecast can help to develop recommendations for the use of different parts of forest ecosystems for the needs of population in the long-term period after the incident and to develop the emergency plan. Data on content of 137Cs in the fruit bodies of mushrooms of different species and weight of different species of mushrooms per 1 sq. km is to be obtained in different forest ecosystems of Fukushima Prefecture. These data enable us to determine species of mushrooms-concentrators of this radionuclide in the forests of Japan and to forecast the expediency of remediation of forest soils in Japan with the help of mushrooms.

2 Modifications of work after April 2014 meeting

In April 2014 in Tokyo, the project manager Zarubina reported on the implementation of project. Supervisory Committee of ISTC/STCU, together with colleagues from Japan recommended that the project participants to conduct work (sampling of fruit bodies of mushrooms and soil) in the territory of Japan, contaminated after the accident at the Fukushima-1 in order to maximize the adaptation of existing data to the immediate problems of mushrooms contamination in the forest of Japan.

In September - October 2014, N. Zarubina on the invitation of Japanese colleagues from the University of Fukushima, Institute of Environmental Radioactivity, visited Japan and jointly conducted sampling of mushrooms and soil from several sampling sites on the territory of exclusion zone of NPP Fukushima-1. Measurements of specific activity of ^{137}Cs in collected samples were carried out by Japanese colleagues at the University of Fukushima, Institute of Environmental Radioactivity. The results obtained were used to update achievements of all three stages of the project.

Also, Supervisory Committee of ISTC/STCU has proposed to revise scope of the second stage - Research of the expedience of remediation of forest 'soils by means of fruit bodies of mushrooms. However, carrying out sampling in the exclusion zone NPP Fukushima-1 and interpreting data, unexpectedly allowed us to get new developments. Even according to preliminary estimates from 2014 sampling session, the effectiveness of fruit bodies remediation method compared to the methods that are used at present at the contaminated areas of Japan has been proved. The feasibility of remediation of forest soil using fruiting bodies of mushrooms based on Japanese data has been confirmed.

Supervisory Committee of ISTC/STCU has recommended verifying project models using data from the Fukushima Prefecture territories. These recommendations were taken into account and the final models were verified using sampling results from Japan.

3 Technical approach, method, experiments, theory, etc

For implementation of the program project it is necessary to collect main objects of forest ecosystems, the preparation of samples for measurements and to measure content of ^{137}Cs and ^{134}Cs in samples.

Fruit bodies of mushrooms are selected on landfills sites quickly (preferably by 1 day) to avoid the influence of weather conditions on the accumulation of radionuclides by mushrooms. The fruit bodies of mushrooms should be selected of an equal maturity level. Fruiting bodies are primary pre-cleaned from surface contamination and placed in plastic bags (for each species) and marked.

In the laboratory, samples of mushrooms are homogenized and placed in tight containers for subsequent gamma-spectrometric studies. The prepared for measurements samples of fruit bodies are stored in a freezer at -18 ^{0} C until measurement.

Along with samples of mushrooms, soil layers are sampled by the envelope method. Each sample is placed in a separate container, is labeled and transported to the laboratory. In the laboratory, soil samples are dried to air-dry weight and necessary part exposed to gamma spectrometric examination.

Depending on the task, samples of higher plants may be taken simultaneously with soil and mushrooms. Samples are packed into bags, labeled and transported to the laboratory. In the laboratory they are dried to air-dry weight, crushed and placed into calibrated bowl for gamma-spectrometric investigations.

Measurements are made on gamma spectrometer installation on the basis of the coaxial semi-conductor detector of GC6020 "CANBERRA" type, with the registration of the radionuclide content on the personal computer.

Specific activity of ^{137}Cs in samples is calculated as follows:

$A(Bq/kg) = (S - S_f)/(\varepsilon * I_\gamma * t_{meas}(s) * m(kg))$,
where S – area of a gamma line of 661.6 keV ^{137}Cs in the spectrum of the sample;
S_f – area of a gamma line of 661.6 keV ^{137}Cs in the background spectrum;
ε - absolute photo-efficiency of registration of gamma-ray 661.6 keV ^{137}Cs with this geometry of measurements
I_γ – quantum yield of gamma rays (for gamma-quantums 661.6 keV ^{137}Cs – 0.851);
t_{meas} – time of the measurement (s);
m(kg) – weight of sample (kg).

The measurement error of the specific activity of ^{137}Cs in the samples is calculated as follows:

$$\delta A = \sqrt{(\sqrt{\delta S^2 + \delta S_\phi{}^2})^2 + \delta\varepsilon^2 + \delta I_\gamma{}^2},$$

where
δA - relative error in determining of specific activity of ^{137}Cs in the sample (%);
δS - relative error in determining of square area of gamma lines of 661.6 keV ^{137}Cs in the gamma spectrum of the sample (%);
δS_ϕ - relative error in determining of the area of ^{137}Cs - gamma lines in the background gamma spectrum (%);
$\delta\varepsilon$ - relative error of absolute photo-efficiency of gamma-ray 661.6 keV ^{137}Cs in this geometry measurements (%);
δI_γ - relative error of the tabular values yield gamma-quantum 661.6 keV for ^{137}Cs (%).

Value ε is calculated for different geometries of measurements using the code MCNP-4C based on the known detector geometries and measurements. The correctness of the calculations was checked by measurements of standard source - exemplary source of special OYSN EM66 with certified specific activity of ^{137}Cs. Size of error was estimated as 5% for 1 (P = 0.68)σ based on the assessment of various factors affecting to calculations by Monte Carlo using the code MCNP-4C.
The value δI_γ was taken from the program package NUCHAT firm CANBERRA and equaled 0.2%.
Error values t_{meas} and m can be neglected.

5.1 Substantiation and working out of recommendations about carrying out of monitoring of forest ecosystems of Japan polluted as a result of failure on the atomic power station Fukushima-1

Regional level radioecological monitoring, which is performed within a radiation accident zone, is aimed at the collection of information about environment radiation state to support the decision-making process during the management of the territory suffered from radioactive contamination. From a general point of view, the territory management is reduced to two tasks: the radiation safety and the employment of the territory. The assurance of the radiation safety includes the following goals:

- The estimation of general parameters for the contamination of natural environments, their critical components (water, air, natural resources);
- The analysis and prognosis of radiation-dangerous situation;
- The estimation and control of radioactive substances' flow out of the territory.

The employment of a territory usually includes the following actions:

- Natural resources use;
- Transport and communication activity;
- Radiological decontamination and other work of similar type;
- Ecological control of the territory (forestry, water-control practice).

Considerably large data bases of up-to-date radioecological information are needed for these activities' safe performance and the estimation of their consequences. The radioecological monitoring may only provide these data.

During the monitoring, initial data are collected, which are transformed into information – databases, cartographic documents, analytic notes and reports, answers for information requests, etc. The form and the sense of the information are mainly determined by the tasks of its users – government executive bodies. At the same time, the existent demand for the information is considered as well as its potential employment for the solution of tasks not requested by practice yet.

The monitoring performs three functions – surveillance, state estimation, and prognosis. The surveillance means preprocessed data obtained from a surveillance network. The state estimation is the information preparation pertaining to control tasks (e.g., surveillance results estimation from the standpoint of national standards for radiation safety). The prognosis means the development of options for the situation change in time basing on the obtained data. Modeling techniques, trend separation and other are employed for this purpose. General scheme of monitoring is shown in Fig. 3.1.1.

