

Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

PNNL-18911

Reconstruction of Dose to The Residents of Ozersk From The Operation of The Mayak Production Association: 1948-2002: Progress Report on Project 1.4

Y Mokrov SI Rovny LR Anspaugh BA Napier

October 2009



Proudly Operated by Battelle Since 1965

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes **any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.**

PACIFIC NORTHWEST NATIONAL LABORATORY operated by BATTELLE for the UNITED STATES DEPARTMENT OF ENERGY under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831-0062; ph: (865) 576-8401 fax: (865) 576-5728 email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161 ph: (800) 553-6847 fax: (703) 605-6900 email: orders@ntis.fedworld.gov online ordering: http://www.ntis.gov/ordering.htm



RECONSTRUCTION OF DOSE TO THE RESIDENTS OF OZERSK FROM THE OPERATION OF THE MAYAK PRODUCTION ASSOCIATION: 1948–2002

Submitted to the Office of International Health Studies, U.S. Department of Energy for the US–Russia Joint Coordinating Committee on Radiation Effects Research

Progress Report on Project 1.4

Principal Investigators:

For Russia: Yuri G. Mokrov Sergey I. Rovny

For the United States: Lynn R. Anspaugh and Bruce A. Napier

Participating Institutions:

For Russia: Mayak Production Association

For the United States: University of Utah, Salt Lake City, UT Pacific Northwest National Laboratory, Richland, WA

TABLE OF CONTENTS

Introduction	1
Tasks Within Project 1.4	1
Progress During the Reporting Period.	7
Task 2.4 (Milestone 4)	7
Tasks 8.3 and 9.4 (Milestone 12)	8
Task 12 (Milestone 11)	9
Task 11.1 (Milestone 10)	9
Milestone Status	10
Other Activities	12
Acknowledgements	12

International Copyright © 2008 Mayak Production Association All rights reserved.

INTRODUCTION

In keeping with the request of November 2008 of the members of the American Scientific Review Group (AmSRG), this Progress Report for Project 1.4 of the U.S.– Russia Joint Coordinating Committee on Radiation Effects Research continues in the abbreviated format of providing details only on the work accomplished during this sixmonth reporting period.

TASKS WITHIN PROJECT 1.4

Task 2: Atmospheric transport

Task 2.4. Chemical/physical form of atmospheric iodine and determination of deposition-rate parameters (L. Anspaugh). Specific physical and chemical forms of radioactive iodine are generally referenced in the literature as being present in the atmosphere due to releases from operational or incidental releases: iodine bound with aerosol particles, elemental iodine gas, and organic gaseous iodine compounds. The forms of iodine may have changed after installation of different processing techniques or cleanup devices. Available data from similar facilities (Hanford, Sellafield) will be acquired and evaluated. Natural analogues will also be considered. Mayak stack measurements will be reviewed. The results of environmental monitoring will be compared with model output, including wet deposition, to see if the results support selection of one or another chemical form. The results of this task will support Subtasks 2.5 and 2.6, which feed Milestone 12. The results will be reported in Milestone 4.

Work on this task has been completed. See discussion below.

Task 7: Uptake from mother's milk

Task 7.1. Mother's milk consumption rates (L. Anspaugh/B. Napier). Referenceconsumption rates for breast-feeding children as a function of age and associated intake of other foods will be developed. The model(s) and parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 7.2. Transfer from mother 's diet to breast milk (L. Anspaugh/B. Napier). Transfer factors for radionuclides from mothers to breast milk will be evaluated based on recent literature publications. The model(s) and parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 8. Calculation of dose from ingestion

Task 8.1. Ingestion dose coefficients (L. Anspaugh/B. Napier). The most recent source of age-dependent ingestion dose coefficients, the ICRP CD-ROM system, will be reviewed and values accepted if appropriate; uncertainty ranges will be applied to the values. The parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 8.2. Pre-natal dose coefficients (L. Anspaugh/B. Napier). The ICRP dose coefficients for unborn persons for ¹³¹I will be reviewed, compared with the current literature, and appropriate values selected with uncertainty ranges applied. The parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 8.3. Calculation of dose to representative persons (Y. Mokrov). Ingestion doses, with uncertainties, will be estimated for reference individuals of various ages, sex, lifestyle, and dietary habits. The results will be reported in Milestone 12.

Work on this task is underway. See discussion in following section on current progress.

