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Proceedings of the Final ISTC/STCU Technical Review Committee Meeting of Fukushima Initiative "On the Environmental Assessment for Long Term Monitoring and Remediation in and around Fukushima" November 5-6, 2015, Tokyo, Japan

(Eds.) Secretariat of the Technical Review Committee Meeting of ISTC/STCU Fukushima Initiative Fukushima Initiative Technical Review Committee

> Environment and Radiation Sciences Division Nuclear Science and Engineering Center Sector of Nuclear Science Research

June 2016

日本原子力研究開発機構

Japan Atomic Energy Agency

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(Eds.) Secretariat of the Technical Review Committee Meeting of ISTC/STCU Fukushima Initiative, Fukushima Initiative Technical Review Committee

> Environment and Radiation Sciences Division Nuclear Science and Engineering Center Sector of Nuclear Science Research Japan Atomic Energy Agency Tokai-mura, Naka-gun, Ibaraki-ken

> > (Received April 15, 2016)

The final ISTC/STCU Technical Review Committee Meeting "On the environmental assessment for long term monitoring and remediation in and around Fukushima" was held in Hitotsubashi University Hall, Tokyo, on November 5 - 6, 2015. This report consists of summary reports of Fukushima Initiative projects, which were presented at the meeting. As regarding to the sections those which will be published elsewhere, only their executive summaries are included.

Keywords: Fukushima Nuclear Power Station Accident, Radioactive Waste, Agriculture, Livestock, Remediation, Phytoremediation, Monitoring, Polymer, ISTC, STCU, CIS, Fukushima Initiative

JAEA-Conf 2016-002

「福島および周辺環境における長期モニタリングと環境回復に関する特別研究(福島イニシア チブ)」についての国際科学技術センターおよびウクライナ科学技術センター技術評価委員会

最終会合報告集

2015年11月5~6日、東京都

日本原子力研究開発機構 原子力科学研究部門 原子力基礎工学研究センター 環境・放射線科学ディビジョン

(編) ISTC/STCU 福島イニシアチブ技術評価委員会会合事務局、 ISTC/STCU 福島イニシアチブ技術評価委員会

(2016年4月15日受理)

平成 27 年 11 月 5 日~6 日の 2 日間にわたり「福島および周辺環境における長期モニタリングと環 境回復に関する特別研究」についての国際科学技術センター(ISTC)およびウクライナ科学技術セン ター(STCU)合同の福島イニシアチブ技術評価委員会最終会合を東京都内において開催した。本論 文集にはそこで行われた報告のうちから、別途論文として投稿されるものを除いて収録している。

Contents

1. Prefac	e1
1.1 (Overview of the ISTC/STCU Fukushima Initiative 'on the environmental assessment for long
term	monitoring and remediation in and around Fukushima'1
1.2 0	Contributing parties
1.3 7	Fechnical Review Committee members
1.4 8	Secretariat of the Technical Review Committee Meeting of ISTC/STCU Fukushima Initiative4
2. Agend	la of the final technical review committee meeting of the Fukushima Initiative5
3. Procee	dings of the Fukushima Initiative Projects11
3.1 0	Organic waste volume reduction with incineration - Compaction of radioactive waste produced by
deco	ntamination of territories polluted due to the accident at Fukushima Daiichi Nuclear Power
Stati	on: STCU Project 595211
	Volodymyr Tokarevskyy, Institute of Chernobyl Problems (Ukraine)
3.2 A	Advanced Polymeric Systems Providing Deactivation of Different Surfaces and Soil from
Radi	oactive Pollutions-Inorganic Waste Volume Reduction with Polymer: ISTC Project A-207130
	Zoya Farmazyan, CJSC "Yerevan SRI "Plastpolymer" (Armenia)
3.3 F	Remediation of Cs-contaminated soils through regulation ¹³⁴ Cs and ¹³⁷ Cs soil-plant-transfer by
poly	meric sorbents: ISTC Project A-2072
	Anna Tadevosyan, Institute of Hydroponics Problems (Armenia)
3.4 I	Development of a set of measures for production of assured quality agricultural goods under
radio	pactive contamination conditions - Experiments of Cesium Uptake by Livestock and Crops: ISTC
Proje	ect K-2085
	Andrey Panitskiy, Institute of Radiation Safety and Ecology, National Nuclear Center of Kazakhstan (Kazakhstan)
3.5 N	Methodology for long-term radiation monitoring to dose assessment using radiological zoning and
mod	eling of radionuclides migration in environmental and food chains -Countermeasures of Cesium
Upta	ke by Farm Crops and Livestock: STCU Project 595355
	Boris PRISTER, Mykola TALERKO, Evgenii GARGER, Viktorija VINOGRADSKAJA, Tatjana
	LEV, Institute for Safety Problems of Nuclear Power Plants of National Academy of Sciences of
	Ukraine (Ukraine)
3.6 N	Monitoring of radioactive pollution of forest ecosystems after accident on Chernobyl NPP –
Reha	abilitation with mushrooms harvesting in forest ecosystem: STCU Project 5954
	Nataliia Zarubina, Institute for Nuclear Research of National Academy of Sciences of Ukraine
(Ukr	raine)

目 次

1.	緒言1
	1.1 ISTC/STCU 共同の特別課題 「福島とその周辺環境の長期モニタリングと回復のための
	環境影響評価に関する活動」の概要1
	1.2 寄与機関
	1.3 技術評価委員会構成
	14 最終会合事務局構成 4
2.	議事次第
3.	福島イニシアチブプロジェクト成果報告11
	3.1 福島第一原子力発電所事故により汚染した地域の除染で生ずる廃棄物の減容研究有機
	廃棄物の燃焼減容: STCU 595211
	Volodymyr Tokarevskyy, チェルノブイリ問題研究所 (ウクライナ)
	3.2 新規高分子材料を用いた、種々の表面材質ならびに土壌に対する除染研究高分子材料
	による無機廃棄物の減容: ISTC-A-2071 –
	Zoya Farmazyan, CJSC 「エレバン SRI プラストポリマー」 (アルメニア)
	3.3 高分子吸着剤による ¹³⁴ Cs および ¹³⁷ Cs の土壌-植物移行の抑制と Cs 汚染土壌の浄化:
	ISTC-A-2072
	Anna Tadevosyan, Institute of Hydroponics Problems (アルメニア)
	3.4 放射性汚染環境において基準を満たした農畜産物を生産するための対策の開発-家畜・
	作物への放射性セシウムの移行試験: ISTC-K-208550
	Andrey Panitskiy, Institute of Radiation Safety and Ecology, National Nuclear Center of
	Kazakhstan (カザフスタン)
	3.5 環境および食物連鎖における放射性核種の移行に係る区分化とモデリングによる長期
	放射線モニタリングの方法論農畜産物への放射性セシウムの移行低減対策:STCU-5953
	Boris Prister, Mykola Talerko, Evgenii Garger, Viktorija Vinogradskaja, Tatjana Lev, ウクライ
	ナ科学アカデミー原子力発電所安全問題研究所 (ウクライナ)
	3.6 チェルノブイリ原発事故後における森林生態系の放射性汚染のモニタリング-森林生態
	系でキノコによる収奪を利用する除染: STCU 5954
	Nataliia Zarubina, ウクライナ科学アカデミー原子力研究所(ウクライナ)

1. Preface

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

Secretariat of the Technical Review Committee Meeting of the Fukushima Initiative

1.1.1 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 Station. 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.

1.1.2 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.

1.1.3 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. Eleven 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

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
 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 Fukushima area: Agricultural Radiation Research Center, Fukushima Branch and Tohoku Agriculture Research Center, National Agriculture and Food Research Organization (NARO); National Forest at Iitate 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.

1.2 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 States Department of Energy (US DOE)

European Commission Directorate-General for International Cooperation and Development (DEVCO)

Ministry of Foreign Affairs of Japan (MOFA)

Technical Contributors

Pacific Northwest National Laboratory (PNNL)

Brookhaven National Laboratory (BNL)

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

Among many others

1.3 Technical Review Committee members

Dr. Sergey Fesenko (International Atomic Energy Agency) Dr. Yasuo Onishi (Pacific Northwest National Laboratory) Prof. Dr. Yuichi Onda (Tsukuba University, Japan) Prof. Dr. Shuji Tsuruoka (Shinshu University, Japan) Prof. Dr. Mitsuru Akashi (Osaka University, Japan) Dr. Shinichi Nakayama (Japan Atomic Energy Agency, Japan) Dr. Keiichi Kawase (Japan Atomic Energy Agency, Japan)

1.4 Secretariat of the Technical Review Committee Meeting of ISTC/STCU Fukushima Initiative

Dr. Takeshi Matsunaga (International Science and Technology Center, Japan Atomic Energy Agency)
Mr. Alan Taylor (International Science and Technology Center)
Dr. Iryna Tomashevska (Science and Technology Center in Ukraine)
Ms. Olga Martin (Los Alamos National Laboratory)
Ms. Ayimgul Kakenova (International Science and Technology Center)
Ms. Anel Kasseinova (International Science and Technology Center)
Ms. Zauresh Dikhanbayeva (International Science and Technology Center)

2. Agenda of the final technical review committee meeting of the Fukushima Initiative

ISTC/STCU Joint Review of Projects 'On the environmental assessment for long term monitoring and remediation in and around Fukushima'

Hitotsubashi University Hall (Tokyo, Japan), November 5-6, 2015

Day 1 - November 5, 2015

PART I: Final Technical Expert Project Review

Opening & Introductions

09:00 - 09:20	Opening Remarks:
	Mr. Yasuo Kishimoto, MEXT
	Mr. Andy Hood, U.S. DOE
	Mr. Curtis (BJ) Bjelajac, STCU
	Mr. David Cleave, ISTC
	Introductions:
	Dr. Matsunaga, ISTC

Session 1: Waste Interim Storage and Waste Volume Reduction

Chair: Dr. Araki, JAEA

09:20 - 9:40	Organic Waste Volume Reduction with Incineration Compaction of radioactive waste produced by decontamination of territories polluted due to the accident at Fukushima Daiichi Nuclear Power Station: STCU-5952 Dr. Volodymyr Tokarevskyy
09:40 - 10:20	Technical Discussion on Project STCU 5952
10:20 - 10:40	Coffee Break
10:40 - 11:00	Inorganic Waste Volume Reduction with Polymer Advanced polymer systems providing deactivation of different surfaces and soil from radioactive pollutions: ISTC-A-2071 Dr. Zova Farmazvan, Dr. Konstantin Pyuskyulyan
11:00 - 11:40	Technical Discussion on Project ISTC A-2071

Session 2: Recovery of Agriculture

Chair: Dr. Onishi, Pacific Northwest National Laboratory, U.S.A.