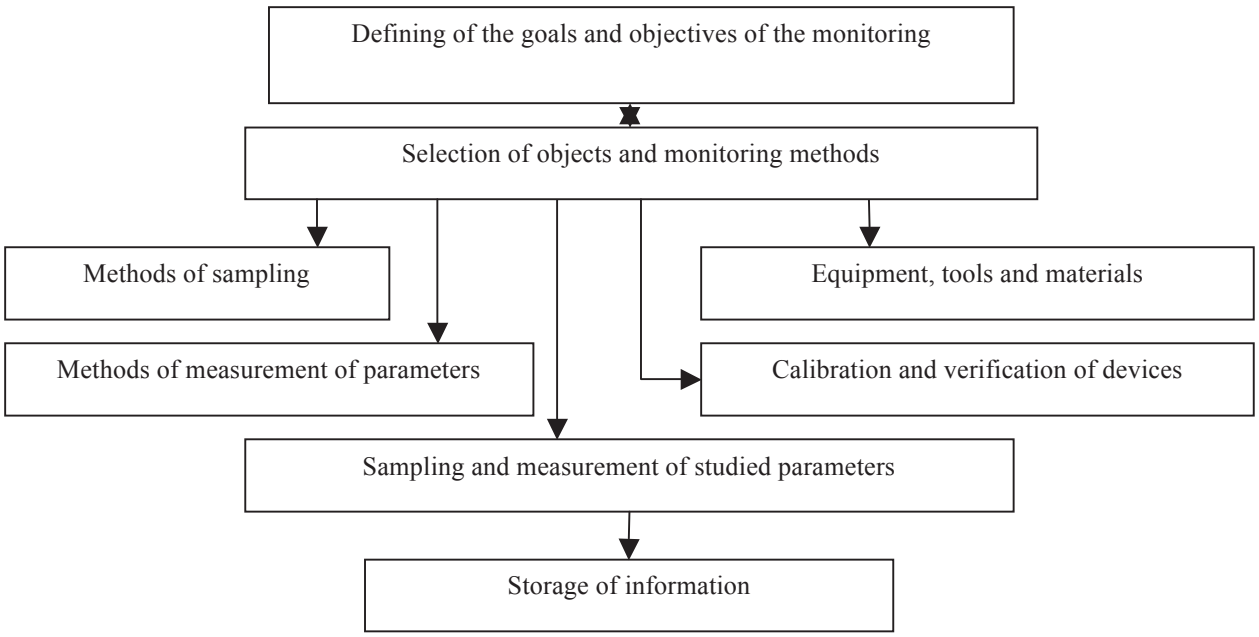


Figure 3.1.1. The general scheme of environmental monitoring.

We have developed a preliminary list of tasks to be performed by the monitoring:

- obtaining of full, detailed, and statistically reliable information about radioactive contamination levels at forest territories;
- the determination of radioactive contamination dynamics at the forest territories;
- the study of radionuclide redistribution in different soil types and landscapes;
- the study of the intensity of radioactive elements migration into wood species, bushes, grass, mushrooms, moss, lichen, and in the system 'soil-forest biota component' (plant species, mushrooms, animals);
- the study of radionuclide migration in the system 'soil – fodder plant – wild animals';
- the study of radionuclide migration along food chains from forest to human beings.

Surveillance network. According to the nature of obtained information, the network is split into two levels – basic and special. The basic level includes all territory under surveillance. It is a set of sampling points located in the nodes of a square 8-km increment grid (Fig. 3.1.2).

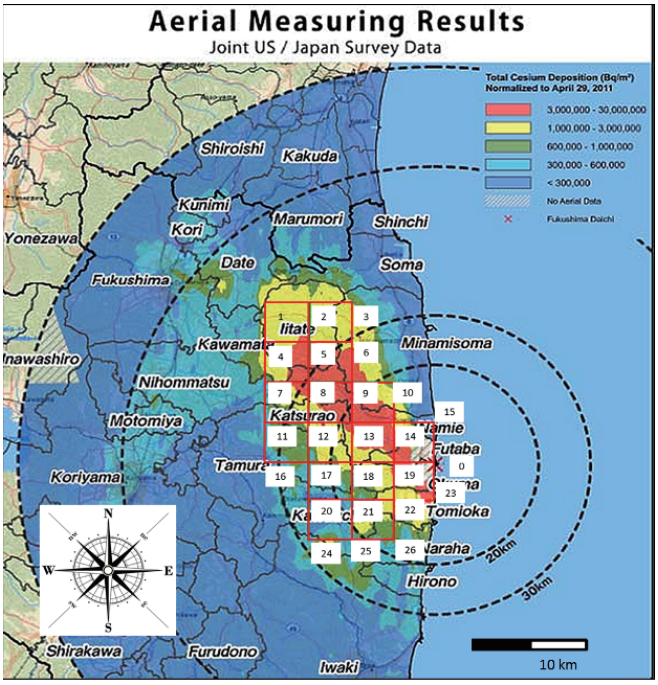


Figure 3.1.2. Basic observation network for the radioecological monitoring of forests

Monitoring performance duration and probe sampling schedule. Monitoring performance duration depends on many factors: radioecological, managerial, and social. As the upper limit, we recommend to consider the period of time needed for practically full radiation decontamination of a territory from anthropogenic radionuclides, which is ten half-decay periods of a radionuclide. It is about 300 years for ^{137}Cs. The lower limit might depend on decisions of managerial authorities, which, taking into account approved local norms, may consider the radiation situation at a given territory safe and suspend or change the performance of the radiation-ecological monitoring. Existent experience of huge radiation accidents (ChNPP in 1986 and PU "Mayak" in 1957) shows that the radiation-ecological monitoring lasts for several dozens of years: 8 and 57 year correspondingly. In any case, the radiation-ecological monitoring of forests in the region of Fukushima-1 NPP is a long-term project, which would possibly last for several dozens of years.

The choice of monitoring objects is done depending on an ecosystem structure. For the observation, the main ecosystem elements are chosen, which could be divided into constitutive parts, if necessary. In a forest ecosystem, we emphasize three basic elements, which become the monitoring objects of the forest ecosystems: soil (layer-by-layer), vegetation, and mushrooms.

Exposure dose rate measurement. In addition to sample collection, it is recommended to measure the **exposure dose rate** at each point. It is a universal express indicator for radiation situation. The measurements are performed

with certified equipment at each point of probe sampling in two positions: at the soil surface and at 1 m height from the surface.

Soil. The experience of practical radioecology shows that soil is the main depot for radionuclides within landscape. We consider soil here as a biological system, which includes the soil itself and the forest waste litter.

Several soil layers are studied during the radioecological monitoring: the ground litter and the upper soil layers till 20 cm deep. If necessary, (the study of radionuclide vertical migration in time), soil samples are collected till 150 cm depth.

Ground litter. In this layer, the processes of organic substances' mineralization and their return into soil occur. It is a key element in locking the biological cycling of substances. It consists of three layers:

1. Litter fall (A01);
2. Semidecomposed litter fall layer (A0f);
3. Decomposed litter fall layer (A0h) [31].

The layers A01 and A0f+A0h are collected separately. Litter fall samples are cut out with a knife or separated by blocks using a metal frame of 25 by 25 cm size. The samples must not contain mineralize soil particles.

Soil itself. Probe sampling is performed employing a cylindrical sampler (Figure 4) till the 20 cm depth in order to estimate the density of the soil surface contamination.

Soil layer separation into layers 0-1 cm, 1-5 cm and so on with the step of 5 cm could possibly be done on-site. In this case, sealed packaging is necessary for each layer with a detailed description and a probe sampling coordinate.

Soil is collected from 5 points employing the envelope technique (4 samples at the corners and one in the center) within a square of 25 m^2 area.

Samples from all points are mixed.

A pit-hole with the dimensions of 1.5 m x 0.8 m x 0.5 m is used for the estimation of radionuclides' vertical migration in soil. Probe collection is performed from a cleaned pit-hole wall with a 5-cm diameter sampler from each 5-cm thick soil layer.

The vegetation of a forest ecosystem is represented by several blocks: the forest stand, the herbaceous-shrub layer, grass, and moss.

The forest stand. Forest forming tree species possess the second place after soil as a radionuclide depot within a landscape. From one side, trees provide the accumulation and the multi-year storage of substances, and, from another side, - the main substance flow from soil into the surface part of the landscape.

Probes of the following forest stand components are sampled: wood, surface and internal bark, leaves, and needles. The wood is collected from a tree trunk using a bore at the 1.5 m height.

The surface and internal bark also are collected at the 1.5 m height. All components are collected from 3-5 trees of the dominating species to compose a mixed sample.

The herbaceous-shrub layer. Because of a short life cycle and low lignin content in tissues, herbaceous plants provide the fast return of mineral substances into soil. Shrubs also provide the every-year return of mineral substances during leaf fall. Branch and leaves samples are collected from 3-5 shrubs of a dominating species.