Task 9. Calculation of dose from inhalation

Task 9.1. Inhalation-dose coefficients (L. Anspaugh/B. Napier). The most recent source of age-dependent inhalation-dose coefficients, the ICRP CD-ROM), will be reviewed and the values accepted, if appropriate for iodine in several chemical/physical forms, with uncertainty ranges applied to the values. The parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 9.2. Inhalation rates (L. Anspaugh/B. Napier). ICRP Publication 66 introduced standard breathing rates for six age groups in the context of four types of physical activities: sleeping, wakeful rest, light physical activity, and heavy physical activity. A series of reference scenarios will be developed for breathing rates; uncertainty ranges will be applied to the values. The parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 9.3. Evaluation of indoor air concentrations (L. Anspaugh/B. Napier). The radionuclide concentration in air inside any building may be lower by a factor of a few than the concentration outside a building. Monthly average indoor-to-outdoor ratios will

be developed. The model(s) and parameterizations selected, with their uncertainties, will be used in Milestone 12.

Work on this task has been completed. See discussion below.

Task 9.4. Calculation of dose to representative persons (Y. Mokrov). Inhalation doses, with uncertainties, will be estimated for reference individuals with various ages, sex, lifestyles, and activity habits. The results will be reported in Milestone 12.

Work on this task is underway. See discussion in following section on current progress.

Task 11: Doses from non-iodine radionuclides

Task 11.1. Internal doses from non-iodine radionuclides (Y. Mokrov). Human-bodyburden data available from the Southern Urals Biophysics Institute for residents of Ozersk will be used to develop screening estimates of internal dose. It is anticipated that the doses will be a sufficiently small fraction of the dose from ¹³¹I so that more intensive methods will not be required. This determination will be justified and documented in Milestone 9.

Work on this subtask is nearly completed. See discussion below.

Task 12: Validation using monitoring data

Task 12.1. Evaluation of monitored air concentrations (P. Stukalov). The Mayak archives will be searched and information related to concentrations of ¹³¹I measured in the atmosphere at various locations and times will be collected. This information will support the activities of Task 12.7 and be reported in Milestone 11.

Work on this task is underway. See discussion below for recent progress.

Task 12.2. Evaluation of monitored vegetation concentrations (P. Stukalov). The Mayak archives will be searched and information related to concentrations of 131 I measured in vegetation at various locations and times will be collected. This information will support the activities of Task 12.7 and be reported in Milestone 11.

Work on this task is underway. See discussion below for recent progress.

Task 12.3. Evaluation of monitored concentrations in milk and milk products (P. Stukalov). The Mayak archives will be searched and information related to concentrations of ¹³¹I measured in milk and milk products at various locations and times will be collected. This information will support the activities of Task 12.7 and be reported in Milestone 11.

Work on this task is underway. See discussion below for recent progress.

Task 12.4. Evaluation of monitored soil concentrations (P. Stukalov). The Mayak archives will be searched and information related to concentrations of ¹³¹I measured in soil at various locations and times will be collected. This information will support the activities of Task 12.7 and be reported in Milestone 11.

Work on this task is underway.

Task 12.5. Evaluation of monitored external exposure rates (P. Stukalov). The Mayak archives will be searched and information related to monitored external exposure rates at various locations and times will be collected. This information will support the activities of Task 12.7 and be reported in Milestone 11.

Work on this task is underway.

Task 12.6. Evaluation of monitored concentrations in animal thyroids (P. Stukalov). The Mayak archives will be searched and information related to concentrations of ^{131}I measured in cow thyroids at various locations and times will be collected. This information will support the activities of Task 12.7 and be reported in Milestone 11.

Work on this task is underway.

Task 12.7. Comparison of monitoring data to calculations (Y. Mokrov). The data collected in Tasks 12.1 through 12.6 will be compared against the concentrations calculated using modeling results. Decision criteria will be developed to determine adequate verification and validation of the calculated results. The comparisons and conclusions will be presented in Milestone 11.

Work on this task is underway.

Task 14: Uncertainty support

Task 14.2: Analysis of parameter variability (O. Alexandrova). The statistical processing and evaluation staff will work with all other task leaders to define and develop uncertainty distributions around key model-input parameters. This task provides assistance and statistical direction to all tasks of the project.

Work on this task is underway.

Task 14.3: Analysis of model uncertainty (O. Alexandrova). The statistical processing and evaluation staff will work with all other task leaders to understand the implications of selection of certain models over others, and the impact on the overall uncertainty of the results. This task provides assistance and statistical direction to all tasks of the project.

Work on this task is underway.

Task 14.4: Support for other tasks (O. Alexandrova). The statistical processing and evaluation staff will work with all other task leaders as needed to provide advice and consultation on issues regarding uncertainty. This task provides assistance and statistical direction to all tasks of the project.

