Risk assessment for human health and terrestrial ecosystem under chronic radioactive pollution near regional radioactive waste storage

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Abstract. An impact of the radioactive waste storage facility at the regional population was assessed under supervision of IAEA. It was made in accordance with the methodology for assessment of doses and risks to human storage using different scenarios of radionuclides releases into the environment. The following scenarios were considered: leakage of fluid, resuspension of dust, fire, flooding. Thy evaluation of radiation doses received and the risks to the human showed that the risk has been acceptable for all scenarios. An approach for an ecological risk assessment for terrestrial ecosystem is presented as five modules: selection of the ecosystem-receptor of radiation effects; determination of reference species of living organisms and their survival indices; the critical load as an absorbed dose rate is calculated from the dependence between the absorbed Sr-90 radiation dose rate and the coefficient of radioactive strontium accumulation in mollusc shells; the critical dose; risk is assessed from a part of the ecosystem territory with increased mollusc loading; uncertainties appeared at each stage of risk assessment are characterized. The risk of exposure to the repository on the ecosystem should be characterized as unacceptable.

1. Introduction

Much attention is being given to optimal human-environment relationships under the conditions of ever-increasing anthropogenic impact on the biosphere.

Thus, the developed sanitary-hygienic principles of rationing are based on an anthropocentric approach which considers a human impact of technogenic factors and is aimed at limiting pollutant intake. The anthropocentric principle of rationing was formulated by the International Commission for Radiological Protection (ICRP) in 1970 and confirmed in 1990 [1, 2]. However in 2007 ICRP [3] had stated that in developing the strategy of environmental radiation safety, it is necessary to give the evidence for protecting not only people and but also living organisms; it reflects the ecocentric principle of radiation factor rationing. Therefore many recent papers have developed the ecological approaches to rationing of radiation effects on ecosystems [4-6].

In this paper are presented the results of the risk assessment for human health and for terrestrial ecosystem under chronic radioactive pollution near regional radioactive waste storage. The territory near a regional radioactive waste storage created in Kaluga region has been chosen as a study area. This object had been put into operation in the 50-70s of the last century. Now the object is a closure.
In 1998-1999 the increased specific activity of Sr-90 had been revealed in the surveying wells and in the following this was stipulated by surface water penetration in one of the facility. At reservoir overfilling, Sr-90 and Cs-137 radionuclide escaped outside. Multi-year studies have showed that Sr-90 contributes much to radioactive contamination on the territory near the storage and in the adjacent territory [7].

2. Materials and methods

2.1. Risk assessment for human health

An impact of the radioactive waste storage facility at the regional population was assessed under [2], supervision of IAEA [8].

It was made in accordance with the methodology for assessment of doses and risks to the human from the effects of storage using different scenarios of radionuclides into the environment. The following scenarios were considered: leakage of fluid, resuspension of dust, fire, flooding. Calculations were carried out with the mathematical models describing selected script.

Radiation risk assessment carried out by calculation of radiation exposure for the population at different ways of radionuclides in the human body. The integral radiation risk factor of malignancy and hereditary health effects was used for the calculations. The magnitude of the risk factor for a uniform irradiation of the whole body is 0.057 Sv⁻¹ [9].

2.2. Risk assessment for terrestrial ecosystem

The ecological risk assessment for terrestrial ecosystem is presented as five modules: 1) selection of the ecosystem-receptor of radiation effects; 2) determination of reference species of living organisms and their survival indices; 3) the critical load as an absorbed dose rate is calculated from the dependence between the absorbed Sr-90 radiation dose rate and the coefficient of radioactive strontium accumulation in mollusc shells; 4) the critical dose; risk is assessed from a part of the ecosystem territory with increased mollusc loading; 5) uncertainties appeared at each stage of risk assessment are characterized.

2.3. Soil sample analysis

Soil samples are collected by a special sampler with a set of Edelman hand soil drills (Eijkelkamp Firm, Netherlands). Soil sample preparation for assessing Sr-90 content had been realized according to the [10].