Moss cover is an important factor for the litter fall transformation: it provides a necessary humidity and pH regime. Moss samples are collected onto a tray in blocks of 25 cm by 25 cm size. The samples must not contain soil particles, litter fall, leaves, and needles.

Herbaceous plants are collected from the area of 1 m^2. A geobotanical frame is employed for this purpose. The surface part only is collected.

Mushrooms as ^{137}Cs contamination indicators for soil ecosystems.

After the accident at ChNPP, mushrooms are the most contaminated biological components of forest ecosystems. Several biological factors influence on ^{13}Cs specific activity levels. Mushrooms nutrition type is one of the main factors (belonging to a certain ecological group). The localization depth of the mycelium main part is the second important factor. This factor plays an important role for mushrooms, which belongs to the symbiotroph ecological group (symbiotic relations with higher plants.

Nutrition type (ecological group). During the first two-three years after radionuclide precipitation, the high intensity of ^{137}Cs accumulation by mushrooms-saprotrophs and xylotrophs is detected. Maximum ^{137}Cs content values are characteristic for these species during the period.

^{137}Cs content increase in mushrooms-symbiotrophs occurs considerably slower. For these mushrooms, the maximum specific activity values of this radionuclide could be registered in 8-12 years after the radiation contamination of an ecosystem.

Mycelium localization depth. For the mushrooms belonging to the symbiotroph ecological group, the main factor influencing the ^{137}Cs accumulated amount is the localization depth of the main part of mycelium, which is characteristic for each species (the quantity of available ^{137}Cs in soil at the localization depth of mycelium of each species).

During the whole period (1986-2014) of the research of forest ecosystem contamination after the accident at Chernobyl NPP, the ^{137}Cs content in mushrooms, which mycelium was localized in the 0-5 cm layer, was considerably higher (sometimes more than 1000 times) than in mushrooms, which mycelium was deeper than 5 cm.

In mushrooms with not deep mycelium location (0-5 cm), the differences between separate species could be quite large, however, they do not exceed one order of magnitude. All mushrooms of this group accumulate cesium in greater quantities that mushrooms with the deep mycelium localization.

The increase of ^{137}Cs concentration in mushrooms with the deep mycelium location could witness about the elevate availability of this radionuclide for other biota species in soil at the depth more than 5 cm and/or about the transfer of the radionuclide main depot at the depth greater than 5 cm.

We recommend the employment of mushrooms as the species-indicators during the performance of monitoring work within forest ecosystems, taking into account the parameters of ^{137}Cs accumulation dynamics for various ecological group mushrooms and the different localization depth of the main part of mycelium in soil.

To perform monitoring work, it is necessary to collect mushrooms fruit bodies keeping in mind the parameters, which influence the accumulation of radionucides. As it has been stated above, the main factors are the belonging to an ecological group and the mycelium localization depth in soil.

The sample collection of higher mushrooms-mushrooms is performed according to techniques, which were developed at the Institute for Nuclear Research NAS or Ukraine after the accident at ChNPP and using commonly known procedures employed for that work.

Maromycetes belonging to the following ecological groups are employed for the research:

1. Symbiotrophs (mycorrhiza-forming);
2. Soil saprotrophs;
3. Litter fall saprotrophs;
4. Xylotrophs-parasites;
5. Xylotrophs-saprotrophs.

During monitoring sampling, it is desirable to collect mushrooms belonging to the symbiotroph ecological group with different localization of the main part of the mycelium in soil, which is characteristic for each species:

- Litter fall layer;
- Soil layer 0 – 5 cm;
- Deeper than 5 cm.

Radioecological monitoring results in some initial data. On the basis of the data, higher level information formats are developed: generalizations, information-analytical materials, prognoses, etc. The whole mechanism for information storage organization is developed mainly around the initial data.

A system of distributed databases employing modern computer infrastructure should be a technical basis for the collection, storage, treatment, and output of the information. This system functioning should be supported by unified software solutions.

The results of forest radioecological monitoring are stored and transformed into the following information objects:

- Data base for monitoring objects' sample measurements;
- Monitoring results data base;
- Verified statistical information;
- Observation rows;
- Information-analytical materials (analytical notes, articles, monographs, etc.)

All these objects are equally important for data saving and information treatment. Data bases for measurement results and monitoring results considerably duplicate each other, but this situation increases the information storage security. Such categorization and information flows distribution considerably simplify the access to data and other information materials. It is important because providing information for decision making is the main task of the monitoring.

3.1 Research of the expedience of remediation of forest' soils by means of fruit bodies of mushrooms

Within the framework of the STCU project # 5954 «Monitoring of radioactive pollution of forest ecosystems after accident on ChNPP», «Research of the expedience of remediation of forest' soils by means of fruit bodies of mushrooms», we have analyzed literature data, experience of forest ecosystems remediation on the example of "Red forest" at the territory of the 30-km zone of ChNPP. We also performed experiments at the territory of Ukraine and Japan.

Mechanical elimination of woody plants and upper soil layer followed by the material "burying" in 1.5-2.0 m depth trenches being at the level of subsoil water was performed at the territory of "Red forest" after the accident at ChNPP. It contributes to prolonged radionuclide contamination of not only the subsoil water, but new vegetation growing at the burial locations. Thus, the "burying" of the "Red forest" solved only instant problems of 1986-1987 years, but promoted the appearance of consequential radioecological problems.

Later, artificial **phytostabilization** (artificial tree planting) as well as natural phytostabilization (natural seeding) played a role in the remediation of the "Red forest" territory. However, a considerable part of the "Red forest" ecosystem has not returned to its initial state because of the **mechanical elimination** of its main components, but transformed into another forest type.

As far as we know, remediation of large territories of forest ecosystems after radionuclide contamination has not been ever performed anywhere in the world. We have considered possibilities for the remediation using **phytoextraction** and **phytostabilization** of ^{137}Cs in forest ecosystems of Japan.

Main approaches for the choice of such methods were conditioned by the principles of minimization of radionuclide contamination influence on personnel and inhabitants. Also, the minimization of additional influence on the developed ecosystem and decrease of economical costs.

Research at testing areas with different radionuclide contamination levels were performed for these purposes at the territory of the 30-km zone of ChNPP in 2013-2015. There were three stations at the testing area "Kopachi" and one station at each of the easting areas "Paryshe', "Leliv", and "Dytiatky". Testing area "Staiky" was developed 150 km from ChNPP as the reference territory. Main components of forest ecosystems were studied.

Together with Japanese scientists, we performed in parallel analogous study at the territory of Japan at the testing areas "Ohgawara", "Namie", and "Iitake" located within the influence zone of Fukushima-1 NPP. These testing areas are similar to the testing areas at the territory of ChNPP 30-km zone according to their character and levels of radionuclide contamination.

Characteristics of the chosen testing areas in Ukraine and Japan are similar in contamination levels and in ^{137}Cs distribution within vertical soil layers. Maximum ^{137}Cs specific activity at these studied testing areas is registered, as a rule, in the ground litter ($A_0l + A_0f + A_0h$), and minimum - within soil layers deeper than 5 cm. It allows sufficiently correct comparison among the testing areas.

Firstly, we are considering a potential possibility for ^{137}Cs phytoextraction fro forest ecosystems using fruit bodies of wild mushrooms-macromycetes (**mycoextraction**). It has been dictated by the fact that the fruit bodies of the fungi-macromycetes are the maximum accumulators of ^{137}Cs in forest ecosystems.

Mycoextraction. Relative simplicity of mushroom collection and minimum interference into the forest ecosystem are the advantages of this technique of forest ecosystems remediation.

Fungi-cosmopolite species widely spread in the world including Ukraine and Japan were chosen for the study. Mainly, we studied representatives of the symbiotroph ecological group.

Mycoextraction results are shortly presented in the tables 3.2.1 and 3.2.2.