JAEA-Conf 2016-002

11:40 - 12:00	Reduction of Cesium Transfer from Soil to Plant with Polymer <i>Remediation of Cs-contaminated soils through</i> <i>regulation</i> ¹³⁴ Cs and ¹³⁷ Cs soil-plant-transfer by polymeric sorbents:ISTC-A-2072 Dr. Anna Tadevosyan, Dr. Nina Tavakalyan
12:00 - 12:40	Technical Discussion on Project ISTC A-2072
12:40 - 13:40	Lunch
13:40 - 14:00	Experiments of Cesium Uptake by Livestock and Crops Development of a set of measures for production of assured quality agricultural goods under radioactive contamination conditions: ISTC-K-2085 Dr. Andrey Panitskiy, Dr. Timur Kozhahanov
14:00 - 14:40	Technical Discussion on Project ISTC K-2085
14:40 - 15:00	Coffee Break
15:00 - 15:20	Countermeasures of Cesium Uptake by Farm Crops and Livestock Methodology for long-term radiation monitoring to dose assessment using radiological zoning and modeling of radionuclides migration in environmental and food chains: STCU-5953 Dr. Mykola Talerko
15:20 - 16:00	Technical Discussion on Project STCU-5953

Session 3: Forest Management and Ecosystem

Chair: Dr. Onda, Tsukuba University, Japan

16:00 - 16:20	Forest Ecosystem – Mushrooms Monitoring of radioactive pollution of forest ecosystems after accident on Chernobyl NPP: STCU-5954 Dr. Nataliia Zarubina
16:20 - 17:00	Technical Discussion on Project STCU 5954
Wrap-Up	
17:00 - 17:30	Summaries from Panel Chairs Day 2 - November 6, 2015

Significance of Studies for Long Term Monitoring and Remediation In and Around Fukushima

Chair: Dr. Kawase, JAEA

9:00 - 9:20	Studies of Soil to Plan Transfer of Radionuclides – The importance of the Japanese Environment <i>Dr. Tsukada, Fukushima University</i>
9:20 - 9:30	Q&A
9:30 - 9:50	Importance and Remaining Issues in Studies of Secondary Migration of Radionuclides and Waste Dr. Onda, Tsukuba University
9:50 - 10:00	Q&A
10:00 - 10:20	Coffee Break

PART II: Fukushima Initiative Closing

Project Results

Chair: Dr. Nakayama, JAEA

10:20 - 10:30	Organic Waste Volume Reduction (STCU-5952) Dr. Volodymyr Tokareyskyy
10:30 - 10:40	Inorganic Waste Volume Reduction (ISTC-A-2071)
10:40 - 10:50	Reduction of Cesium Transfer From Soil to Plant With Polymer (ISTC-A-2072)
	Dr. Anna Tadevosyan
10:50 - 11:00	Experiments of Cesium Uptake by Livestock and Crops (ISTC-K-2085)
	Dr. Andrey Panitskiy
11:00 - 11:10	Countermeasures of Cesium Uptake by Farm Crops and Livestock (STCU-5953)
	Dr. Mykola Talerko
11:10 - 11:20	Forest Ecology - Mushroom (STCU-5954)
	Dr. Nataliia Zarubina

Next Steps for Cooperation

Chair: Ambassador Lehman, ISTC, Kazakhstan

11:20 - 12:10	Applicability of Project Results for Application In and Around Fukushima and Next Steps for Promoting Project Results <i>Comments from Technical Experts</i>
12:10 - 12:30	Lessons Learned for Future Cooperative Initiatives Example of Joint Research Outcome (Dr. Onda,, Dr. Zarubina) Facilitated Discussion
12:30 - 12:45	Opportunities for Future Collaboration and Targeted Initiatives through ISTC and STCU Facilitated Discussion

Closing Remarks

12:45 - 13:15	Closing Remarks & Observations:
	Dr. Yasuo Kishimoto, MEXT
	Mr. Andy Hood, DOE
	Dr. Masanori Araki, JAEA (SAC, Chair)
	Mr. Noritsugu Takahashi, MOFA
	Ambassador Ron Lehman, ISTC Chair

Follow-up Meetings

Afternoon open for ISTC/STCU Project Participants arrange meetings with interested Japanese experts

List of Participants

1. Technical Review Committee

Dr. Sergey Fesenko, International Atomic Energy Agency
Dr. Yasuo Onishi, Yasuo Onishi Consulting (Ex. DOE-PNNL) (U.S.A.)
Dr. Shinichi Nakayama, Nuclear Safety Research Center, JAEA (Japan)
Dr. Keiichi Kawase, Fukushima Environmental Safety Center, JAEA (Japan)
Prof. Dr. Yuichi Onda, Tsukuba University (Japan)
Prof. Dr. Shuji Tsuruoka, Shinshu University (Japan)
Prof. Dr. Mitsuru Akashi, Osaka University (Japan)

2. National Representatives

DOE, US

Ms. Regina Carter, Senior Policy Advisor, Office of Nuclear Controls, Office of Nonproliferation and International Security **Mr. Andy Hood**, Senior Advisor to DOE

Ms. Olga Martin, Los Alamos National Laboratory

Mr. Koichi Uchida, Energy Specialist, US DOE, Resident in Tokyo, Japan

EU

Dr. Leonidas Karapiperis, Minister-Consular, Head of Science & Technology Section, Delegation of the European Union to Japan

ISTC GB, STCU Board

Mr. Ronald F. Lehman, Chair, ISTC GB; Executive Committee of STCU Board

ISTC

Mr. David Cleave, Executive Director of ISTC.

STCU

Mr. Curtis (BJ) Bjelajac, STCU Executive Director

SAC, ISTC

Dr. Masanori Araki, JAEA, Chair, ISTC Science Advisory Committee

MEXT, Japan

Mr. Yasuo Kishimoto, Director General
Mr. Takashi Imura, Deputy Director
Ms. Ayaka Imamura, Secretary, International Science and Technology Affairs Division, Science and Technology Policy Bureau

MOFA, Japan

Absent

3. Extra-committee Experts

Prof. Dr. Hirofumi Tsukada (Fukushima University) (Invited Lecturer)
Prof. Takuya Okamoto (Shinshu University) (Ex. ISTC)
Dr. Makoto Nakatani (by a substitute), Ministry of Agriculture, Forestry and Fishery
Dr. Tomoko Nakanishi, University of Tokyo

Project Representatives

STCU- 5952: Dr. Volodymyr Tokarevskyy, Institute of Chernobyl Problems (Ukraine)

ISTC- A-2071: Dr. Zoya Farmazyan, CJSC "Yerevan SRI Plastpolymer" (Project Manager, Armenia) Dr. Konstantin Pyuskyulyan CJSC "Yerevan SRI Plastpolymer" (Researcher, Armenia)

ISTC- A-2072: Dr. Anna Tadevosyan, Inst. Hydroponics Problems (Project Manger, Armenia) Dr. Nina Tavakalyan, Inst. Hydroponics Problems (Researcher, Armenia)

ISTC- K-2085: Dr. Andrey Panitskiy, National Nuclear Center of Kazakhstan (Project Manager, Kazakhstan) Dr. Timur Kozhahanov, National Nuclear Center of Kazakhstan (Researcher, Kazakhstan)

STCU-5953: Dr. Mykola Talerko, Institute for Safety Problems of Nuclear Power Plants (Ukraine)

STCU-5954: Dr. Nataliia Zarubina, Institute of Nuclear Research (Ukraine)

ISTC/SCTU Secretariat

ISTC Dr. Takeshi Matsunaga, ISTC Program Manager

STCU Dr. Iryna Tomashevska, STCU Program Manager 3. Proceedings of the Fukushima Initiative Projects

3.1 Organic waste volume reduction with incineration– Compaction of radioactive waste produced by decontamination of territories polluted due to the accident at Fukushima Daiichi Nuclear Power Station

STCU Project 5952

Tokarevskyy V.V. Institute for Chernobyl Problems

Project Information

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:	TOKAREVSKYY, 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
Participating Institutes:	Name: EKSIS Ltd, Ukraine Director: Rovensky A. I. Address: 61040, 43, Skorostnaia str, Kharkiv, Ukraine Phone: +380577556901 Email: guandr@mail.ru
Foreign Collaborators:	Dr. Yasuo Onishi (Pacific Northwest National Laboratory, U.S.A.)

1. 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 the 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.

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).

JAEA-Conf 2016-002



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



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: $8 \sim 100 \text{ kBq/kg}$ 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, e.g. 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 (¹³⁷Cs, ⁹⁰Sr,) throughout the volume of soil in the temporary location of radioactive waste (> 700,000 m³), 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 ¹³⁴Cs and ¹³⁷Cs. Technological scheme for decontamination of soil comprises 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 pH 1.

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 2.

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 pH 3 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 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, Fig. 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.

JAEA-Conf 2016-002



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

3.3 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)

3.4 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 $100 \sim 150$ 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.

3.5 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 ¹³⁷Cs and direct combustion in the installation of municipal type EKSYS Ltd. In recent years attention-grabbing alternative composite techniques that extend to Japan.

3.5.1 Technology of decrease volume and weight

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.

3.5.2 Volume reduction technology

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 ¹³⁷Cs 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 \in .

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).

3.5.3 Separation technology

Technology of separation of ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs and impurities in the soil, accelerating, rate of removal of ¹³⁷Cs in soil is 80% ~99.9% (at 1000 ~ 1300°C). In Fig. 6 is a diagram of thermal separation technology ¹³⁷Cs from soil.



Fig. 6. Scheme of thermal separation technology ¹³⁷Cs 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.

3.6 Justification of combined wet technology

Indicator "reduce volume" (K₁), which was defined as the ratio of the total soil (M₁) to the amount of solid radioactive waste arising from decontamination (M₂): $K_1 = M_1/M_2.$ (1)

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.

(2)

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.



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.

JAEA-Conf 2016-002





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 ¹³⁴Cs, ¹³⁷Cs between particle size fractions of soil.
- Results of calculations of the equilibrium distribution of ¹³⁴Cs, ¹³⁷Cs 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 ¹³⁴Cs, ¹³⁷Cs 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), the combustion will occur in the shaft furnace in a dense layer under the upward flow of air-oxidant, thus forming following zones: drying, pyrolysis, carbon gasification, combustion, ash cooling, etc.

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. The design capacity of one reactor is 500 kg/h, and the block of 4 reactors is 2000 kg/h. Compaction ratio of organics is 90 ~ 100 times. Combustion technology is performed with simultaneous destruction of organic and inorganic materials; adsorption and coagulation, and immobilization of Cs on the aluminosilicates the in the process of interaction with clays, zeolites, 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 ¹³⁷Cs 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 (Table 1).

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	$95 \sim 98\%$ (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	to 99.99% according to the standard requirements
particles and aerosols quality of emissions	
The concentration of 137 Cs in the air torch output (D)	$< 8.10^{-1} \text{ Bq/m}^3.$
Costs:	
capital (in 2004 prices).	20 million €.
block of 4 reactors	(1 main reactor 10 million €, 3 auxiliary 10 million
	€).
Exploitation	1 million € per year.
Cost of processing 1 ton	166€

Table 1 Contaminated wood of Chernobyl Exclusion Zone

5.2 Step 1 – 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 ¹³³Cs, which was introduced into the charge in an amount that simulates a specific activity of 8 kBq/kg. This shows the following results (Table 2):

Table 2 Experience	processing of	contaminated w	with toxic materials	charge of	f municipal waste.
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Compaction ratio of the charge	90 ~ 95 times		
The transition rate ¹³³ Cs in ash	to 80%		
6 stepped gas cleaning with thermo-catalytic	to 99.99%		
combustion and hermetic ash capture provides			
cleaning of solid particles and aerosols			
Quality emissions	according to the requirements		
The experiments also showed technology provides	to 1×10^{-1} Bq/m ³ at a rate of D $8 \cdot 10^{-1}$ Bq/m ³ .		
clean the aerosols ¹³³ Cs			

The technology is licensed. The installation is made commercially by "Energostal", Kharkov, Ukraine. Costs (in prices 2006):

Capital	199 million UAH (8.8 million USD)
Operational	12.87 million UAH (7.3 million USD);
Cost of processing of 1 ton	288 UAH (12.7 USD).

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 ¹³⁴Cs and ¹³⁷Cs. 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 ¹³⁴Cs and ¹³⁷Cs. 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 ¹³⁴Cs and ¹³⁷Cs, 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.