Work on this task is underway.

PROGRESS ON TASKS DURING THE REPORTING PERIOD

During the reporting period, Mayak experts performed the following work:

Task 2.4 (Milestone 4)

Work was completed on the reconstruction of I-131 formation in technological areas of the radiochemical plant and atmospheric releases of different species of I-131 (aerosol, vapor, and organic forms). The summarized results are presented below.

	Iodine speciation, % by volume			
Years	aerosol	vapor-gas phase (molecular iodine)	organic	
1948 - 1952	50-80	1-35	0.5-30	
	(65)*	(20)	(15)	
1953-1963	60-80	1-25	0.5-30	
	(70)	(15)	(15)	
1963-1976	60-75	1-10	20-30	
	(70)	(5)	(25)	

I-131 Release Speciation at the B Plant

• Here and below values that are used for further calculations are given in parentheses

I-131 Release Speciation at the DB Plant

	Iodine speciation, % by volume			
Years	aerosol	vapor-gas phase (molecular iodine)	organic	
1959 - 1960	50-80	1-35	0.5-30	
	(65)	(20)	(15)	
1961-1968	60-80	1-20	0.5-30	
	(70)	(12)	(18)	
1969-1987	60-95	0.5-5	0.5-30	
	(80)	(5)	(15)	

Detailed description of the reconstruction results is given by I.A. Istomin, *Reconstruction of I-131 Release Speciation in Main Technological Areas of the Mayak PA in the period 1948 -1987*, which is currently undergoing the Mayak clearance process at the Rosatom security agency.

Tasks 8.3 and 9.4 (Milestone 12)

Based on the studies undertaken to reconstruct I-131 speciation in the composition of releases from the radiochemical plant (IA Istomin, discussed under Task 2.4), input data files for the atmospheric model were refined; several calculation options were prepared to account for uncertainty in the source term (parameters of the release source) and in the percentage of various species of radioiodine. Operation of the computer modules used for I-131 atmospheric transport calculation, radioiodine distribution in the food chain, and internal dose calculations is being coordinated.

A new report by Rovney et al. *Methods for calculating thyroid doses to the residents* of Ozersk due to ¹³¹I releases from the stacks of the Mayak Production Association (being distributed with this progress report) describes special software that was developed which contains a database of reference data by age, various diets, dose factors, a database of input data, and a database of calculated output data for each exposure pathway (inhalation, ingestion, and external). The primary functions of the software include:

- Import of files with daily values of the RATCHET-calculated surface concentrations and depositions to a database electronic table. This is designed for future use, when coordinates of a required receptor point are specified, corresponding time series of data will be chosen from the table and tables of input data will be formed;
- Determination of daily activity accumulation in compartments of the radioiodine migration model soil surface layer, soil root layer, outer and inner parts of plants. The results are transmitted to the corresponding table of the database;
- Calculation of external absorbed doses due to immersion in the cloud and due to deposition on the soil. The results are transmitted to the corresponding tables of the database;
- Calculation of absorbed doses due to ¹³¹I inhalation. The results are transmitted to the corresponding table of the database;
- Calculation of absorbed thyroid doses due to consumption of foodstuffs containing ¹³¹I. The results may be aggregated into a unified table, or data may be provided for each type of foodstuffs.

Preliminary estimates of equivalent internal doses to thyroid from releases of ¹³¹I were obtained for different age groups of Ozersk population:

- dose due to inhalation– 7-10 mSv;
- dose from all pathways (cow's milk, vegetables, etc.) 1-2 Sv;
- dose to a limited fraction of children population (born in 1948-1954) who consumed goat's milk from Metlino village (located in the vicinity of the Techa River) on a regular basis– 5-7 Sv.

This report includes an interim selection of some parameters, final selection of which is

described in the report *Minor Parameters Needed for Individual-Dose Calculations* by LR Anspaugh and BA Napier, which is being distributed with this progress report. Future calculations will be made with the updated parameter selections.

Populated area	External radiation	Dose to thyroid ⁽¹⁾	
	dose	Inhalation	Ingestion (milk,
			vegetables, etc.)
Ozersk	$0.01 - 0.015^{(2)}$	0.004 - 0.01	0.5 - 1.5
Metlino	0.10	0.02 - 0.04	3 - 6
Novogorny	0.020	0.015 - 0.025	2-4
Kasli / Kyshtym	0.005	0.003 - 0.005	0.3 - 0.8
Production site	0.5	~ 0.2	-

Preliminary calculated estimates of radiation doses to the population (1948-2002), Sv

⁽¹⁾ for different age groups of the population

⁽²⁾ for different districts of the town

Development of the software for visualizing the calculation results is underway.