Table 3.2.1. The results of mycoextraction performed at stations of the testing area "Kopachi" during the season October 15 – November 15, 2013 (Ukraine)

Station #	**Station area (m^2)**	**^{137}Cs Total content in the station soil (Bq)**	**^{137}Cs extracted from the station soil by mushrooms during the season (Bq)**	**^{137}Cs extracted from the station soil by mushrooms during the season (% of total content)**
1	100	68962000	1536440	2,228
2	100	16778000	34248	0,204
3	100	201583000	313934	0,156

Table 3.2.2. The results of mycoextraction performed at stations of the testing area "Ohgawara" (Japan) during single sampling on September 28, 2014.

Station #	**Station area (m^2)**	**^{137}Cs Total content in the station soil 28.09. 2014 (Bq)**	**^{137}Cs extracted from the station soil by mushrooms during single collection on 28.09.2014 (Bq)**	**^{137}Cs extracted from the station soil by mushrooms during single collection on 28.09.2014 (% of total content)**
1	1,92	2371920	581	0,025
2	1,05	1473150	28824	0,196
3	2,70	6197850	15547	0,251

Similar results for the mycoextraction were obtained at the testing area "Namie".

It should be pointed that the results for the "Ohgawara" testing area were obtained during a single collection. The efficiency of ^{137}Cs mycoextraction would be higher t regular mushroom collection. In addition, mushroom yield during productive years, especially at mushroom-rich plots, could exceed the mean during a season by dozens or hundreds times, allowing, therefore, a successful remediation.

Thus, considerable amount of ^{137}Cs is eliminated from a forest ecosystem (up to several % a year of the total content in soil) during the mycoextraction with no damage to the system.

Phytoextracton with the use of trees. The forest at the station #1 of the "Leliv" testing area has been developed mainly by artificial planting of Pinus sylvestris (L.), performed in 1946-1955. We estimated ^{137}Cs content at the station in 2014 at the value of 26,250,590 Bq/100 m^2.

Theoretical calculations of ^{137}Cs phytoextraction efficiency through potential total elimination of surface part of the main forest-forming species (Pinus sylvestris (L.)) obtained during the study at the "Leliv" testing area are presented in the Table 3.2.3.

Table 3.2.3. The results of potential total phytoextraction at the station #1 of the "Leliv" testing area employing Pinus sylvestris (L.) in 2014.

Organ or tissue	**Weight, kg/100 m^2**	**^{137}Cs (Bq/kg)**	**Potential ^{137}Cs phytoextraction from soil (Bq)**	**Potential ^{137}Cs phytoextraction from soil (%) of total content in soil for 28 years**
Needles	80	160 ± 17	12800	0,049
Branches	140	115 ± 23	16100	0,061
Wood	1010	335 ± 26	338350	1,289
Bark	90	2269 ± 194	204210	0,778

The potential total phytoextraction of ^{137}Cs at the testing area "Leliv" using Pinus sylvestris (L.) from the area of 100 m^2 28 years after the accident at ChNPP was 571,460 Bq or 2.955 % of the total content. It equals approximately 0.1 % per year.

However, ^{137}Cs specific activity in fungi fruit bodies at the testing area "Leliv" is considerably higher in 2014 and varies from 1830 to up to 62,300 Bq/kg, exceeding the radionuclide specific activity in wood by 5-300 times and sometimes even more. Mainly, this exceeding is from 30 to 250 times for different fungi species. Such ^{137}Cs specific activity exceeding in mushrooms fruit bodies is consistently characteristic for all studied testing areas within 30-km ChNPP zone starting 1989-1990 until now (2015).

3.2 Creation of model of migration of ^{137}Cs in the forest ecosystems polluted as a result of accident on the Chernobyl atomic power plant and the atomic power plant Fukushima-1

The model is a dynamic compartment model considering various processes of radionuclide migration in the system "soil – mushrooms". The main ***objects*** of the modeled system are the following: soil and mushrooms, which characteristics are a soil type, sol contamination density σ, time t of radionuclide presence in soil, and mushroom species. The following ***processes*** determine the value of ^{137}Cs intake by the mushrooms from soil:
1. Radionuclide radioactive decay.
2. Soil-based:
ground litter decomposition;
fuel particles destruction caused by soil factors;
vertical migration of ^{137}Cs ions along the soil profile caused by diffusion and convective transfer of Cs^+ ions by soil moisture;
processes of ion "sorption – desorption".
3. Biological: surface sorption by mushroom mycelium, metabolism, etc.
Temporal limits of the model use are determined from the moment of radioactive precipitates $t_0 = 0$ *ad infinitum* $t_{max} = \infty$.
Sphere of use allows the employment of the model parameters for: a set of experimentally studied mushrooms; various soil types with the use of values for sorption – desorption parameters, diffusion, and convective transfer for them.

The model was developed on the basis of the data on contamination of mushrooms and soils on testing area Dytiatky. The testing area Dytiatky is located on the southern trace of the 30-km zone of radionuclide emission after the accident at Chornobyl NPP. Density of soil contamination on May 10, 1986 was 185-370 kBq•m^2. The testing area is represented by sod-weakly-podzol clay-sandy soil type with acidic soil solution pH_{KCl} 4.9, low humus content around 1%, and cation exchange capacity of 3.4 mg-eq. per 100 grams of soil. In total, data for 11 mushrooms obtained during a long period after the accident at ChNPP were used, which allows building a model for radiocesium transfer from soil into the mushrooms for the whole period after the radiation accident.

Basing on the insight, a conceptual scheme for a model on radiocesium accumulation by mushrooms has been proposed (Figure 3.3.1). Basic principles for the radionuclide behavior model in the system "soil – mushrooms" have been formulated:

^{137}Cs gets into a mushrooms immediately from the soil solution, and its concentration in mushroom is proportional to the concentration of the radionuclide water-soluble WS form in soil, which dynamics is determined by the processes of the radionuclide release from fuel particles, its redistribution among sorption sites, and by vertical migration of the ion along the soil profile. The coefficient of the radionuclide transfer from soil into mushrooms TF, which value was determined for the moment of their fruit bodies maturing, was used as a quantitative

characteristic of mushroom species peculiarities. The coefficient is the final integral estimation of all metabolism processes.

The accumulation of radiocesium by various mushroom species is dependent on the concentration of the radionuclide water-soluble forms in soil layer containing the mycelium of a specific mushroom species. Thus, mycelium of the studies mushrooms is localized in the following soil layers: *Xerocomus badius*, *Paxillus involutus*, *Lucoperdon perlatum*, *Macrolepiota procera*, *Armillariella melea* – in the ground litter $A_0l+A_0f+A_0h$, *Suillus luteus*, *Russula xerampelina*, *Lactariusturpis* – in the soil layer A (0 – 5 cm), *Boletus edulis*, *Leccinum scabrum*, *Tricholoma flavovirens* – in the soilllayer B (5 – 10 cm).