3.2 Advanced Polymeric Systems Providing Deactivation of Different Surfaces and Soil from Radioactive Pollutions –Inorganic Waste Volume Reduction with Polymer

ISTC Project A-2071

Zoya Farmazyan CJSC "Yerevan SRI "Plastpolymer"
Project Information

Title of the Project:	Advanced Polymeric Systems Providing Deactivation of Different Surfaces and Soil from Radioactive Pollutions	
Commencement Date:	July 1, 2013	
Duration:	2 years	
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1. Brief description of work: Objectives, and expected results

1.1 Project goals and objectives

At the beginning of the project, two main problems were investigated:

(1) Development of polymer systems for the 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 the top soil layers on fields.
- In examinations of polymers solutions for the decontamination of radionuclides ¹³⁷Cs and ¹³⁴Cs from soils;
- For the study of the effect of combining polymers with microscopic soil fungi (MSF) on radionuclide fixation.

1.2 Expected results and their application

1.2.1 Original setting up

Originally, the project had following expected results.

(1) Deactivating strippable films

The development of new polymer systems implies the possibility of their use for the decontamination of porous surfaces as strippable films instead of the commonly used high-pressure water means.

(2) Soil decontamination

Several variants of the 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 a narrow layer of soil before the soil is removed from the fields.
- To use polymeric systems for the decontamination of the removed soil wastes.

1.2.2 Results obtained before April 2014

The main results of the works performed on deactivating strippable films before April 2014 are summarized below.

- New polymeric systems for use as strippable films for the decontamination of ¹³⁷Cs and ¹³⁴Cs from porous surfaces were developed
- The basic technological process of the new polymers was designed. The parameters of the technological process ensure the quality of the films with the desired operational properties, such as adhesion, strippability, plasticity-elasticity, and depth of penetration into the porous surface.
- The properties of the films were studied on 4 porous surfaces, differing in chemical composition and porosity. For the tested samples that penetrate into pores, the plasticity/elasticity of the films formed after drying allows the film to be pulled from the surface layers. The percentage of removal of 137Cs from the porous surface was found to reach up to 35%.
- The developed technological process and properties of the products are competitive with one of the known deactivating compositions- the water dispersion ALARA-1146 of Carboline production, which we used as an analogue.

2. Modifications of work after the April 2014 meeting

On April 13-18, 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 the long-term monitoring and remediation in and around Fukushima, Fukushima and Tokyo, Japan.

As a result, in view of the current situation in Fukushima, the Japanese side noted that cleanup of hard surfaces with polymers is of no interest at this time, whereas the mechanism of trapping and moving of ¹³⁷Cs in soil is of great interest and importance. Therefore, the work with hard surfaces was stopped, and all further efforts were focused on investigations of the behavior of polymer solutions in soils. Nevertheless, by the end of the third quarter, the principal variant of polymer synthesis was found to provide removal of polymer films from porous surfaces. In addition, a part of the works on soils was performed. The main tasks after April were determined to be as follows:

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

(2) Possibility of using the results of these studies within the common problem to reduce the amount of radioactive soil wastes, which were removed from the fields and stored in bags.

To perform the new tasks, the methodology of the experiments was modified according to the previously obtained results. In addition, the number of objects under study was increased.

3. Technical approach, methods, experiments, and theory

3.1 Technical works structure

The stages of work performed after April 2014 can be described with the following flowchart:



3.2 Soil decontamination studies

3.2.1 Materials

(1) Soil

The first soil sample (sANPP) was sampled at the area around the Armenian nuclear power plant (ANPP, Ararat plain) and corresponds to the type of a soil around Fukushima (Gray Lowland soils). According to the XRD analysis, the clay fraction consists mainly of montmorillonite (MMT; $Al_{1.67}Mg_{0.33})Si_4O_{10}(OH)_2Na_{0.33}$. The soil composition data are provided in Table 1.

In addition to sANPP (enriched with MMT), two other soil types containing muscovite and illite were sampled from different locations around the Tsaghkunyats Mountains: Meghradzor (sMdz) and Tsaghkadzor (sTh).

According to the XRD analysis, these soils contain the following:

Mica-montmorrilonite $-((Al_9Ti_9Fe)Al_{0.68}Si_{9.32})O_{10}(SOH_2)(K_8Na)_{0.43}X_{0.17}$

Illite- (K9Na9Ca)1.36(Al,Mg)4.17 (Si,Al)8(O,SOH)24

Muscovite - (K₉Na) (Al₉Mg₉Fe)₂ (Si 2.1 Al 0.9)O10(SOH)₂

Illite- (K9Na9Ca)203.33 (Mg9 Mn) O.4.3 (Al9Fe 9 Ti)2 O2 16 (Si9Al) O2 4H2O

Soils sMdz and sTh are highly similar in mineralogical composition but differ in mechanical composition (Table 1).

Soil sampling	Content of particles of different size (mm) in soil suspension (%)					
location	(Kachinski method)					
	1-0.25	0.25-0.05	0.05-0.01	0.01-0.005	0.005-	< 0.001
					0.001	< 1 µm
					5-1 µm	
Ararat plane	4.2	21.84	23.1	12.0	33.58	5.28
(sANPP soil)						
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

Table 1. Mechanical properties of soils

The mechanism of Cs binding by polymeric solutions was refined on pure clay minerals (illite and vermiculite). The reasons for the choice of these minerals are as follows: 1-these minerals primarily bind ¹³⁷Cs and ¹³⁴Cs in soils; 2-Fukushima soils (clay fractions) primarily contain these minerals; and 3-Japanese specialists plan to sieve the contaminated soils to separate the highly contaminated silt and clay fractions.

Vermiculite (Vm) was purchased as the raw material with large particles of 1-2 cm; the samples were crushed and sieved through 0.25 mm meshes (Vm-0.2 mm). The illite-enriched fraction was separated from 2 mm soil sMdz-2 mm by sieving through a 0.2-0.25 mm sieve (sMdz-0.2 mm).

(2) Preparation of soil contaminated with radionuclides

All soil samples throughout the project were artificially contaminated with radionuclides ¹³⁷Cs and ¹³⁴Cs. The source of the radioactive contamination was radioactive water. The sieved soil was mixed in

several doses with radioactive water at a ratio of 1:0.4 by weight and then air-dried. The entire sample preparation process lasted 8-10 days.

All soil samples with particle sizes of 2-3 mm were contaminated with ¹³⁷Cs and ¹³⁴Cs, as described above. The level of Cs activity in soils was within the ranges of 6,000-10,000 Bq/kg (sANPP) and 5,000-6,000 Bq/kg (sMdz). For contamination of sMdz-0.2 mm and Vm-0.2 mm, weighed samples were placed into packets from the filtration material and then immersed into the radioactive water. Twenty-four hours later, the contaminated soil and Vm were removed from the bags and then air dried. The Cs activities in the samples of sMdz-0.2 mm and Vm-0.2 mm were approximately 6,000 and 2,500 Bq/kg, respectively.

(3) Polymers

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

i) Two variants of polyvinyl alcohol (PVA) samples

This is the known industrially produced grades and our own elaborations as modified polyvinyl alcohol (MPVA). PVA-s contains mainly hydroxyl and acetate functional groups at different ratios; MPVA also has carboxyl-containing units. PVA and MPVA samples are water-soluble powders; their solutions of concentrations (0.5-3.0%) were used for the soil tests.

ii) A newly developed VA-AA polymer system based on vinylacetate (VA-AA copolymers)

These copolymers are various forms of water-dispersion products. Variations in the ratio of comonomers and the synthesis conditions allow polymers to be obtained over a wide range of properties, from solutions to dispersions. Diluted solutions/dispersions (0.5-3.0%) were used for the soil test. Hereafter, both polymeric systems will be referred to as "polymer solutions".

3.2.2 Methodology of the soil experiments before April 2014

At the beginning of the works, we have supposed to study the downward stream of polymer solutions in the soil, i.e. the polymer solutions should be applied over the top layer of soil. The height of the experimental vessels (6-7 cm) was chosen based on knowledge that the main amount of radioactive Cs is distributed in the upper layers of the soil (up to 3-5 cm). A plastic tube (height 6 cm, diameter D-6, 2 cm) was filled with a contaminated and moistened soil sample (~ 250 g). Next, 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 then, the second portion of polymer solutions was applied over the second layer of the plastic tube with a height of 1-1.5 cm was cut. The radioactivity of the initial soil, all of the layers, and the remaining part of the soil were measured. This volume of experiments is referred to as Stage 1.

3.2.3 Main results and conclusions from the soil experiments (before April 2014)

Solutions of several samples of PVA, MPVA and VA-AA copolymers were tested using the above-described methodology. Two series of experiments were also performed with the same solutions, to which water-spore suspensions of soil fungi (Aspergillus niger, Aspergillus flares, Aspergillus apricalis,

Alternaria alternata, and Rhizopus stolonifer) were added.

The following primary effects were discovered at this stage of the experiments:

i) Increasing activity of ¹³⁷Cs and ¹³⁴Cs in the upper soil layers was noted in the case when some polymer solutions (two grades of polyvinyl alcohol) were poured over the top soil layer.

ii) For all other tested solutions, the opposite effect was observed: increasing of Cs activity in the lower layer, consistent with the movement of solutions.

iii) The additives of fungal spores to the polymer solutions showed a significant change in the dynamics of motion of Cs. The presence of fungi led to increasing Cs, mainly in the lower layers.

The basic conclusions are:

i) The behavior of polymers in soil is similar to water, with manifestations of capillary and gravitational movement.

ii) Under the experimental conditions (solutions flow top-down), the capillary and/or gravitational movement of radionuclides in the soil occurs under the influence of polymer solutions.

iii) 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 the increase of ¹³⁷Cs in the upper or lower layers due to binding with polymers, further studies would be conducted to identify the role of each parameter of the polymeric systems.

iv) The observation of increasing ¹³⁷Cs in the topsoil as a result of the capillary movement of polymer solutions demonstrated the need for a separate study of the capillary and gravitational behavior of polymers.

A series of experiments was performed in Stage 1, but the polymer solutions were applied so that they penetrated into the soil upward (from bottom to top). The results of all experiments with different polymer solutions (on one type of soil, sANPP) have identified the main factors regulating the efficiency of the redistribution of 137 Cs in soils:

i) Initial soil moisture (ISM)

ii) Volume of the polymer solutions

iii) Ratio of the hydrophilic and hydrophobic functional groups in polymers and the presence of ionogenic groups

iv) Morphological state of the polymer systems (solutions or dispersions)

The conclusions based on analysis of all of the above-described findings formed the basis of detailed studies after April 2014.

3.2.4 Radiocesium interception potential (RIP)

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

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

Table 2 RIP values for different soils

The data obtained are correlated with the data of RIP values for various soils and clay minerals in Fukushima provided in the several documents (in Japanese) from Dr. Onishi [1, 2].

3.2.5 Polymer systems study after April 2014

Based on the results before April 2014, a set of studied polymer systems was changed.

1) The number of PVA samples was supplemented with new grades, which, together with the previously selected samples, allowed the estimation of the impact of various polymer characteristics on the behavior of polymer solutions in soil, such as the ratio of hydrophilic/hydrophobic functional groups, viscosity and molecular weights.

2) Based on the test results of the PVA and VA-AA copolymers, the methods for introducing hydroxyl groups into VA-AA copolymers were determined.

As a result, a new technology of synthesizing the hydrolyzed VA-AA copolymers (VA-AA-H) was developed. These copolymers have the optimal properties for use in soils from the perspective of the project tasks.

3) The developed technology enables the synthesis of polymers with reproducible characteristics. A pilot sample VA-AA-H-80 was synthesized by this technology; this pilot sample was then used in all of the batch tests (see Stage 4 below).