Preliminary results of dose calculations for the population were provided by Yu.G. Mokrov, D.A. Beregich, P.M. Stukalov, I.A. Ivanov, I.I. Teplyakov, S.I. Rovny, L.R. Anspaugh, and B.A. Napier, *The Results of JCCRER Project 1.4: Reconstruction of Dose for the Residents of Ozersk from the Operation of the Mayak Production Association: 1948-2002*, presented at the JCCRER Meeting in Washington, D.C., in September 2009.

Task 12 (Milestone 11)

The period from 1960 to 1965 was chosen for verification of calculated doses due to iodine-131 on the basis of monitoring data. During this period of time, as a result of reduction in the holdup time of uranium blocks delivered for reprocessing to the DB plant, there was an increase of radioiodine in the technological process, which caused higher radioiodine content in the ground-level atmosphere of the Mayak PA buffer area and monitoring area.

This period of time is characterized by:

- availability of quite reliable experimental and calculated data on iodine-131 release rate;
- large amounts of experimental data related to contamination of air, soil, vegetation and milk with radioactive iodine.

Preliminary estimates demonstrate satisfactory agreement between the calculated and experimental results.

Task 11.1 (Milestone 10)

For retrospective reconstruction of doses to the residents of Ozersk due to ingestion of Sr-90 and Cs-137 from 1951 to 1975, data were collected on contamination of the foodstuffs and the territory and aerosol fallouts for these radionuclides, as well as on the dietary habits of the town residents during the period under study.

Generalization was made of numerical parameters of contamination of agricultural products versus contamination of the territory. Processing and analysis of the collected data demonstrated that doses due to ingestion of Sr-90 and Cs-137 during 1959 (the year that is characterized by the maximum doses following the tank farm explosion accident) were reduced by no less than a factor of 1.5-2 as a result of measures taken to eliminate or reduce consumption of agricultural products from the contaminated territories.

Radiation doses due to ingestion of Sr-90 and Cs-137 in the subsequent years were caused primarily by contemporaneous contamination of collective and individual farming lands. The most significant reason for formation of such doses was consumption of agricultural products grown at the collective farmlands located in the proximity of the production site, as well as foodstuffs produced in the territory of Ozersk by individual farmers involved in animal husbandry and poultry breeding. Contamination of these food products, as well as of the territories used for their production, was caused mainly by the ongoing releases from the Mayak PA during the entire period considered.

In the subsequent years of the 1960s through 1980s, reduction in the internal radiation doses to the residents of Ozersk due to ingestion of such radionuclides as Sr-90 and Cs-137 to 0.05-0.1 mSv/year in the 1990s, is caused by the decrease in the amount of airborne releases from the facility and of radioactive fallout in the adjacent territories.

Preliminary estimates of internal radiation doses due to intake of radionuclides other than iodine-131 with foodstuffs demonstrated that maximum doses occurred from 1950 to 1954 and were on the order of 10-30 mSv for the critical population groups.

The results of the study of foodstuffs contamination and internal radiation exposure of the residents of Ozersk will be provided in a report *Reconstruction of internal radiation doses to the residents of Ozersk due to ingestion of* ⁹⁰Sr and ¹³⁷Cs, by Yu.G. Mokrov, A.I. Alexakhin, and I.A. Ivanov, which has been prepared and is currently undergoing the Mayak clearance process at the Rosatom security agency.

MILESTONE STATUS

All the activities under this project including production of the final report are scheduled for completion in the first half of year 2010.

1. Monthly iodine deliveries to reprocessing plants.

This milestone has been completed through publication of *Reconstruction of the Rate* of Atmospheric Releases of Noble Radioactive Gases and Assessment of Iodine-131 Accumulation in Irradiated Uranium at Mayak Reactor Facilities in 1948-1965 - Part I, Calculation Method Development and Justification, and Part 2, Calculated Results, distributed in October 2006, and Reconstruction of Atmospheric Releases of I-131 from Mayak Radiochemical Plant Stacks for the Period from 1948 to 1970, Part 1: Improvements in the Calculation Methods for the Determination of I-131 Delivery to the Radiochemical Plant distributed in October 2008.

2. Modeling atmospheric transport for releases from the MPA

This milestone has been completed through publication of Assessment of data from weather stations located near Mayak PA for atmospheric dispersion calculations

(weather stations at Verkhny Ufaley, Nyazepetrovsk, Brodokalmak, Kunashak, Argayash, Chelyabinsk, ESRS, and Mayak PA) and Selection of Atmospheric Transport Model and Transfer and Installation of the Selected Atmospheric Dispersion Model to MPA in April 2006.