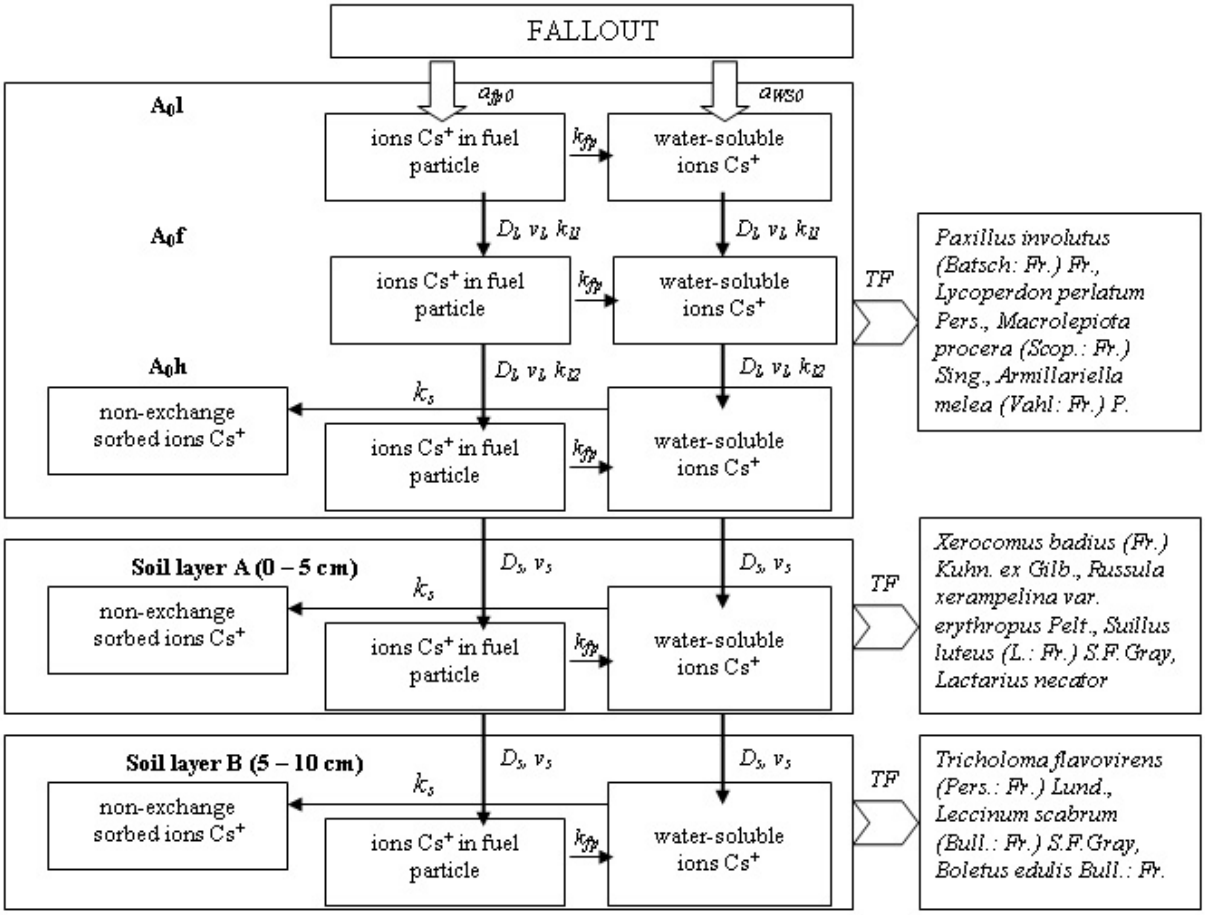


Figure 3.3.1. Conceptual scheme for the model of ^{137}Cs migration in the system "soil – mushroom"

Taking into account the conceptual scheme and the main principles of the model, radionuclide balance in an elementary soil layer has been examined and balance simultaneous equations have been developed:

$$Q_{WSi}^{m} = Q_{WSi}^{m-1} + k_{fp} \cdot Q_{fpi}^{m-1} + k_{l1} \cdot Q_{WSi-1}^{m-1} - k_{l2} \cdot Q_{WSi-1}^{m-1} - k_s \cdot Q_{WSi}^{m-1} - \lambda \cdot Q_{WSi}^{m-1} + (D_{l(s)} \cdot \frac{Q_{WSi-1}^{m-1} - Q_{WSi}^{m-1}}{\Delta x^2} +$$

$$+ v_{l(s)} \cdot \frac{Q_{WSi-1}^{m-1} + Q_{WSi}^{m-1}}{2 \cdot x}) \cdot \Delta t - (D_{l(s)} \cdot \frac{Q_{WSi}^{m-1} - Q_{WSi+1}^{m-1}}{\Delta x^2} + v_{l(s)} \cdot \frac{Q_{WSi}^{m-1} + Q_{WSi+1}^{m-1}}{2 \cdot x}) \cdot \Delta t$$

Then, the concentration of ^{137}Cs in mushrooms, mycelium which is different soil layers will be equal:

Litter $$SA_l = TF_0 \cdot \sum_{i=0}^{n} Q_{wsi} \quad (3.16)$$

Soil layer 0 – 5 см $$SA_{0-5} = TF_0 \cdot \sum_{i=n}^{m} Q_{wsi} \quad (3.17)$$

Soil layer 5 – 10 см $$SA_{5-10} = TF_0 \cdot \sum_{i=m}^{d} Q_{wsi} \quad (3.18)$$

where TF_0 - values extrapolated to zero time t = 0 ^{137}Cs transfer coefficients from soil to mushrooms $(Bq \cdot kg^{-1})/(kBq \cdot m^{-2})$.

It performed the initial condition that the amount of radionuclide the initial time:

in fuel particles $$Q_{fp0} = a_{fp0} \cdot \sigma \quad (3.14)$$

in water – soluble form $$Q_{ws0} = (1 - a_{fp0}) \cdot \sigma \quad (3.15)$$

The value of the model parameters were tridiagonal matrix algorithm using data obtained by other researchers in the study of individual processes involved in radionuclide migration in the system "soil - mushrooms" and presented in Table. 3.3.1, 3.3.2.

Table 3.3.1. Parameters of ^{137}Cs migration in the "soil - mushrooms" system models

Parameters			Unit of measurement	Value
Density of soil contamination		σ	$kBq \cdot m^{-2}$	185
Rate decomposition	A_0l	k_{l1}	y^{-1}	0,6
	A_0f+A_0h	k_{l2}		0,4
Depth of the litter layer		n	cm	3
Depth of soil layer 0-5 cm		m		8
Depth of soil layer 5 – 10 cm		d		13
The fate of cesium in the composition of fuel particles		a_{fp0}		0,80
Rate destruction of fuel particles	$A_0l+ A_0f+A_0h$	k_{fpl}	y^{-1}	0,06
	soil	k_{fps}		0,16
Rate of convection soil moisture	$A_0l+ A_0f+A_0h$	v_l	$cm \cdot y^{-1}$	0,46
	soil	v_s		0,86
Diffusion coefficient	$A_0l+ A_0f+A_0h$	D_l	$cm^2 \cdot y^{-1}$	0,34
	soil	D_s		0,62
Rate of non-exchange radionuclide sorption		k_s	y^{-1}	0,28
Decay rate		λ	y^{-1}	0,022

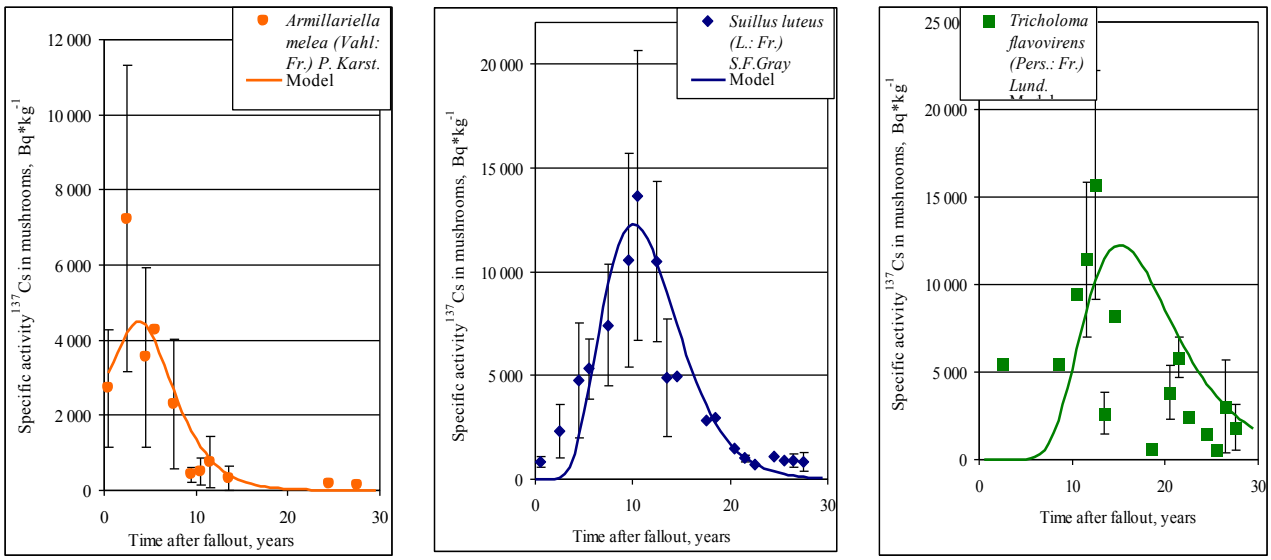
Table 3.3.2. Value extrapolated to the ^{137}Cs fallout time transfer from soil to mushrooms TF_0, $(Bq \cdot kg^{-1})/(kBq \cdot m^{-2})$

Mushrooms	TF_0
Saprotroph ($A_0l+A_0f+A_0h$)	
Paxillus involutus (Batsch: Fr.) Fr.	1 150
Macrolepiota procera (Scop.: Fr.) Sing.	125
Lycoperdon perlatum Pers.	115
Armillariella melea (Vahl: Fr.) P. Karst.	65
Symbiotroph (0 – 5 cm)	
Xerocomus badius (Fr.) Kuhn. ex Gilb.	1 120
Russula xerampelina var. erythropus Pelt.	750
Suillus luteus (L.: Fr.) S.F.Gray	610
Lactarius necator	470
Symbiotroph (5 – 10 cm)	
Tricholoma flavovirens (Pers.: Fr.) Lund.	2 650
Boletus edulis Bull.: Fr.	1 830
Leccinum scabrum (Bull.: Fr.) S.F.Gray	1 650

In the Fig. 3.3.2 shows dynamics of ^{137}Cs specific activity after the accident and the model curve describes them in saprotroph and symbiotroph.