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

3.2.6 Experimental stages

The experimental stages for the soil tests throughout the project are defined as Stages 1–4 *Stage 1* Experiments in vessels (height of 6 cm, D of 6.5 cm), as described above.

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

Stage 3 Experiments in columns with heights of 35-40 cm, similar to the experiments of Stage 2. Soil samples with weights of 1.2-1.5 kg were used.

Based on the analysis of all results, the parameters influencing the processes in these volumes under the experimental conditions cannot be evaluated comprehensively.

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

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

Thus, all experiments at the ANPP within 7-8 quarters were performed under conditions of stage 4 as small batch tests. Both downward and upward experiments were performed in sANPP, sMdz, and sTh (soil particle sizes of 2-3 mm); in addition, downward experiments were performed in soils 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%) are 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 a tray on a holder (ring or mesh with large pores) with a height of 0.5-1 cm. In the upward experiment, the tray is filled with a required amount of polymer solution. In the downward experiment, a solution is poured over the top layer gradually. In both cases, the time of complete absorption of solution into soil is registered.

For this experiment, the method of soil sampling (similar geological core) by a sharpened plastic tube with an inner diameter of 3.5 cm and a height of 30-40 cm was used.

The tube is inserted into the studied soil to the bottom, pulled out, and then cut into several pieces, from which a soil sample is taken for analysis (¹³⁷Cs, ¹³⁴Cs). Three samplings are performed with intervals of 24, 48 and 120 hours. After sampling the first tube with soil, an empty tube is inserted into the empty soil space so as to not disturb the equilibrium of the system; the same procedure is performed for the second and third samplings.

3.2.7 Mathematical analysis - Application of Washburn equations

The Washburn equation was used to compare the capillary rise of polymer solutions for all tested soils; the equation 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 the surface tension of liquid (polymer solution), θ is the wetting angle, *h* is the length of the capillary region filled by solution at the current moment, η is the viscosity of liquid, and *r* is the radius of the pores.

The values (η, σ) for all of the tested polymers solutions were experimentally determined. The kinetic curves of the polymer solution rise were used to assess the pore size *r* and wetting angles θ .

The method was implemented to select the pairs of contact $angle\theta$ and pores diameter *r* such that the theoretical kinetic curves of the solution rise coincide with the experimental ones. This approach allows the pore sizes through which the polymer solutions penetrate in the soil capillaries to be estimated.

3.3 Works with polymer solutions

3.3.1 Methodology after April 2014

Based on the work performed within the quarters 7-8, and on a comparative analysis of all experiments performed in the polymer laboratory and at the ANPP, a method of experiments was obtained in which the capillary tube data serve as guidance for the scaled experiments. This methodology includes the following steps:

i) First, in tubes with soil (of known ISM), the kinetics of the capillary rise of polymer solutions are determined, along with the humidity distribution and the volumes of absorbed liquids.

ii) Next, reasoning from the data of step 1, optimal samples (polymer solutions with the required surface tension and viscosity that provide capillary rise of solutions) are selected.

iii) Next, the selected solutions are tested in tubes with contaminated soils at the ANPP (upward experiments). Based on the obtained data, the polymer solutions for scaled experiments (batch tests) are selected.

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

3.3.2 Results after April 2014

In the following, an example of this methodology is described in the case of soil sMdz-0.2 mm with ISM of 6.5%, soil weight of 4 kg, and top-down (downward) polymer solution application. A series of VA-AA-H copolymers with lower viscosity was synthesized for testing on sMdz-0.2 mm and Vm-0.2 mm.

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 2-3 mm. Sample PVA GF is one of the reference samples in the development of a series of copolymers of VA-AA-H and is shown for comparison.

Sample	Content of functional groups,		
VA-AA-H	% weight		
("Tests")*	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

Table 3 VA-AA- H copolymers

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

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

1 5 71				
Parameters	Solutions, 0.23-0.24%			
	VA-AA-H	VA-AA-H	VA-AA-H	PVA
	80	83	87	GF
Surface				
tension,	58	71	62	51
mN/m				
Viscosity,	1.42	1.57	1.38	1.25
mm ² /sec				
Density,	0.9986	0.9987	0.9989	0.9986
d g/cm ³				

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

Kinetic curves of the rise of the polymer solutions in soil Mdz-0.2 mm were determined in the tubes (Stage 2) (Fig. 1a).



Fig. 1 (a)- Kinetic curves of the rise of the polymer solutions (a) and pore-size distribution (b), sMdz-0.2 mm

The radius of pores through which capillary rise occurs are defined for each solution based on the kinetic curves. To accomplish this task, we built a theoretical curve of the Washburn equation, substituting the experimental data from Table 4 and adding selecting values for r so that the experimental and theoretical curves coincide. The contact angle θ is 89.8^o (this value was obtained according to our numerous definitions).

The pore-size distribution with height (Fig. 16) shows that up to a height of 20-25 cm, the kinetics of the rise of solutions 83 and 87 were close to that of water; however, the polymer solutions penetrate through the larger pores.

Polymer solutions 83 and 87, as well as PVA GF, were tested under the same conditions in tubes with contaminated soils at the ANPP. Four tubes were filled with soil, and two of these tubes were used for Test 83. The soils in tubes for Test 87, PVA GF and one tube for Test 83 were analyzed after 24 hours, with the second one being analyzed after 48 hours after the experiment began (Fig. 2).

Fig. 2 shows distributions of the specific activity of ¹³⁷Cs in the soil layers depending on the duration of the experiment.



Fig. 2 Distribution of ¹³⁷Cs in sMdz-0.2 mm with height, upward experiment (tubes)

An unexpected result was that sample 83 promotes the movement of Cs down, and a tendency of capillary rise of Cs was not observed, even after 4 days. Sample PVA GF (low-molecular-weight polyvinyl alcohol) is similar to sample 83;

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

Solution VA-AA-H 87 (0.24%) was tested on contaminated soil at the ANPP in the batch experiment. The difference in 137 Cs activity between layers is approximately 30%. The polymer solution penetrates into the pores only to a depth of 10 mm. Equilibrium is observed in the system after 48 hours.

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

3.3.3 Discussions on mechanism

The main conclusions resulting from the conducted research: binding Cs with polymer solutions and movement in soil occurs via the same mechanism, regardless of the type of clay mineral (montmorillonite, illite or vermiculite).

A possible explanation of this phenomenon at this stage of investigation is the similarity with the known processes of intercalation and exfoliation of the clays resulting from adsorption of the polymers molecules from its solution.

To include Cs into the polymer solution, the Cs binding with the active centers of the clay lattice should be weakened or broken. The roles of the polymeric macromolecules in Cs binding are as follows:

To weaken the binding of Cs with the minerals due to binding with its own groups;

- To keep Cs in solution by coordinating with its group and absorption to prevent the reversible binding with clays;
- Remove Cs from the inter-packet space and drag it into the pores, where capillary and gravitational forces appear.

The tested polymers 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 the nano-scale level is one of the important factors to enhance the efficiency of Cs removal from the soil.

After the Fukushima accident, Japanese authors and other authors examined the mechanisms of Cs adsorption, especially on pure minerals, such as illite, vermiculite, in detail using 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 the structure of Cs-containing clay particles at the level of approximately 1 nm and active centers of clay minerals. Consequently, a more accurate understanding of the reasons for weak desorption of Cs from illite and vermiculite was obtained.

Currently, the main conclusion from these investigations is that Cs cannot be easily removed from interlayer spaces. Any effective remediation measures must degrade the illite/vermiculite clay fraction. One possibility to accomplish this task is to expand the interlayers via 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 be used to develop methods to improve the efficiency of removal of Cs from clay minerals.

3.3.4 Evaluation of polymer biodegradation in soil

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

PVA is known as a biodegradable polymer and is included in the Japan BioPlastics catalog as GreenPla. Any modifications to PVA continue to be studied; moreover, standardization of methods for determining biodegradability, including the techniques of ISO, is in progress.

Samples of PVA and its carboxyl derivatives used in the project generally correspond to the classification GreenPla. The newly developed VA-AA-H copolymer systems have functional groups that are standard for biodegradable polymeric systems and are suitable for use in soil.

To assess the extent of biodegradation of the VA-AA-H polymers in soil we used one of the accepted methods of such studies - effect of soil microorganisms (fungi and bacteria) on the polymer samples. In this case, changes in the amount of polymer in the soil over time, as well as changes in the composition of soil fungi under the influence of the polymers, are studied.

Several studies with durations of 14, 28, or 60 days were performed in soils sMdz-2 mm and sANPP-2 mm. In each series of experiments, the newly developed VA-AA-H samples were tested in parallel with PVA-grade "Mowiol", which is considered a fully degradable polymer. The experimental results indicated that polymers undergo degradation in soil (likely the stepwise conversion of the polymer and destruction); in line with this, favorable conditions are created in soil for the activation of specific

species of fungi responsible for specific reactions. The dependence of the results on soil type as well as on the soil sampling season (summer or autumn) was revealed.

The results for the newly developed VA-AA-H series are similar to the results of "Mowiol". Based on alterations in the compositions of fungi, there are no dramatic changes in soils from the mycology perspective due to the polymer "intervention". The results of the experiments demonstrate that according to variation in compositions of fungi, the newly synthesized copolymers undergo biodegradation by similar mechanisms as those of "Mowiol". Thus, at this stage of investigation, it can be concluded that VA-AA-H copolymers are not harmful to soil microorganisms.

4. Conclusions

i) For the first time a redistribution of radioactive Cs in the radiation-contaminated soils influenced by polymers solutions is discovered.

ii) Increased activity of ¹³⁷Cs and ¹³⁴Cs in the upper layers of soils due to certain polymer solutions was observed. This phenomenon is due to the manifestation of the capillary movement of polymer solutions in soils.

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

iv) Based on these results, the mechanism for Cs binding and movement with the polymer solutions is the same for all of the studied minerals. The mechanism of this process was suggested.

v) The types of polymeric systems and parameters responsible for ¹³⁷Cs and ¹³⁴Cs binding were determined. vi) The technology for the synthesis of such copolymers at the laboratory scale was developed and refined in the process of scaling the experimental volumes. A pilot sample was synthesized by the developed technology and used in several batch tests at the Armenian NPP.

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

viii)The methodology for the preliminary assessment of the efficiency of polymeric systems to bind and move Cs in soils was elaborated.

5. Applicability and Recommendations for Japan

i) The results of the studies showed the principal possibility of using the revealed regularities of increasing ¹³⁷Cs and ¹³⁴Cs in the top layers of soils under the influence of polymer solutions to reduce the volume of contaminated soils removed from the fields.

ii) Several series of "small batch mode tests" at the ANPP were performed to define the main technological parameters and their influence on the efficiency of ¹³⁷Cs and ¹³⁴Cs redistribution in soil. Difference in ¹³⁷Cs between layers reached up to 40-42% in sANPP (enriched by montmorillonite), and 30-32% in sMdz (enriched by illite).

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

iv) A further change in the experimental volume should be regulated by increasing the vessel height without significantly widening the vessel. At this stage, all parameters to ensure an effective process are

JAEA-Conf 2016-002

expected to be defined. More specifically, the "small batch tests" can be considered as the stage at which the results can be used to predict expected changes in the case of further scaling of the experimental volume. v) Analysis of the published works related to the mechanisms of Cs adsorption-desorption on illite and vermiculite shows that more in-depth studies of the interaction of polymer molecules with clay minerals and Cs on the molecular level may be required to improve the efficiency of Cs redistribution in soil.