Soil mass of 1 kg was counter balance weighed and uniformly distributed on a stainless steel griddle in the layer of 20-30 mm in thickness. To dehumidify, the griddle with samples had been kept in a drying box at 110 °C during 5-7 hours. Dried samples were disintegrated and mixed in a mortar to the grain size of 2-3 mm. Then the mean average sample (mass 50-300 g) of air-dried soil was analytical balance weighed. The sample had been annealed in a muffle furnace at 700 – 750 °C to transform the basic mass into oxide. Strontium carrier solution (50 mg of metal) and 5 M HNO₃ of 200-300 cm³ in volume were added in a glass with annealed soil. The soil/solution mixture was boiled in a sand-bath during half an hour. A solution was separated from a solid phase by filtration. Solid soil was twice treated by boiling in 4-5 M HNO₃ during half an hour. After filtration the solid soil sediment had been washed three times with distilled water in a filter. After filtration the solutions were combined and evaporated to 100 cm³.

A version of oxalate precipitation was used to separate strontium from other radionuclides and chemical elements and to concentrate it. Ethanedioic acid solution (95 g/l H₂C₂O₄) in the quantity of about 100 cm³ was added to the solution obtained after desalinization and evaporation. pH=4 in a solution was set by deacidification with an aqua ammonium solution. The solution with an oxalate precipitate was stayed for 4 hours, further a precipitate was elutriated and washed twice with hot distilled water in a filter. A precipitate and a filter were dried and then annealed at 750°C in a muffle furnace during an hour. Hereafter a precipitate was put into a glass and dissolved in 3-4 M HNO₃, the solution acidity was set at 3.0 M/l (the solution volume is preferably made up to 5-10 cm³). When the
accompanying components were separated, Sr-90 was cleaned by extraction. Nitric-acid solution was put in a separating funnel of 50-100 cm$^3$, poured with chloroform in a 2:1 ratio of the aqueous and organic phases, shaked up and phased. After phase separation, the aqueous solution was added by 5-10 cm$^3$ dicyclohexane-18-crown-6 in 0.2 M of a chloroform solution and extracted by shaking up during 30 s. The organic phase was washed with two portions of 2M HNO$_3$ solution in a 1:1 ratio; the aqueous phase was rejected. The extract (organic phase) or its aliquot part was put on a target and dried under an infrared lamp. Dry matter $\beta$-activity on the target was measured.

2.4. Plant sample analysis

Plant – great nettle (Urtica dióica) - samples were collected at the same sites (Figure 1). Before analysis, fresh crude plant samples were washed from dust and soil first with tap water, then with distilled water. Then the plant mass was dried up in desiccators at 80 – 85°C. A dried plant sample was milled. The optimal size of plant fragments had to be about 1-5 mm. Plant mineralization was realized by dry ashing. After digestion a sample had undergone acid treatment according to the above procedure [10].

2.5. Mollusc sample analysis

Terrestrial molluscs Bradybaena fruticum were collected from plants (great nettle (Urtica dióica)) and soil under plants at the tested sites. At least 25 specimens were chosen in each mollusc sample. First soft tissues were removed from shells. Shells were grinded in a mortar, hereafter the direct Sr-90 activity measurements were performed. Samples had been incinerated in a muffle furnace at 450°C during 6 hours. Radiochemical separation and Sr-90 content assessment were realized according to the above procedure.

2.6. Calculation of annual exposure doses for molluscs and statistical data

The annual doses of external $\beta$-exposure of mollusc tissues due to 90Sr and it daughter 90Y radionuclide contained in shells were calculated with software package ERICA Tool.

Statistical data processing was realized with R software programs [11].

3. Results and discussion

3.1. Risk assessment for human health

Assessment of the impact of regional radioactive waste storage facility was carried out taking into account the environmental monitoring data, including specific activities of artificial radionuclides in soil, vegetation, groundwater and surface waters [7].

It was used different scenarios of radionuclides into the environment in according with the methodology for assessment of doses and risks to the public from the effects of storage.

In the analysis of the data taken into account the recommendations of IAEA, which indicated that the radioactive waste storage facility should be designed so that the average dose to the critical group does not exceed the dose limit of 0.3 mSv/year, which corresponds to the level of risk of $10^{-5}$ per year [12].

The results obtained by calculation of total radiation doze from multiple paths of radionuclides in the human body for scenario «leakage of fluid» have shown the following. Total radiation dose from multiple paths of radionuclides in the human body is well below the regulated limit dose and up to Cs-137 – 1.20E-08 Sv/year and for Sr-90 – 4.66E-08 Sv/year. The main contribution to the dose for the population brings the oral route of radionuclides into the body with food.