The employment of all abovementioned criteria allows concluding that the developed model of ^{137}Cs migration in the element “soil – mushrooms” at the testing area Dytiatky is adequate in the whole, and its determined parameters are statistically significant with sufficient significance levels for 8 mushrooms: saprotroph and symbiotroph, mycelium which is located in soil layer 0 - 5 cm. It should be noted that the model must be implemented as a software code that will allow to conduct numerical calculations with smaller steps in the depth of the soil and time and specify the parameters of the model. Thus, the diagnostic check of the model has proved

the model conformance to the real process that is its theoretical basis and the conceptual scheme are correct, and the model parameters could be used for prognosis of mushrooms contamination with radionuclides.



a *b* *c*

Fig. 3.3.2. The dynamics of ^{137}Cs specific activity after the accident and the model curve describes them in: *a* - saprotroph, micelles which are in forest litter (A_0l + A_0f + A_0h), *b* - symbiotroph, which mycelium in the soil at a depth of 0 - 5 cm, *c* - symbiotroph, which mycelium in the soil at a depth of 5 - 10 cm.

The formal statistics of forecast quality check have shown that the predicted and actual rows correlate with each other, and the model accuracy for ^{137}Cs migration from soil into mushrooms is within 100% limit for 60% of cases.

There is no formal test for the determination of turning points; therefore, they could only be defined while visually checking the forecast and actual rows. The check has allowed determining inflexion points for the model of ^{137}Cs migration in the element "soil – mushrooms". The presence of the points could be explained by the radionuclide vertical migration along the soil profile and by the presence of various sorption sites in the soil, which are characterized by different velocities od the cesium ion "sorption-desorption" process. Because of uncertainty in the velocity values, the turning point on the prediction curve corresponds to the moment of equilibrium settlement between the velocities of vertical soluble forms migration processes in the soil profile and non-exchange sorption of radionuclide SAC. The turning points occur at different moments for different soil layers! Thus, a compromise between the accuracy and dynamics mushroom contamination forecast while using a model of ^{137}Cs migration in the element "soil – mushrooms" need be found to get acceptable accuracy in average along the whole trend.

The sensitivity of the model for cesium and strontium radionuclide migration from soil into mushrooms to starting data could be eliminated through correct assignment of mushroom species to a soil layer, where its mycelium is localized, and through the analysis and consideration of other factors immediately influencing on the forecast results. The estimation of sensitivity of the model for cesium migration from soil into mushrooms to the change of starting data and parameters has confirmed that the model is of high quality as a whole.

The analysis of experimental data and predictive Leliv and Paryshev polygons leads to the conclusion that the reliability calculation ^{137}Cs concentration in mushrooms on the developed model is between 100% and can be considered satisfactory in predicting the radiation environment.

Due to the fact that the result of calculation mushrooms ^{137}Cs contamination of the model is sensitive to input data, have developed the basic requirements. To predict the contamination mushrooms must have the following information:

1. The density of contamination of soil ^{137}Cs, soil depth from which samples were taken to assess the density of soil contamination.
2. Form fallout ^{137}Cs: condensation, fuel, their part.
3. Type of soil, advisable agrochemical soil properties by which we can determine the kinetic parameters of radionuclides in the soil. Soil type specified in the national classification and according to the soil classification of the Food and Agriculture Union of Nations (FAO UNESCO).

4. Forest category (type) with indication tree species composition of litter (coniferous, deciduous), the thickness and the weight of forest litter on square 1 m^2.
5. Mushrooms type with indication soil layer in which it is placed mycelium, mushrooms state at the time of collection (old, dry, etc.)
6. Time sampling mushrooms, with indicating the exact date.
7. Methods of sampling soil and mushrooms. Be sure to indicate the degree of soil samples and conjugated mushrooms. It is desirable to specify the number of fruiting bodies used for measurement.
8. Geographic coordinates of sampling that enables the use of the presence of digital maps of soil types or soil contamination density.

An important issue is the accuracy and credibility of experimental data. It is advisable to sample "mushrooms - soil - year" contain at least 3 values radionuclide content in fruit bodies.

4 Conclusions

4.1. As forest ecosystems occupy 71 % of the area of prefecture Fukushima, they are critical ecosystems in providing radiation safety at the contaminated territories. Therefore, managing of forest ecosystems is a priority one at the territories contaminated with radionuclides.

During execution of the project, we have developed a plan for monitoring of forest ecosystems adapted to the natural conditions of contaminated territories after the accident at the Fukushima-1. In proposed scheme, at least 26 nodal points should be selected for monitoring reliability. Location of the nodal points should be identified depending from the landscape and can be adjusted directly on the sampling site. These points will serve for detailed sampling: soil (layers-to-layers), plants, mushrooms, etc. This approach allows obtaining sufficient information about dynamics of radionuclides spreading and gives data to build into models to predict year-by year behavior of radiocesium in the forests. Based on primary results, the number of sampling sites can be corrected based on pollution level of the territory.

Due to cooperation with Japanese colleagues from the Fukushima University, Institute of Environmental Radioactivity, monitoring on 4 sampling sites (out of 26 optimal) at exclusion zone of NPP Fukushima-1 was carried out in 2014 and 2015, which obviously is not enough. However, these few data were used by us to prove the possibility of soil remediation on some polygons, as well as to provide verification of the developed models taking into account local conditions.
We consider it is very important to continue monitoring of forest ecosystems in the exclusion zone of Fukushima-1 NPP in close cooperation with our Japanese colleagues.

4.2. We assume that total deforestation is less effective comparing to the mycoextraction because trees extract less ^{137}Cs than mushrooms per year under current conditions of chronic radionuclide contamination at forest ecosystems of Japan.

The advantages of **phytoextraction** (total deforestation):

The velocity of radionuclide elimination from the contaminated territory;
High level of mechanization of the clearance process;
Potential capability for technical use of the wood: piles, construction material for unattended or rarely attended premises, constructions and buildings (mooring berths, storage facilities, hangars, etc.)

Phytoextraction disadvantages:

Total destruction of forest ecosystem structure;
Destruction of forest buffer function in radionuclide retention;
The absence of phytostabilizing function of woody plants accelerates the vertical and horizontal migration of radionuclides, contributes to ecosystem erosion and degradation;
Huge volume and weight of radioactive material, especially at exceeded maximum permissible levels, multiply the complexity of the material processing, radionuclide concentration, fixation and/or burying of the obtained material. It results in additional irradiation of personnel and in work cost increase;
High costs of artificial remediation of the ecosystem, which will be regenerating to initial level during many years.

Advantages of **mycoextraction** (harvesting of fungi fruit bodies):

Minimum influence on the forest ecosystem. Practically, no real interference into the ecosystem;
High specific activity of the fungi fruit bodies allows extracting considerable amount of ^{137}Cs from contaminated territories;
During rich years, 0.5 -2 % and more of the total ^{137}Cs content in soil could be extracted using the fungi fruit bodies at contaminated territories;
The collection of objects with high ^{137}Cs concentration and relatively low weight assumes lower transportation and preliminary storage costs;
Drying of the fungi fruit bodies reduces their weight and volume approximately by 10 times making certain storage and following burying methods easier.