Acknowledgement

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

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3.3 Remediation of Cs-contaminated soils through regulation ¹³⁴Cs and ¹³⁷Cs soil-planttransfer by polymeric sorbents

ISTC Project A-2072

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Project Information

Title of the Project:	Remediation of Cs-contaminated soils through regulation ¹³⁴ Cs and ¹³⁷ Cs soil-plant transfer by polymeric sorbents	
Commencement Date:	June 1, 2013	
Duration:	2 years	
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Executive Summary Report of ISTC Project A-2072

1. Objectives / scope of work and technical approach / expected results

1.1 Project Goal

The project goal 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 <u>water – soil – plant</u> system.

1.2 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.
- Assessment of the effectiveness of polymers in soil and hydroponic culture of plants;
- determination of radioactive cesium content in <u>water soil plant</u> and <u>water nutrient solution plant systems;</u>
- determination of migration and accumulation of radioactive cesium in soil layers (0-5; 5-10; 10-20cm), above-ground and under-ground parts of plants depending on application of the polymer; revealing the dependence of redistribution of radionuclides in systems <u>water soil plant</u> and <u>water nutrient solution plant</u> on the type of a polymer material, variety of a plant and number irrigation.

1.3 Methods

For copolymers obtaining: radical (co)polymerization of vinyl monomers; plant production by hydroponics techniques and traditional soil culture. Low-background gamma spectrometer with pure Ge detector (Canberra production) and supporting "Linx" and "GENIE-2000" software, Low-background beta-spectrometer supporting "PROGRESS" software, Low-background *i*Matic[™] Automatic LB Counting System (Canberra production) were used.

Analyses: chemical, radiochemical, physiological-biochemical analyses.

1.4 Expected results:

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

2. Obtained results

In this research novel composite adsorbents consisting of inorganic filler (bentonite, zeolite) and binding polymer were prepared. The composite was evaluated for removal of radioactive cesium from aqueous solutions and soils. The developed polymer sorbents demonstrated efficiency of actions for Cs-contaminated water purification without post filtration. Despite the type of polymer matrix the additions of filler and ferrocyanides increased adsorption of radioactive cesium.

Some effective polimeric sorbents were chosen for plant cultivation. At the beginning of study

Sample 13 had maximal high water purification coefficients for ¹³⁷Cs and ¹³⁴Cs and was used for the first and second vegetative seasons of plant growing. Sample 13 was the most stable one. Upon further investigations new compositions with highest uptake of radioactive cesium were developed. Compositions 13* and 73* have the greatest level of absorption of radioactive cesium and were selected for second vegetative seasons of plant growing. Sample 13, Sample 13*, and Sample 73* have excellent mechanical property, and easy separation from aqueous solutions and soils. At the given period Sample 13 has the potential commercial application.

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

• Applying of polymers in Japanese basil culture alowes to cut in half nourishment frequency in hydroponics and in 25% irrigating frequency in field, and ensure the same and even high yield of leaves (for food) with high quality indices.

• Applying of polymers in Redroot pigweed soilless 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 lowered accumulation of 1¹³⁷Cs in basil leaves both in hydroponics and soil.

• Sample 13 and 13* polymers lowered ¹³⁷Cs concentration in the soil layer by 10-20% compared with control.

• The use of Sample 13 and 13* in soil and hydroponics will support collection more ecologically sound biomass for food.

• Separate application of Sample 11 and Sample 13 polymers in hydroponics promoted the accumulation of ¹³⁷Cs in Redroot pigweed biomass and decrease ¹³⁷Cs concentration in the soil layers, but their joint application, on the contrary, prevented ¹³⁷Cs accumulation in biomass of Redroot pigweed both in hydroponic culture and in soil, as well as didn't influence on the ¹³⁷Cs concentration in the soil layers.

• Co-growing system wasn't effective: polymers promoted the decrease of ¹³⁷Cs accumulation both in basil leaves and in pigweed whole biomass (we expected ¹³⁷Cs content decreasing in basil leaves and increasing in pigweed biomass).

• Based on our initial experiments there is an opportunity to obtain ecologically sound soybean in Cscontaminated soils by using polymeric sorbents (additional investigations are necessary).

• In Cs-contaminated soil by radiocaesium absorbance capacity polymers have the following trend: 73*>13*>13.

The present work describes potential countermeasures to control radioactive Cs transfer from soil to agricultural plants. In conclusion, we propose new countermeasures to Fukushima, namely, the use of polymeric sorbents for Cs-contaminated water and soils.

During the project implementation compositions were synthesized by room-temperature polymerization technology. These compositions have high efficiency uptake for ¹³⁷Cs and ¹³⁴Cs from water. The compositions have not been tested in the field conditions, as their physical-and-chemical properties were studied in the end of the 2nd year of the project implementation.

The room-temperature polymerization technology is of great interest as more feasible method of

gels obtaining. Therefore, the search for adsorbents obtained through this technique will be continued in future as a follow-up of our investigations.

Keywords: radioactive cesium, accumulation and transfer, polymeric sorbent, composition, stability, purification coefficient, soil and hydroponics conditions, plant productivity.

3.4 Development of a set of measures for production of assured quality agricultural goods under radioactive contamination conditions – Experiments of Cesium Uptake by Livestock and Crops

ISTC Project K-2085

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Project Information

Title of the Project:	Development of a set of measures for production of assured quality agricultural goods under radioactive contamination conditions	
Commencement Date:	September 1, 2013	
Duration:	2 years	
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Participating Institutes:	none	
Foreign Collaborators:	Dr. Yasuo Onishi (Pacific Northwest National Laboratory, U.S.A.)	
	Dr. Fesenko Sergey (International Atomic Energy Agency)	

Executive Summary Report of ISTC Project K-2085

1.1 Objectives / scope of work and technical approach / expected results

1.1.1 Project goal

The project goal is to obtain quantitative data on the characteristics of radionuclides transfer in agricultural products and the impact s of various factors and events on these processes. Based on these data, to develop recommendations for production of assured quality agricultural goods at their production in the conditions of radioactive contamination.

1.1.2 Scope of work and technical approach.

The basis for obtaining quantitative variables was field-scale experiments with farm animals and plants in the STS radioactive contamination conditions. The experiments were carried out in the ecosystems with elevated radionuclide concentrations in the components of the natural environment. During the experiments with farm animals we considered radionuclide uptake by the experimental animals with soil, water, plants. We also studied radionuclide distribution on separate organs of the crops and influence of some physical and chemical properties of soils on the ¹³⁷Cs accessibility to the feed components of the species studied.

1.1.3 Expected result

The studies provide the following results:

• data on the distribution of radionuclides in pigs and poultry (chickens) at various forms of radionuclides uptake with components of the diet (soil, water and plant food);

• transfer factors of radionuclides in pig and poultry products at various forms of radionuclides uptake with components of the diet (soil, water and plant food);

• dynamics radionuclides concentrations in pork and crop products at various forms of radionuclides uptake with components of the diet (soil, water and plant food);

• dynamic of radionuclides excretion from the body of pigs and chickens. Half- periods in radionuclides concentrations in pig and poultry products at transition to a "clean" diet (uncontaminated);

• transfer factors of radionuclides in crops (potatoes, beets, cabbage, wheat), being the main in a food basket and diet of farm animals (pigs and chickens);

• dependencies of radionuclide transfer in the investigated species of wild and agricultural plants on the physical and chemical parameters of the soil;

• recommendations on the production of assured quality agricultural goods to be produced in conditions of radioactive contamination.

1.2 Obtained results

Study of the ¹³⁷Cs distribution in the piglets' bodies showed that unlike the tropic radionuclides, ¹³⁷Cs is distributed in the body in all tissues and organs. However, the difference between the highest and lowest ¹³⁷Cs activity in certain tissues and organs of the body can be up to 8 times. With long-term uptake the greatest concentrations of ¹³⁷Cs recorded in the muscle tissue and kidneys, the lowest in the brain and blood. At long-term uptake the ¹³⁷Cs activity in the body decreases in the series: kidneys> muscle> tongue> heart> liver> bone tissue> spleen> lungs> brain> blood. At the same time, the nature of ¹³⁷Cs distribution in organs and tissues is not dependent on the form and source of the radionuclide uptake by the body. In general, one can say that after prolonged entry of ¹³⁷Cs in the body its distribution has a specific pattern that allows the prediction of this radionuclide concentration in one organ by its concentration in another.

In the conditions of long daily uptake of ¹³⁷Cs by the pigs with different components of the environment, the concentration of ¹³⁷Cs in organs increases to a certain value, followed by a dynamic balance between accumulation and excretion. No further increase in ¹³⁷Cs activity in organs occurs. Thus, the equilibrium state for the various organs and tissues comes on the 28th day. After a long uptake with water and feed, ¹³⁷Cs activity concentration half-life after the animals shifted to a "clean" diet does not exceed 14 days and on the 56th day ¹³⁷Cs is excreted almost entirely. At uptake with the soil the activity concentrations half-life do not exceed 7-10 days, and on the 56th – excreted almost completely.

Comparative studies showed no obvious differences in the factors of transfer into domestic and wild pigs, regardless of the radionuclide species in the soil, which allows the use of the transfer factors obtained for the domestic pigs to evaluate the possible Cs concentrations in the wild pigs.

The state of equilibrium for the muscle tissue is characterized by 28 days, for the liver – 8 days. The half-life for the muscle tissue of chickens was 8 days. In the chicken's liver on the 2^{nd} day the birds shifted to a "clean" diet, the radionuclide concentration does not exceed 50% of the maximum. The state of equilibrium in the egg at different sources of uptake occurs on the 21^{st} day and no further increase in activity concentrations occur. After the chickens shifted to a "clean" diet the ¹³⁷Cs concentration in the egg on 6th day does not exceed 50% of the maximum.

The studies found that the obtained transfer parameters for the crops are lower than the international data on beet, potato, sunflower, barley and wheat by 2 orders of magnitude (more than 100 times), corn, buckwheat, saikon and colza - 1 order of magnitude (more than 10 times). ¹³⁷Cs TF for all the studied crops is within the range of $n \times 10^{-2}$ - $n \times 10^{-4}$.

High doses of chemicals (KCl, CaCl₂ and Na₂SO₄) lead to an increase in ¹³⁷Cs TF for wheat: high doses of KCl (more than 439 g/m² (higher the potash norm by 16 times)) leads to increased accumulation of ¹³⁷Cs in the vegetative organs of wheat by 1.3-5.3 times in comparison with the control (without application of chemicals; adding CaCl₂ (over 360 g/m²) leads to increased accumulation of ¹³⁷Cs in all organs of wheat up to 4 times, and a larger dose of CaCl₂ (735 g/m²) on the contrary reduces the accumulation of radionuclide by 1.2-2.1 times; higher dose of Na₂SO₄ (470 g/m²) increases ¹³⁷Cs TF in comparison to control for wheat stems by up to 6 times and reduces ¹³⁷Cs TF for head and root by 1.1 times and 1.4 times, respectively; high dose of Na₂SO₄ (940 g/m²) does not affect the ¹³⁷Cs accumulation in the roots, and fro the above-ground parts of wheat on the contrary increases the accumulation of radionuclide by 4.9 times.

The use of fertilizers in accordance with the recommended agricultural norms does not affect the ¹³⁷Cs TF or has little effect for tomato (an exception is potassium fertilizer –TF increase up to 1.9 times in leaves). Adding $\frac{1}{2}$ norm of fertilizers leads to increased accumulation of ¹³⁷Cs from 2.9 to 5.2 times in the tomato leaves. Adding to the norms of nitrogen fertilizers and manure results in a decrease of ¹³⁷Cs by 1.2 times and 2-2.5 respectively.