3. Description of models used for estimation of dynamic concentrations of ¹³¹I in terrestrial plants and animals

This milestone has been completed through publication of *Determination of* quantitative parameters for the Iodine-131 model for transfer to vegetation, farm animal products, and agricultural products in April 2006.

4. Evaluation of chemical forms of ¹³¹I involved in atmospheric transport

This milestone has been completed through publication of a supporting report, provided as an Appendix to the April 2006, and by *Evaluation of chemical forms of* ¹³¹*I involved in atmospheric transport* in October 2008.

5. Releases of noble gases to the atmosphere from reactor operations

This milestone has been completed through publication of *Reconstruction of the Rate* of Atmospheric Releases of Noble Radioactive Gases and Assessment of Iodine-131 Accumulation in Irradiated Uranium at Mayak Reactor Facilities in 1948-1965 - Part I, Calculation Method Development and Justification, and Part 2, Calculated Results in October 2006 and Milestone 10 Part 2, Reconstruction Of External Dose Caused By Atmospheric Releases Of Noble Radioactive Gases From The Stacks Of The Mayak Reactors In 1948–1989 in April 2009.

6. Analysis of age-dependent food consumption patterns in Ozersk

This milestone has been completed through publication of *Changes in Population* Food Ration and Demographic Parameters for Ozersk In 1948–2002 in October 2008, which was transmitted in April 2009.

7. Releases of 131 I to the atmosphere from reprocessing plants

This milestone has been completed through publication of *Reconstruction of*¹³¹*I* Atmospheric Releases from the Stacks of the Mayak Radiochemical Plant in 1948-1970, Part II, Results of the Reconstruction of ¹³¹*I* Releases from the Stacks of the Reactor and Radiochemical Plants in October 2008.

8. Analysis of food production and distribution networks

This milestone has been completed through publication of *Production and Delivery of Food to Ozersk* in October 2007.

9. Evaluation of 129 I as a tracer for 131 I

This milestone has been completed through publication of *Potential for I-129* Application as a Tracer Element for Assessment of Internal Dose to Thyroid for Residents of the Territories Contaminated with Iodine-131 in October 2008.

10. Screening dose estimates for non-iodine radionuclides

This milestone is nearly completed, through publication of Screening Calculation of External Doses to the Residents of Ozersk (Not Associated with Releases of Noble Gases

and Iodine -131) in October 2008 and a second report *Reconstruction of internal* radiation doses to the residents of Ozersk due to ingestion of ^{90}Sr and ^{137}Cs in Rosatom review as discussed above.

11. Validation of ¹³¹I environmental transport modeling results with historical monitoring data

Work is underway as discussed above with a report in preparation. This report is also supported by the report *Methods for calculating thyroid doses to the residents of Ozersk due to*¹³¹*I releases from the stacks of the Mayak Production Association* being distributed with this progress report.

12. Representative doses for Ozersk residents

Scheduled for October 2008 but delayed due to budget restrictions. Initial work leading to this milestone is described in the report *Methods for calculating thyroid doses to the residents of Ozersk due to* ¹³¹*I releases from the stacks of the Mayak Production Association* and in the report *Minor Parameters Needed for Individual-Dose Calculations*, both of which are being distributed with this progress report, and in the presentation made at the JCCRER meeting in Washington, D.C., *The Results of JCCRER Project 1.4: Reconstruction of Dose for the Residents of Ozersk from the Operation of the Mayak Production: 1948-2002*. This report has been rescheduled for mid-2010.

OTHER ACTIVITIES

The US collaborators had planned to meet with the Russian team in Chelyabinsk in October. However, due to a communication breakdown (apparently caused by mass August vacations at Mayak) a new Rosatom regulation was not fulfilled requiring at least 45-day notice to security officials prior to their meeting with foreign nationals (e.g., the US team). As a result, our meeting was cancelled, causing substantial frustration. We held discussions by telephone and email, and believe that the project is progressing reasonably well, but transmittal of milestone reports to the US collaborators has encountered substantial delays caused by the Rosatom review requirements. Yuri Mokrov has promised receipt of final documentation in early 2010.

ACKNOWLEDGEMENTS

Work on the study is being funded by the US Department of Energy's Office of International Health Studies, and has had the general support and cooperation of the Federal Agency for Atomic Energy of the Russian Federation. The work on this project was performed under the auspices of the US–Russia Joint Coordinating Committee on Radiation Effects Research.