Disadvantages of **mycoextraction** (harvesting of fungi fruit bodies):

The yield of wild fungi depends on many factors and varies from year to year at different areas and territories. Therefore, the development of accurate forecasts for annual parameters of ^{137}Cs mycoextraction efficiency at certain areas is complicated;
Relatively high activity of fungi fruit bodies preconditions special requirements for personnel and safety of technological schemes of subsequent processing and storage (utilization) of the obtained material.

4.3. Distinctive feature of the developed model is the housing account мицелия various kinds of mushrooms in soil: in a debris layer, a layer of earth of 0-5 cm both 5-10 cm. Frame and properties of a forest litter have been considered, as it is impure dynamic mediuim and sharply differs from layers of earth below 0 cm on a vertical.

For the first time, the conceptual scheme of model including all migratory soil processes and features of mushrooms has been developed. The assessment of sensitivity of model to change of the initial data and parameters has confirmed that as a whole this model is qualitative.

It is spent model verification on the experimental data received on other polygons in the remote season after failure on ChNPP. Model verification has shown satisfactory accuracy of forecasting of the maintenance of radiocesium in fruit bodies of mushrooms on other test polygons, including polygon «Namie» in terrain of Japan.

The model can be used on polluted after accident on the Fukushima-1 NPP forest ecosystems of Japan.

For correct adaptation of model to conditions of Japan it is necessary to use the concrete data for each research polygon:

type of a forest;
predominant species of plants and mushrooms,
thickness and compound of a litter,
type and properties of soil.

The listed characteristics influence values of parameters of migration ^{137Cs} on a soil profile and its sorption a soil-absorbing complex.

5 Applicability and Recommendations for Japan

5.1. During performance of the project we had been developed system of monitoring of forest ecosystems of Japan polluted as a result of the accident on the Fukushima-1 NPP. The monitoring system has been developed taking into account geographical and environmental conditions of this region of Japan. In the developed system of monitoring we have considered a role of mushrooms in accumulation and redistribution of radioactive nuclides in forest ecosystems.

5.2. During the project realization, we have used our multi-year experience of radioecological research of forest ecosystems within ChNPP influence zone. Open literature data and information on the radioecological state of forest ecosystems of Japan, obtained in collaboration with Japanese colleagues during the project performance, have been analyzed. On this basis, we recommend an approach to the remediation of forest ecosystems of Japan at the Fukushima-1 NPP influence zone.

Main principles of our approach to the forest ecosystem remediation declare the minimization of artificial (anthropogenic) influence on the developed ecosystem aiming the required effect of sustainment, regeneration (curing) of the ecosystem. Natural processes of self-rehabilitation start working, when there is no need in artificial external influence, and if the ecosystem was not in a critical state.

We believe that the fundamental principle is noninterference or minimum influence on an ecosystem, except the situations when an ecosystem state poses threat to human health and life or to the existence of the ecosystem.

Phytoextraction could be used in a case of dangerous or critical situation for humans, followed by planting of new trees and by regeneration of the vegetation cover. However, these are long-term and costly programs, which could be ineffective.

Mycoextraction, namely the collection of fungi fruit bodies followed by their processing and utilization, applies minimum negative influence onto the forest ecosystem. At sufficient mushroom yield, the mycoextraction of radionuclides, as well as many other pollutants, is more effective and less harmful for the forest ecosystem comparing to the phytoextraction.

In forest ecosystems, **natural phytostabilization** regulates horizontal and vertical flows of mineral substances (including radionuclide compounds), the formation of soil and of the forest ecosystem itself. In addition, self-rehabilitation functions start working in forests at inconsiderable natural and anthropogenic interferences.

Artificial phytostabilization, including planting of aboriginal woody and shrubby plant species, is possible at negative but non-critical parameters of influence on forest ecosystems of Japan.

This might result in the solution of non-critical radioecological problems with minimum influence on the environment.

5.3. During the execution of the project manager STCU number 5954 NE Zarubina together with Japanese colleagues Radiological station in the village near Okuma conducted soil sampling and mushrooms in Japan in September 2014 (9.28.2014). Measurement of 137Cs specific activity in selected samples conducted by Japanese colleagues. The main focus is on the training ground Ohgawara, posted 7 km east of NPP Fukushima-Daichi 1.

On the Ohgawara polygon 3 stations were studied forest ecosystem with total area 495 m^2. Compact stations are not more than 30 m apart. In experimental stations dominated by deciduous forest on brown forest soils.

According to Japanese colleagues in some areas selected polygon layer of the forest floor ($A_0l+A_0f+A_0h$) some time after the accident was removed. In September 2014 this layer began to recover naturally. At the time of sampling on site polygon dominated by the environmental group symbiotrophs. Parallel studies conducted on polygon Namie, which environmental conditions were similar to the polygon Ohgawara.

It estimated the possibility of model verification of ^{137}Cs contamination mushrooms using the data obtained in the above ranges. In the Table. 5.1 presents offered in Japan for the model parameters of ^{137}Cs migration in the system "soil – mushrooms".

Table 5.1. Parameters of ^{137}Cs migration in the system "soil - mushrooms" models for Japan

Parameters			Unit of measurement	Value	
				Ohgawara, Litate	Namie
Density of soil contamination		σ	$kBq \cdot m^{-2}$	1 235 – 2 295	13 910
Rate decomposition	A_0l	y^{-1}	$year^{-1}$	0,6	0,6
	A_0f+A_0h			0,4	0,4
Depth of the litter layer		n	cm	3	6
Depth of soil layer 0-5 cm		m		8	11
Depth of soil layer 5 – 10 cm		d		13	16
Rate of convection soil moisture	$A_0l+ A_0f+A_0h$	v_l	$cm \cdot y^{-1}$	0.46	0,86
	soil	v_s		0.54	0,94
Diffusion coefficient	$A_0l+ A_0f+A_0h$	D_l	$cm^2 \cdot y^{-1}$	0.26	0,62
	soil	D_s		0.30	0,48
Rate of non-exchange radionuclide sorption		k_s	y^{-1}	0,31	0,31
Decay rate		λ	y^{-1}	0,022	0,022

Thus, with literature provides the agrochemical properties of brown forest soils and, consequently, their absorption properties are similar to grey forest soils, common in Ukraine. Therefore, we used non-exchange rate value of radionuclide sorption for grey forest soils. Table settings values no fuel particles speed degradation since it is known that Fukushima-1 nuclear power plant accident fallout were condensing form. Table 5.2 presents experimental data of ^{137}Cs specific activity in mushrooms and calculated values using the developed model.

Table 5.2. The ^{137}Cs specific activity in mushrooms, taken on Ohgawara and Namie testing sites 28.09.2014 and forecast data using models.