For the sunflower the use of fertilizers in accordance with the norms and $\frac{1}{2}$ norm lead to a decrease of ¹³⁷Cs in leaves by 1.1-16 times and 1.7-3.3 times, respectively. Adding $\frac{1}{2}$ norm of phosphate and potash

fertilizers, norms of potash fertilizer and manure leads to increased accumulation of ¹³⁷Cs by 2.5-2.8 times, 1.4 times and 1.8 times, respectively, in the roots of sunflower.

Additionally, the studies have established that application of the fertilizer half the norm, reduces 137 Cs accumulation for the buckwheat, compared with the control, for the seed and leaves – more than 1.1 times and 2.6 times, respectively. The use of fertilizers in the recommended rate reduces 137 Cs transfer by 5.9 times for leaves, and for the seeds – 7.1 times. However, the using potash in norms increases the 137 Cs transfer by 3.2 times in buckwheat seeds. Adding to the double norm of fertilizer reduces the 137 Cs transfer into the seeds and leaves from 3.4 to 11 times. It should be noted that using double norms of nitrogen fertilizers significantly increases the radionuclide transfer into the buckwheat seeds and is more than 50 times higher than the control.

Keywords:

Semipalatinsk Test Site, agricultural products, cesium-137, food safety

3.5 Methodology for long-term radiation monitoring to dose assessment using radiological zoning and modeling of radionuclides migration in environmental and food chains – Countermeasures of Cesium Uptake by Farm Crops and Livestock

STCU Project 5953

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Project Information

Title of the Project:	Methodology for long - term radiation monitoring to dose assessment using radiological zoning and modeling of radionuclides migration in environmental and food chains
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Foreign Collaborators:	Dr. Yasuo Onishi (Pacific Northwest National Laboratory, U.S.A.)

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 the April 2014 meeting

Participants of the project are based on the thesis that the developed methodology of radioecological 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 ¹³⁷Cs from 1 to 1000 kBq \cdot 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 - 37.934°, S- 37.686°, W - 140.465°, E - 140.766°. Area test site 728.7 km², 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.

JAEA-Conf 2016-002



Fig. 1. The test polygon on the map of soil ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs resuspension in the contaminated zone around the Fukushima-1 NPP.

(4) Evaluation of the ¹³⁷Cs 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





Fig. 2. The structure of the system of predicting the radiation situation, agrosphere monitoring and radiation quality control. It reads "Conclusion and submission of results" on the bottom ring in purple.

• *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:

 \circ consistent interaction and successive execution of the models included in the subsystem "Radiation situation forecast and modeling",

 \circ space analysis and evaluation of the radiation situation, formation of a monitoring network at regional and local levels,

 \circ 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 (<u>http://nomads.ncdc.noaa.gov</u>). 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 (1³¹I, 1³⁷Cs, ⁹⁰Sr) 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 ¹³¹I, ¹³⁷Cs, ⁹⁰Sr.

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 <u>the</u> <u>model of plant aerial contamination AeralPlant</u> is used (Priester, 1975 [1], 2008 [2]). 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:

$$C(t) = \sigma \cdot PF \cdot (a_1 \cdot e^{-0..693 \cdot \frac{t}{T_1}} + (1 - a_1) \cdot e^{-0.693 \cdot \frac{t}{T_2}})$$
(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 ¹³⁷Cs concentration in leafy vegetables within the selected polygon was high and reached 120 - 6 000 kBq \cdot kg⁻¹. The operation intervention level OIL6 (IAEA, GSG-2) under the emergency is 2 kBq \cdot 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs. These calculations make it possible to recommend for Japanese specialists to reconstruct the concentration of ¹³⁷Cs and ¹³¹I 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 ¹³⁷Cs 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 ¹³⁷Cs contamination of leafy vegetables after the accident at Fukushima-1 NPP in these monitoring points, as well as the dynamics of ¹³¹I 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.



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 (Priester, 2008 [4]) (curves): a - ¹³¹I for monitoring points en_2.7 (1), en_2.6 (2) and en_2.3 (3);



(b)

Fig. 7. (continued):

b - ¹³⁷Cs for monitoring points en_2.1, en_2.8 and en_2.4.

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.

As one could be seen in Fig. 7, the contamination dynamics are well approximated by twoexponent 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 ¹³¹I 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 <u>the root contamination of crop production</u> 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 ¹³⁷Cs transfer from the soil of 4 types in harvest of 16 crops (Priester *et al.*, 2003 [3]).

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

$$SA(t) = \sigma \cdot TF(0) \cdot \left\{ \frac{k_1 - (2 \cdot k_2 + \frac{k_3}{2})}{k_1 - k_2} \cdot e^{-(k_1 + \frac{k_3}{2}) \cdot t} + \frac{k_2 + \frac{k_3}{2}}{k_1 - k_2} \cdot e^{-(k_2 + \frac{k_3}{2}) \cdot t} \right\}$$
(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* (Priester, 2002 [4]; Priester *et al.*, 2003 [3]). 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:

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

The calculation of the ¹³⁷Cs 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 ¹³⁷Cs 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.



Milk contamination in pastures (below the standard 50 Bq/kg) 1.1 to 35

Fig. 8. Calculations of the ¹³⁷Cs 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 ¹³⁷Cs concentration in milk in 1 year after the accident, when the grass contamination caused by root path, do not exceed 35 Bq \cdot kg⁻¹, which is less the limit level LL = 50 Bq \cdot 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 Figs. 8 and 9 show the concentration ranges of crops contamination, compared with the national norm LL. For all crops, except for milk, LL of ¹³⁷Cs content in the product is 100 Bq \cdot 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 ¹³⁷Cs 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 ¹³⁷Cs contamination. In large areas the radionuclide concentration is in the range 70 - 300 Bq \cdot kg⁻¹ or even more. For cabbage a small number of excess of 70 Bq \cdot kg⁻¹ has been obtained.

As for the test site in Ukraine, the concentration of ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 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 Sirakawa Oya compared with the modeling results.



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



Fig. 11. The forecast values of ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs soil contamination density. The data of control held in Tomino in 1 year after the accident show that the ¹³⁷Cs concentration in soybeans did not exceed 100 Bq \cdot kg⁻¹. The considerable variability in the properties of density and soil contamination was found, which are confirmed by the control data: ¹³⁷Cs 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 \cdot 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 ¹³⁷Cs 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 ¹³⁷Cs 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 worstcase 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 \cdot 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 ¹³¹I 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

4.1 Conclusions for general cases

(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.

4.2 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³¹I and ¹³⁷Cs 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 ¹³¹I and ¹³⁷Cs concentration in leaves could be adequately described by the two-exponent model. The periods of leaves semi-cleaning for ¹³¹I (2.8 and 6.0 days) and ¹³⁷Cs (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 ¹³¹I and 6.6 and 59 days for ¹³⁷Cs.

The large absolute values of ¹³¹I ($0.29 - 12.3 \text{ MBq} \cdot \text{kg}^{-1}$) and ¹³⁷Cs concentration ($0.045 - 2.90 \text{ MBq} \cdot \text{kg}^{-1}$) 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 ¹³¹I 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 ¹³¹I and ¹³⁷Cs in leafy vegetables.

(13) The estimation of the ¹³⁷Cs 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 \cdot kg⁻¹. Conservative estimates made using the maximum values of the ¹³⁷Cs metabolism parameters in the chickens body show that the ¹³⁷Cs concentration in chickens meat and eggs at outdoor keeping will be below the norm in Japan 100 Bq \cdot kg⁻¹.

5. Applicability and Recommendations for Japan

5.1 Applicability 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).

5.2 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 ¹³⁷Cs 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 ¹³⁷Cs 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.

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3.6 Monitoring of radioactive pollution of forest ecosystems after accident on Chernobyl NPP-Rehabilitation with mushrooms harvesting in forest ecosystem

STCU Project 5954

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Project Information

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Commencement Date:	September 01, 2013	
Duration:	2 years	
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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 ¹³⁷Cs 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 ¹³⁷Cs. The forests remediation problem could not be solved without preliminary thorough study of ¹³⁷Cs accumulation in various forest and soil objects. In addition, ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs in forest ecosystems to create a model of long-term behavior of ¹³⁷Cs 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 ¹³⁷Cs contents in different objects of forest ecosystems from Fukushima Prefecture.

The project main objective was to forecast the behavior and redistribution of ¹³⁷Cs 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 the 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 ¹³⁷Cs 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.

The technical Review 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

3.1 General methodology

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 ¹³⁷Cs and ¹³⁴Cs 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 semiconductor detector of GC6020 "CANBERRA" type, with the registration of the radionuclide content on the personal computer.

Specific activity of ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs in the samples is calculated as follows:

$$\delta \mathbf{A} = \sqrt{\left(\sqrt{\delta \mathbf{S}^2 + \delta \mathbf{S}_{\phi}^2}\right)^2 + \delta \varepsilon^2 + \delta \mathbf{I}_{\gamma}^2},$$

where

 δA - relative error in determining of specific activity of ¹³⁷Cs in the sample (%);

 δ S - relative error in determining of square area of gamma lines of 661.6 keV ¹³⁷Cs in the gamma spectrum of the sample (%);

 δS_{ϕ} - relative error in determining of the area of ¹³⁷Cs - gamma lines in the background gamma spectrum

(%);

 $\delta\epsilon$ - relative error of absolute photo-efficiency of gamma-ray 661.6 keV ¹³⁷Cs 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 ¹³⁷Cs. 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_{ν} was taken from the program package NUCHAT firm CANBERRA and equaled 0.2%.

 $\label{eq:error} \text{Error values } t_{\text{meas}} \text{ and } m \text{ can be neglected.}$

3.2 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

3.2.1 Monitoring program

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. 1.



Fig. 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.

3.2.2 Surveillance network

(1) Network level

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. 2).



Fig. 2 Basic observation network for the radioecological monitoring of forests

(2) 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 ¹³⁷Cs. 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.

(3) The choice of monitoring objects

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.

(4) 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.

3.2.3 Samples on and below the ground surface

(1) 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.

(2) 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).0

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.

(3) Soil itself (mineral soil layer)

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.

3.2.4 Samples over the ground surface

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

(1) 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.

(2) 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.

(3) Moss cover

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.

3.2.5 Mushrooms as ¹³⁷Cs 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 ¹³Cs 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.

(1) Nutrition type (ecological group)

During the first two-three years after radionuclide precipitation, the high intensity of ¹³⁷Cs accumulation by mushrooms-saprotrophs and xylotrophs is detected. Maximum ¹³⁷Cs content values are characteristic for these species during the period.

¹³⁷Cs 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.

(2) Mycelium localization depth

For the mushrooms belonging to the symbiotroph ecological group, the main factor influencing the

¹³⁷Cs accumulated amount is the localization depth of the main part of mycelium, which is characteristic for each species (the quantity of available ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 radionuclides. 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.

3.3 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 ¹³⁷Cs 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 ¹³⁷Cs distribution within vertical soil layers. Maximum ¹³⁷Cs specific activity at these studied testing areas is registered, as a rule, in the ground litter $(A_0 l + A_0 f + A_0 h)$, 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 ¹³⁷Cs 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 ¹³⁷Cs in forest ecosystems.

3.4 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 1 and 2.

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 ¹³⁷Cs 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 ¹³⁷Cs 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.

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 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 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
2	2 70	6197850	15547	0.251

Table 2 The results of mycoextraction performed at stations of the testing area "Ohgawara" (Japan) during single sampling on September 28, 2014.

3.5 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 ¹³⁷Cs content at the station in 2014 at the value of 26,250,590 Bq/100 m².

Theoretical calculations of ¹³⁷Cs 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.

Table 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 m² 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, ¹³⁷Cs 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 ¹³⁷Cs 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).

3.6 Creation of model of migration of ¹³⁷Cs 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 ¹³⁷Cs 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.