Cs-137 is dose-forming radionuclide for the scenario "resuspension of dust" with the storage trench type. Inhalation and food intake are the main routes of public exposure. Radiation doze for the population is much less than 0.3 mSv/year: for Cs-137 is 5.8E-13 Sv/year, for Sr-90 is 1.7E-13 Sv/year.
Sr-90 is dose-forming radionuclide and the oral route is major public exposure for the scenario "flooding".

In the scenario "fire" in a trench type storage total radiation dose is determined by the radionuclide Sr-90 (75%). The inhalation and food intake are the main pathways of exposure. The total dose was 2.7E-12 Sv/year.

Radiation risk of occurrence of genotoxic effects for public health are significantly lower than the acceptable risk to the public (10^{-6}) (table 1).

| Scenario            | Risk of Cs-137 irradiation | Risk of Sr-90 irradiation | Total risk |
|---------------------|----------------------------|----------------------------|------------|
| Leakage of fluid    | 6.9E-10                    | 2.7E-09                    | 3.39E-09   |
| Resuspension of dust| 3.3E-14                    | 9.7E-15                    | 4.27E-14   |
| Flooding            | 2.8E-20                    | 1.9E-16                    | 1.90E-16   |
| Fire                | 3.5E-14                    | 1.2E-13                    | 1.55E-13   |

It is worth noting that the greatest interest is the scenario of «leakage of fluid», since it is such a scenario corresponds to an accident situation in the storage. In this case the total risk from multiple ways of intake of radionuclides was for Cs-137 is 6.9E-10, and for Sr-90 is 2.7E-09. The main contribution to the overall risk for the population brings a way of intake of radionuclides with food.

3.2. Risk assessment for terrestrial ecosystem

Radiation and chemical monitoring of the storage territory has been carried out to determine the degree of biotope pollution. The specific activity of natural (Ra-226, Th-232, K-40) and technogenic (Sr-90, Cs-137) radionuclides in soil samples collected in the storage territory and cross-border regions has shown that artificial radionuclides make a major contribution to the radioecological situation. In addition, the specific activity of natural radionuclides is compared with the background ones, therefore their contribution to radioactive pollution is insignificant.

The concentration of heavy metals Mn, Zn, Ni, Cu, Cr, Co, Cd, Pb, Fe in the samples studied does not exceed the value of maximum allowable concentration, therefore chemical pollution is of no concern in terms of priority pollution.

Identification of reference species and indices most properly reflecting the degree and the character of pollution in a studied territory is the next stage to assess a biotope of the radioactive waste storage. The terrestrial mollusc (Bradybaena fruticum) has been chosen as a reference species; it turned out to be rather sensitive to radioactive pollution in the territory studied. It should be noted that freshwater and terrestrial molluscs are being profitably employed as test-objects of ecosystem Sr-90 pollution for a long time. It is shown that practically the whole Sr-90 content is concentrated in mollusc shells and its activity is governed primarily by the level of territory pollution and does not undergo systematic long-term, seasonal and taxonomical changes. It is also worthy of note that the choice of similar reference species is supported by other researchers [13-15]. Thus, it is assumed that terrestrial molluscs and their response (lowing Sr-90 accumulation in shells) should be considered as reference species for the terrestrial ecosystem polluted by radionuclides.

Critical radionuclide loads on the reference species and indices have to be determined for assessing the ecological risk. In this case by the critical load we shall basically mean such radionuclide ingress into the ecosystem which does not cause irreversible alterations in its biogeochemical structure, biodiversity and productivity during a long period. A method has been applied to reveal the critical loads by plotting “dose-effect” dependencies and to analyze them [16]. To assess the storage impact on terrestrial molluscs from experimental data, the dose dependences with respect to a load gradient have been plotted. In this case the maximum permissible load is the critical point on a dose-effect
curve which connects the input loads and variations in the coefficient of radioactive strontium accumulation in shells.

![Figure 1. Accumulation coefficients of Sr-90 of mollusc shells versus radioactivity aggregation index.](image)

The dose dependence has been analyzed to find the site with such an aggregation index value at which a 10-fold decrease in the accumulation coefficient of Sr-90 (critical load) takes place; this analysis allowed one to obtain the critical aggregation index value (18.3) corresponding to the critical coefficient value (3.99) of radionuclide accumulation (in this case, the higher the AC values, the better is for ecosystem). The excess critical value of Sr-90 accumulation coefficient causes alterations in the life-sustaining activity of molluscs, decreased activity of their nutrition and, hence, changes in the need for shell building materials and Sr-90 is proved to be one of them (Figure 1).