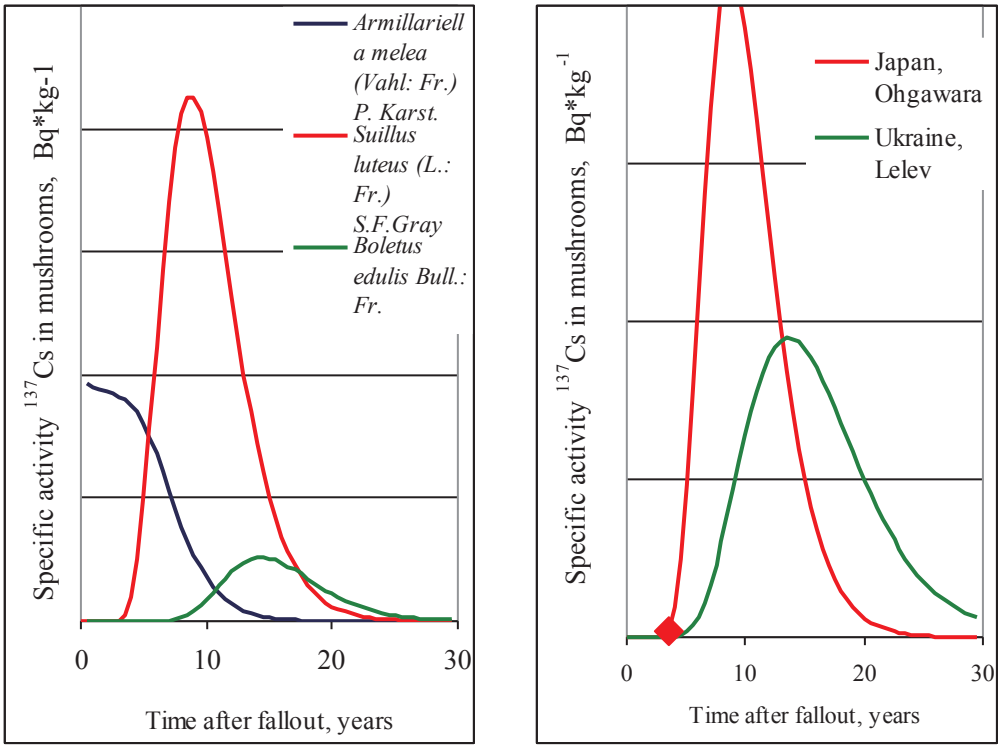
№ stantion	Mushrooms species	^{137}Cs specific activity in mushrooms, $Bq \cdot kg^{-1}$		Relative deviation, ± %
		experimental data	forecast data	
Sampling site «Ohgawara»				
Symbiotroph (0 – 5 cm)				
1	*Lactarius subvellereus, Lactarius rufus*	1 970 – 49 293	1 190	65
2, 3	*Russula emetica*	1 634 – 52 974	1 900	16
Symbiotroph (5 – 10 cm)				
2, 3	*Sarcodon aspratus*	12 592 - 15 031		
2	*Ramaria spp.*	12 459		
Sampling site «Namie»				
Symbiotroph (0 – 5 cm)				
	Suillus luteus	49 111	17 530	280
Symbiotroph (5 – 10 cm)				
	Sarcodon scabrocus, Sarcodon aspratus	22 535 - 35 265		
	Ramaria spp.	5 425		

The table shows that the ^{137}Cs concentration in the same form mushrooms differs from 4 to 32 times in even at Ohgawara polygon. For example, the cesium specific activity for *Russula emetica* is from 1 634 to 52 974 $Bq \cdot kg^{-1}$ in even the same polygon station №1. Such large differences could be explained in the territories in which the radionuclide is composed of the fuel components. Then would be made local emissions of ^{137}Cs specific activity. Presented in Table differences due, most likely, indistinct experimental polygon features that affect migration patterns. In particular, it is necessary to clearly define whether the litter layer was removed and when. If so, what was the initial density of soil contamination.

The same conclusions can be made by comparing data between two different Ohgawara and Namie polygons. When the difference in the density of soil contamination up to 10 times the ^{137}Cs concentration in mushrooms - almost the same and as for *Ramaria spp.* - greater density of 2 times less.

All this confirms once again the need to formulate clear requirements to input data for verification model of radionuclide migration in “soil - mushrooms” system in other territories.

Forecast in Table. 5.2 obtained under the condition that litter does not have deleted. Then, in Fig. 5.1*a* presents forecast of mushrooms contamination of differ environmental groups on the above conditions at the site Ohgawara.



a *b*

Fig. 5.1. Forecasted of ^{137}Cs concentration in mushrooms: *a* - at the site Ohgawara, Japan, *b* - Ukraine and Japan polygons with same soil contamination

The lowest concentration of radionuclides characteristic of symbiotroph, mycelium which is 5-10 cm soil layer.

Figure shows that the ^{137}Cs concentration in:

- saprotroph gradually decreases with time after the fallout;
- symbiotroph, mycelium which is in soil layer 0 - 5 cm - increases ranging from 3-4 years, most occur in the 9-10 year
- symbiotroph, mycelium which is in soil layer 5 - 10 cm - increasing from 8-9 years to a maximum of 15-16 years.

Fig. 5.1*b* presented for comparison curves predicted of ^{137}Cs concentration in *Russula xerampelina var. erythropus Pelt.* for Ukraine and Japan polygons are the same level of soil radionuclide contamination after the accidents at Chernobyl and Fukushima-1 NPP. The figure shows that the ^{137}Cs concentration in symbiotroph to 2 times less at the site Leliv in Ukraine than at the site Ohgawara in Japan. This can be explained by the fact that the radionuclide at the site Leliv in Ukraine was a part of the fuel particles that also migrate through the soil profile.

Special Session

Significance of Studies for Long Term Monitoring and Remediation In and Around Fukushima

Transfer of Radionuclides in Soil-plant System

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Transfer of Radionuclides in Soil-plant System

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The soil-to-plant transfer of radionuclides is an important process for estimating the internal radiation dose through food ingestion including agricultural plants and livestock feeds. Transferability of radionuclides from soil to plant depends on several factors such as plant species, soil types, fertilizer applications, weather conditions, and so on. The soil-to-plant transfer factor used for model application is defined as the concentration of radionuclides in the plant divided with that in the soil. It has very wide ranges. For example, radiocesium in sandy and peat soils is very mobile and is easily taken in by plants. Therefore, the transfer factor of radiocesium in these soil types generally shows higher values compared to that in mineral soils where radiocesium is not as mobile. Radiocesium interception potential (RIP) represents the selective adsorption capacity of Cs, which shows a negative correlation with the transfer factor.

The radionuclides transferred into the plant body are translocated among the plant components and the radionuclides are distributed to each part of the plant body. Rice is a staple food in Asian countries and ingestion of polished rice is the most important pathway of radionuclides into humans. The non-edible rice plant components are returned to the soil as fertilizer and are used to as an ingredient of livestock feed. The concentration of ^{137}Cs in rice plant components varied by one order of magnitude with the lowest and highest values in in polished rice and bice bran, respectively. The lowest concentration of ^{90}Sr in rice plant is in polished rice, which is similar to that of ^{137}Cs, whereas the highest concentration of ^{90}Sr is found in the straw, which is two orders of magnitude higher than that in polished rice. Consequently it is important to clarify the transfer of radionuclides in the soil-plant system for a better understanding of the fate of radionuclides in the terrestrial environment. The presence of radionuclides in the agricultural environment of Fukushima is also included in this report.

Time Changes in radiocesium wash-off from various landuses and transport through river networks after the Fukushima Daiichi NPP accident

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Time Changes in radiocesium wash-off from various landuses and transport through river networks after the Fukushima Daiichi NPP accident

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Due to Fukushima Daiichi Nuclear Power Plant accident, radioactive materials including ^{134}Cs and ^{137}Cs were widely distributed in surrounded area. The radiocesiums have been transported in river networks. The monitoring started at 6 sites from June 2011. Subsequently, additional 24 monitoring sites were installed between October 2012 and January 2013. Flow and turbidity (for calculation of suspended sediment concentration) were measured at each site, while suspended sediments and river water were collected every one or half month to measure ^{134}Cs and ^{137}Cs activity concentrations by gamma spectrometry.

Also detailed field monitoring has been condcuted in Yamakiya-district, Kawamata town, Fukushima prefecture. These monitoring includes, 1) Radiocesium wash-off from the runoff-erosion plot under different land use, 2) Measurement of radiocesium transfer in forest environment, in association with hydrological pathways such as throughfall and overlandflow on hillslope, 3) Monitoring on radiocesium concentration in soil water, ground water, and spring water, 4)Monitoring of dissolved and particulate radiocesium concentration in river water, and stream water from the forested catchment, and 5)Measurement of radiocesium content in drain water and suspended sediment from paddy field.

Our monitoring result demonstrated that the ^{137}Cs activity concentration in eroded sediment from the runoff-erosion plot has been almost constant for the past 3 years, however the ^{137}Cs activity concentration of suspended sediment from the forested catchment showed slight decrease through time. On the other hand, the suspended sediment from paddy field and those in river water from large catchments exhibited rapid decrease in ^{137}Cs activity concentration with time. The decreasing trend of ^{137}Cs activity concentration were fitted by the two-component exponential model, differences in decreasing rate of the model were compared and discussed among various land uses and catchment scales. Such analysis can provide important insights into the future prediction of the radiocesium wash-off from catchments from different land uses. The decreasing trend of river system varied with catchments. Our analysis suggests that these differences can be explained by upstream landuse with different decreasing trend.