<u>*Temporal limits*</u> of the model use are determined from the moment of radioactive precipitates $t_0 = 0$ ad infinitum $t_{max} = \infty$. <u>Sphere of use</u> 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·m². 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 (Fig. 3). Basic principles for the radionuclide behavior model in the system "soil – mushrooms" have been formulated:

¹³⁷Cs 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 $A_0I+A_0f+A_0h$, *Suillus luteus, Russula xerampelina, Lactariusturpis* – in the soil layer A (0 – 5 cm), *Boletus edulis, Leccinum scabrum, Tricholoma flavovirens* – in the soillayer B (5 – 10 cm).



Fig. 3 Conceptual scheme for the model of ¹³⁷Cs 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:

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

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

Litter

$$SA_{l} = TF_{0} \cdot \sum_{i=1}^{n} \mathcal{Q}_{wsi}$$

$$(3.16)$$

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

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

where,

TF₀ - values extrapolated to zero time $t = 0^{137}$ Cs transfer coefficients from soil to mushrooms (Bq·kg⁻¹) / (kBq·m⁻²).

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

in fuel particles	$Q_{fp0} = a_{fp0} \cdot \sigma$	(3.14)
in water – soluble form	$Q_{ws0} = (1 - a_{fp0}) \cdot \sigma$	(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 Tables 4 and 5.

Parameter	Unit of measurement	Value		
Density of soil contamination		σ	kBq·m ⁻²	185
Rate decomposition	A ₀ l	k _{l1}	y ⁻¹	0.6
	A ₀ f+A ₀ h	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				0.80
Rate destruction of fuel particles $A_0 l + A_0 f + A_0 h$		$\mathbf{k}_{\mathrm{fpl}}$	y ⁻¹	0.06
	soil	k _{fps}		0.16
Rate of convection soil moisture $A_0 l + A_0 f + A_0 h$		\mathbf{v}_{l}	cm·y ⁻¹	0.46
	soil	Vs		0.86
Diffusion coefficient	A ₀ l+A ₀ f+A ₀ h	D _l	$cm^2 \cdot y^{-1}$	0.34
	soil	Ds		0.62
Rate of non-exchange radionuclide sorption k _s			y ⁻¹	0.28
Decay rate λ			y ⁻¹	0.022

Table 4 Parameters of ¹³⁷Cs migration in the "soil - mushrooms" system models

Table 5 Value extrapolated to the 137 Cs fallout time transfer from soil to mushrooms TF₀, (Bq·kg⁻¹)/(kBq·m⁻²)

Mushrooms	TF ₀	
Saprotroph (A ₀ l+A ₀ f-	$+A_0h)$	
Paxillus involutus (Batsch: Fr.) Fr.	1150	
Macrolepiota procera (Scop.: Fr.) Sing.	125	
Lycoperdon perlatum Pers.	115	
Armillariella melea (Vahl: Fr.) P. Karst.	65	
Symbiotroph $(0 - 5 \text{ cm})$		
Xerocomus badius (Fr.) Kuhn. ex Gilb.	1120	
Russula xerampelina var. erythropus	750	
Pelt.		
Suillus luteus (L.: Fr.) S.F.Gray	610	
Lactarius necator	470	

Symbiotroph $(5 - 10 \text{ cm})$			
Tricholoma flavovirens (Pers.: Fr.)	2650		
Lund.			
Boletus edulis Bull.: Fr.	1830		
Leccinum scabrum (Bull.: Fr.) S.F.Gray	1650		

Fig. 4 shows dynamics of ¹³⁷Cs 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 ¹³⁷Cs 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.

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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs: 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 Significance of forest ecosystem

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 Advantages of mycoextraction

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

The advantages of phytoextraction (total deforestation) are as follows:

- 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.)

The disadvantages of phytoextraction are as follows:

- 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) are as follows:

- 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 ¹³⁷Cs from contaminated territories;

- During rich years, 0.5 -2 % and more of the total ¹³⁷Cs content in soil could be extracted using the fungi fruit bodies at contaminated territories;

- The collection of objects with high ¹³⁷Cs 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) are as follows:

- 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 ¹³⁷Cs 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. Features of transport model for mycoextraction

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 above influence values of parameters of migration ^{137Cs} on a soil profile and its sorption a soil-absorbing complex.

5. Applicability and Recommendations for Japan

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.

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.

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 $(A_0I+A_0f+A_0h)$ 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 ¹³⁷Cs contamination mushrooms using the data obtained in the above ranges. In the Table 6 presents offered in Japan for the model parameters of ¹³⁷Cs migration in the system "soil – mushrooms".

	U	-		*	
			Value		
Paramet	ers		Unit of measurement	Ohgawara,	Namie
				Iitate	rtuine
Density of soil contamination		σ	kBq⋅m ⁻²	1235 - 2295	13 910
Rate decomposition	A ₀ l	y ⁻¹	year ⁻¹	0.6	0.6
	A ₀ f+A ₀ h			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 ₀ l+A ₀ f+A ₀ h	\mathbf{v}_{l}	cm·y ⁻¹	0.46	0.86
	soil	Vs		0.54	0.94
Diffusion coefficient	A ₀ l+A ₀ f+A ₀ h	D _l	$\text{cm}^2 \cdot \text{y}^{-1}$	0.26	0.62
	soil	D _s		0.30	0.48
Rate of non-exchange radionuclide sorption		ks	y ⁻¹	0.31	0.31
Decay rate		λ	y ⁻¹	0.022	0.022

Table 6 Parameters of ¹³⁷Cs migration in the system "soil - mushrooms" models for Japan

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 7 presents experimental data of ¹³⁷Cs specific activity in mushrooms and calculated values using the developed model.

The table shows that the ¹³⁷Cs 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·kg⁻¹ in even the same polygon station Nol. 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 ¹³⁷Cs 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 ¹³⁷Cs concentration in mushrooms - almost the same and as for *Ramaria spp.* - greater density of 2 times less.

No.	Mushrooms species	¹³⁷ Cs specific activ Bq·l	Relative		
Station		experimental data	forecast data	deviation, ± 70	
	Sampling s	site «Ohgawara»			
	Symbiot	$\operatorname{coph}(0-5 \mathrm{cm})$			
1	Lactarius subvellereus, Lactarius rufus	1970 - 49293	1190	65	
2, 3	Russula emetica	1634 - 52974	1900	16	
	Symbiotro	oph (5 - 10 cm)			
2, 3	Sarcodon aspratus	12592 - 15031			
2	Ramaria spp.	12459			
	Sampling	g site «Namie»			
	Symbiot	$\operatorname{coph}(0-5 \mathrm{cm})$			
	Suillus luteus	49111	17530	280	
Symbiotroph $(5 - 10 \text{ cm})$					
	Sarcodon scabrocus, Sarcodon aspratus	22535 - 35265			
	Ramaria spp.	5425			

Table 7 The ¹³⁷Cs specific activity in mushrooms, taken on Ohgawara and Namie testing sites 28.09.2014 and forecast data using models.

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 obtained under the condition that litter does not have deleted. Then, in Fig. 5*a* presents forecast of mushrooms contamination of differ environmental groups on the above conditions at the site Ohgawara.



Fig. 5 Forecast of ¹³⁷Cs concentration in mushrooms: a - at the site Ohgawara, Japan, b - Ukraine and Japan polygons with same soil contamination

As a common result on the forcast, the lowest concentration of radionuclides characteristic of symbiotroph, mycelium which is 5-10 cm soil layer. Moreover, Fig. 5 shows following features of the ¹³⁷Cs concentration:

1) in saprotroph, the concentration gradually decreases with time after the fallout;

2) in symbiotroph, mycelium which is in soil layer 0 - 5 cm, the concentration increases ranging from 3-4 years, most occur in the 9-10 year;

3) in symbiotroph, mycelium which is in soil layer 5 - 10 cm, the concentration increases from 8-9 years to a maximum of 15-16 years.

Fig. 5b presented for comparison curves predicted of ¹³⁷Cs 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 ¹³⁷Cs 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.

4. Concluding remarks

Secretariat of the Technical Review Committee Meeting of the Fukushima Initiative

ISTC and STCU have been well positioned to tap into unique and substantial regional experience and research related to radionuclide releases into the environment including studies of the Chernobyl power plant nuclear accident and its implications, the impact of uranium tailings in the environment, environmental monitoring of former nuclear test sites, and other areas. Based upon this historical body of knowledge, ISTC and STCU have facilitated the conducting of targeted research projects designed to help Japan in specified areas of nuclear remediation and long-term monitoring, as Japan responds to the challenges posed by the Fukushima accident. This is the ISTC/STCU Fukushima Initiative.

During the Expert Review Meeting, final conclusions of the following six projects were discussed with Japanese national experts alongside their international counterparts to determine possible benefits of the research to the Japanese authorities:

I. Studies of secondary migration of radionuclides and waste treatment

1. Monitoring of radioactive pollution of forest ecosystems after accident on ChNPP (STCU 5954).

2. Methodology for long - term radiation monitoring to dose assessment using radiological zoning and modeling of radionuclides migration in environmental and food chains (STCU 5953).

3. Compaction of radioactive waste produced by decontamination of territories polluted due to the accident at Fukushima Daiichi Nuclear Power Station (STCU 5952).

4. Advanced polymeric systems providing deactivation of different Surfaces

and soil from radioactive pollutions with the use of polymers (ISTC A-2071).

II. Studies to minimize transfer of radionuclides in soil-plant system

5. Development of a set of measures for production of assured quality agricultural goods under radioactive contamination conditions (ISTC K-2085).

6. Project Remediation of Cs-contaminated soils through regulation ¹³⁴Cs and ¹³⁷Cs soil-plant transfer by polymeric sorbents (ISTC A-2072).

Project representatives presented the outcomes of their work with critical review designated experts from Japan, the United States, and the IAEA. The six research projects of the Initiative provided unique understanding and practical approaches to monitoring and remediation following the Fukushima Daiichi nuclear accident in Japan. The projects were funded by the European Union, the Ministry of Education, Culture, Sports, Science and Technology of Japan, and the U.S. Department of Energy's National Nuclear Security Administration.

The review session of the Fukushima Initiative recognized the following achievements:

1. The Initiative facilitated transfer of scientific knowledge from Armenia, Kazakhstan, and Ukraine which have a robust nuclear program as well as they have studied incidences of severe environmental contamination with the former Soviet Union.

2. The sharing of this knowledge is valuable, as it provides greater insight into the complex relationship of such events within the local natural environment and its related ecosystems.

3. The projects' results can be utilized in providing more comprehensive monitoring and remediation of the affected environment in and around the Fukushima site.

4. Although the results are considered to be at a basic fundamental stage of development through continued study, they can be refined in such a manner as to be considered for application in the future.

5. As a result of bilateral collaborative studies between the scientists and their Japanese counterparts, it is possible to derive novel monitoring and remediation technologies with application for the Fukushima area.

6. International collaboration to understand the realities of a severe nuclear accident is important, as it can contribute to more complete mitigation and remediation.

7. Expanded knowledge contributes to the development of more effective operating structures for preventing future events as well as for responding to emergencies.