To characterize the ecological risk from critical loads, GIS maps for the ecosystem studied have been plotted; these maps are used for determining excess critical loads. A space satellite (Google Company) image with a co-ordinate reference has been used as the GIS basis. All data on the ecosystem load as well the critical load are plotted on the digitized map using R program [R development, 2010]. The total territory area according to chosen co-ordinates is 0.54 ha. A part (%) of the area with excess critical loads is recognized by the software. A 95% ecosystem protection when the area with excess critical loads does not exceed 5% of the total value (0.54 ha) is taken as the permissible adverse storage impact on the environment [16]. The analyzed plotted GIS maps show that in a studied territory the area characterized by excess the critical AC is 61% (that exceeds the permissible value (5%)). So, the risk of waste storage effects on a surrounding terrestrial ecosystem should be characterized as inadmissible that highlights the unstable state in the territory studied within the next few years.

When assessing the ecological risk, some uncertainties have to be considered in such investigations. First, interspecies extrapolation, i.e. data for radionuclide effect on individual reference species have been extrapolated to the whole ecosystem. Second, in the critical load assessment the form of radionuclides found in soil has not been taken into account at the stage of data collection. Third, the process of radionuclide return into soil as a result of plant die-away and mollusc loss has not been considered.

4. Conclusions
Risk assessment for the population and the ecosystem allows us to conclude the following.

   Radiation doses and risks for the population, or habitually resident in the territory adjacent to the waste repository show that the radioactive storage is "relatively" safe target for the population.

   Hence, the environmental impact of this storage should be characterized as inadmissible and implies unstable conditions and the expected loss of current biocenosis in the studied territory in the near future.

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References
[1] ICRP Publication 26. The 1977 Recommendations of the International Commission on Radiological Protection. Ann.ICRP1977. V.1, No.3. p.1-53.
[2] ICRP Publication 60. The 1990 Recommendations of the international Commission on Radiological Protection. Ann.ICRP 1991.V.21. p.1-201.
[3] ICRP Publication 103. The 2007 Recommendations of the International Commission on Radiological Protection. Ann ICRP. 2007. V.37. No. 2-4, p. 1-332.
[4] Alexakhin, R., Fesenko, S. 2004 Radiation biology. Radiology. V.44, No.1, p.93-103.
[5] Ecological Risk Assessment 2007 \ edited by Glenn W. Suter II. – 2nd edd.(ISBN-10: 1-56670-634-3).
[6] Biya E A, Kazakov S V, Linge I I 2003 Development of ecological approaches to regulating radiation effects on aquatic ecosystems. IBRAE preprint. M.: RAS Institute of safe development problems of nuclear power engineering, 24 pp.
[7] Lavrentyeva G V 2014 Journal of Environmental Radioactivity № 135, pp. 128-134
[8] IAEA-TECDOC-1380. 2003 Derivation of activity limits for the disposal of radioactive waste in near surface disposal facilities, 145 p.
[9] RSS: Radiation Safety Standards. RSS - 99/2009, 2009. Moscow. (in Russian).
[10] Instruction : Procedure of radiochemical strontium-90 assessment in soil and plant samples, 1994. Instruction of Rosleskhoz of Russia from 05.09.1994 № 192. Moscow (in Russian).
[11] www.r-project.org
[12] IAEA – International Atomic Energy Agency. The Principles of Radioactive Waste Management, Safety Series No.111-F, IAEA, Vienna, 1995.
[13] Gudkov D I, Derevets V V, Zub L N 2005 Rodioecology, Radiation biologyV.45, No.3, p 271-280.
[14] Frantsevich L I, Panjkov I V, Ermakov A A 1995 Ecology p. 57-62.
[15] Lavrentyeva G V, Mirzeabasov O A, Synzynys B I 2014 Int. J. Environ. Res., 8(4): p 961-970
[16] Vorobejchik E L 2004 Ecological rationing of toxic loads on terrestrial ecosystems: Doctor’s thesis Ekaterinburg 362 pp.