8. The Initiative was a successful showcase of the benefits of joint activity of the ISTC and the STCU.

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表 1. SI 基本単位						
甘大昌	SI 基本ì	単位				
本平里	名称	記号				
長さ	メートル	m				
質 量	キログラム	kg				
時 間	秒	s				
電 流	アンペア	Α				
熱力学温度	ケルビン	Κ				
物質量	モル	mol				
光度	カンデラ	cd				

表 2. 基本単位を用いて表されるSI組立単	位の例
AI 立 是 SI 組 立 単位	
名称	記号
面 積 平方メートル	m ²
体 積 立方メートル	m ³
速 さ , 速 度 メートル毎秒	m/s
加 速 度メートル毎秒毎秒	m/s^2
波 数 毎メートル	m ⁻¹
密度,質量密度キログラム毎立方メートル	kg/m ³
面 積 密 度 キログラム毎平方メートル	kg/m ²
比体積 立方メートル毎キログラム	m ³ /kg
電 流 密 度 アンペア毎平方メートル	A/m ²
磁 界 の 強 さ アンペア毎メートル	A/m
量 濃 度 ^(a) , 濃 度 モル毎立方メートル	mol/m ⁸
質量濃度 キログラム毎立方メートル	kg/m ³
輝 度 カンデラ毎平方メートル	cd/m ²
屈 折 率 ^(b) (数字の) 1	1
比 透 磁 率 ^(b) (数字の) 1	1
(a) 量濃度 (amount concentration) は臨床化学の分野では	t物質濃度

(substance concentration)ともよばれる。
 (b) これらは無次元量あるいは次元1をもつ量であるが、そのことを表す単位記号である数字の1は通常は表記しない。

表3. 固有の名称と記号で表されるSI組立単位

			SI租工单位	
組立量	名称	記号	他のSI単位による 表し方	SI基本単位による 表し方
平 面 鱼	ラジアン ^(b)	rad	1 ^(b)	m/m
立体鱼	ステラジアン ^(b)	$sr^{(c)}$	1 (b)	m^2/m^2
周 波 数	ヘルツ ^(d)	Hz	-	s ⁻¹
力	ニュートン	Ν		m kg s ⁻²
压力,応力	パスカル	Pa	N/m ²	$m^{-1} kg s^{-2}$
エネルギー,仕事,熱量	ジュール	J	N m	$m^2 kg s^2$
仕 事 率 , 工 率 , 放 射 束	ワット	W	J/s	m ² kg s ⁻³
電荷,電気量	クーロン	С		s A
電位差(電圧),起電力	ボルト	V	W/A	$m^2 kg s^{-3} A^{-1}$
静電容量	ファラド	F	C/V	$m^{-2} kg^{-1} s^4 A^2$
電気抵抗	オーム	Ω	V/A	$m^2 kg s^{\cdot 3} A^{\cdot 2}$
コンダクタンス	ジーメンス	s	A/V	$m^{2} kg^{1} s^{3} A^{2}$
磁東	ウエーバ	Wb	Vs	$m^2 kg s^2 A^1$
磁束密度	テスラ	Т	Wb/m ²	$\text{kg s}^{2} \text{A}^{1}$
インダクタンス	ヘンリー	Н	Wb/A	$m^2 kg s^2 A^2$
セルシウス温度	セルシウス度 ^(e)	°C		K
光東	ルーメン	lm	cd sr ^(c)	cd
照度	ルクス	lx	lm/m ²	m ⁻² cd
放射性核種の放射能 ^(f)	ベクレル ^(d)	Bq		s ⁻¹
吸収線量,比エネルギー分与,	ガレイ	Gv	J/kg	m ² e ⁻²
カーマ		Gy	ong	
線量当量,周辺線量当量,	シーベルト (g)	Sv	J/kg	$m^2 e^{-2}$
方向性線量当量,個人線量当量		50	5/Kg	III 8
酸素活性	カタール	kat		s ⁻¹ mol

酸素活性(1) ダール kat [s¹ mol]
 (w)SH接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはや コヒーレントではない。
 (h)ラジアンとステラジアンは数字の1に対する単位の特別な名称で、量についての情報をつたえるために使われる。 実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の1は明 示されない。
 (a)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。
 (d)へルツは周期現象についてのみ、ペラレルは放射性核種の統計的過程についてのみ使用される。 セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。それシウス度とケルビンの
 (a)やレシウス度はケルビンの特別な名称で、温度器や温度開隔を表す整備はどもらの単位で表しても同じである。
 (b)放射性核種の放射能(activity referred to a radionuclide) は、しばしば誤った用語で"radioactivity"と記される。
 (g)単位シーベルト(PV,2002,70,205) についてはCIPM物告2(CI-2002)を参照。

表4.単位の中に固有の名称と記号を含むSI組立単位の例

	S	[組立単位	
組立量	名称	記号	SI 基本単位による 表し方
粘度	パスカル秒	Pa s	m ⁻¹ kg s ⁻¹
カのモーメント	ニュートンメートル	N m	m ² kg s ⁻²
表 面 張 九	リニュートン毎メートル	N/m	kg s ⁻²
角 速 度	ラジアン毎秒	rad/s	m m ⁻¹ s ⁻¹ =s ⁻¹
角 加 速 度	ラジアン毎秒毎秒	rad/s^2	$m m^{-1} s^{-2} = s^{-2}$
熱流密度,放射照度	ワット毎平方メートル	W/m^2	kg s ⁻³
熱容量、エントロピー	ジュール毎ケルビン	J/K	$m^2 kg s^{2} K^{1}$
比熱容量, 比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	$m^{2} s^{2} K^{1}$
比エネルギー	ジュール毎キログラム	J/kg	$m^2 s^2$
熱伝導率	「ワット毎メートル毎ケルビン	W/(m K)	m kg s ⁻³ K ⁻¹
体積エネルギー	ジュール毎立方メートル	J/m ³	m ⁻¹ kg s ⁻²
電界の強さ	ボルト毎メートル	V/m	m kg s ⁻³ A ⁻¹
電 荷 密 度	クーロン毎立方メートル	C/m ³	m ⁻³ s A
表面電荷	「クーロン毎平方メートル	C/m ²	m ⁻² s A
電東密度, 電気変位	クーロン毎平方メートル	C/m ²	m ² s A
誘 電 辛	コアラド毎メートル	F/m	$m^{-3} kg^{-1} s^4 A^2$
透 磁 率	ペンリー毎メートル	H/m	m kg s ⁻² A ⁻²
モルエネルギー	ジュール毎モル	J/mol	$m^2 kg s^2 mol^1$
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	$m^2 kg s^{-2} K^{-1} mol^{-1}$
照射線量(X線及びγ線)	クーロン毎キログラム	C/kg	kg ⁻¹ s A
吸収線量率	ダレイ毎秒	Gy/s	$m^{2} s^{3}$
放 射 強 度	ワット毎ステラジアン	W/sr	$m^4 m^{-2} kg s^{-3} = m^2 kg s^{-3}$
放射輝度	ワット毎平方メートル毎ステラジアン	$W/(m^2 sr)$	m ² m ⁻² kg s ⁻³ =kg s ⁻³
酵素活性濃度	カタール毎立方メートル	kat/m ³	$m^{-3} s^{-1} mol$

表 5. SI 接頭語						
乗数	名称	記号	乗数	名称	記号	
10^{24}	э 9	Y	10 ⁻¹	デシ	d	
10^{21}	ゼタ	Z	10^{-2}	センチ	с	
10^{18}	エクサ	Е	10^{-3}	ミリ	m	
10^{15}	ペタ	Р	10^{-6}	マイクロ	μ	
10^{12}	テラ	Т	10^{-9}	ナノ	n	
10^{9}	ギガ	G	10^{-12}	ピコ	р	
10^{6}	メガ	М	10^{-15}	フェムト	f	
10^3	+ 1	k	10^{-18}	アト	а	
10^{2}	ヘクト	h	10^{-21}	ゼプト	z	
10^{1}	デカ	da	10^{-24}	ヨクト	v	

表6.SIに属さないが、SIと併用される単位					
名称	記号	SI 単位による値			
分	min	1 min=60 s			
時	h	1 h =60 min=3600 s			
日	d	1 d=24 h=86 400 s			
度	۰	1°=(π/180) rad			
分	,	1'=(1/60)°=(π/10 800) rad			
秒	"	1"=(1/60)'=(π/648 000) rad			
ヘクタール	ha	1 ha=1 hm ² =10 ⁴ m ²			
リットル	L, 1	1 L=1 l=1 dm ³ =10 ³ cm ³ =10 ⁻³ m ³			
トン	t	$1 t=10^3 kg$			

表7. SIに属さないが、SIと併用される単位で、SI単位で

表される数値が実験的に得られるもの					
名称 記号			記号	SI 単位で表される数値	
電子	ボル	ŀ	eV	1 eV=1.602 176 53(14)×10 ⁻¹⁹ J	
ダル	- F	\sim	Da	1 Da=1.660 538 86(28)×10 ⁻²⁷ kg	
統一原	子質量単	単位	u	1 u=1 Da	
天 文	単	位	ua	1 ua=1.495 978 706 91(6)×10 ¹¹ m	

表8. SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	1 bar=0.1MPa=100 kPa=10 ⁵ Pa
水銀柱ミリメートル	mmHg	1 mmHg≈133.322Pa
オングストローム	Å	1 Å=0.1nm=100pm=10 ⁻¹⁰ m
海 里	Μ	1 M=1852m
バーン	b	$1 \text{ b}=100 \text{ fm}^2=(10^{-12} \text{ cm})^2=10^{-28} \text{ m}^2$
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位しの粉結的な間径は
ベル	В	対数量の定義に依存。
デシベル	dB -	

表9. 固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値		
エルグ	erg	1 erg=10 ⁻⁷ J		
ダイン	dyn	1 dyn=10 ⁻⁵ N		
ポアズ	Р	1 P=1 dyn s cm ⁻² =0.1Pa s		
ストークス	St	$1 \text{ St} = 1 \text{ cm}^2 \text{ s}^{\cdot 1} = 10^{\cdot 4} \text{ m}^2 \text{ s}^{\cdot 1}$		
スチルブ	$^{\mathrm{sb}}$	$1 \text{ sb} = 1 \text{ cd cm}^{-2} = 10^4 \text{ cd m}^{-2}$		
フォト	ph	1 ph=1cd sr cm ⁻² =10 ⁴ lx		
ガ ル	Gal	1 Gal =1cm s ⁻² =10 ⁻² ms ⁻²		
マクスウエル	Mx	$1 \text{ Mx} = 1 \text{G cm}^2 = 10^{-8} \text{Wb}$		
ガウス	G	1 G =1Mx cm ⁻² =10 ⁻⁴ T		
エルステッド ^(a)	Oe	1 Oe ≙ (10 ³ /4 π)A m ⁻¹		
(a) 3元系のCGS単位系とSIでは直接比較できないため、等号「 ▲ 」				

は対応関係を示すものである。

		表	₹10.	SIに 帰	禹さないその他の単位の例
	名	称		記号	SI 単位で表される数値
キ	ユ	IJ	ſ	Ci	1 Ci=3.7×10 ¹⁰ Bq
$\scriptstyle u$	\sim	トゲ	\sim	R	$1 \text{ R} = 2.58 \times 10^{-4} \text{C/kg}$
ラ			ド	rad	1 rad=1cGy=10 ⁻² Gy
$\scriptstyle u$			Д	rem	1 rem=1 cSv=10 ⁻² Sv
ガ	3	/	7	γ	$1 \gamma = 1 \text{ nT} = 10^{-9} \text{T}$
フ	x	N	111		1フェルミ=1 fm=10 ⁻¹⁵ m
メー	ートルヌ	系カラ:	ット		1 メートル系カラット=0.2 g=2×10 ⁻⁴ kg
ŀ			N	Torr	1 Torr = (101 325/760) Pa
標	進っ	大気	圧	atm	1 atm = 101 325 Pa
カ	П	IJ	ļ	cal	1 cal=4.1858J(「15℃」カロリー), 4.1868J (「IT」カロリー), 4.184J(「熱化学」カロリー)
3	カ		\sim		$1 = 1 = 10^{-6} m$