

*This paper reports the results of environmental studies conducted in May–June 2021 in 16 prefectures of Japan, over which radioactive clouds spread as a result of the accident at the Fukushima-1 nuclear power plant in March 2011, namely: Aomori, Miyagi, Fukushima, Niigata, Ishikawa, Ibaraki, Kanagawa, Shizuoka, Osaka, Kyoto, Okayama, Tottore, Shimane, Kagoshima, Nagasaki, and Ehime. Some effects of the impact of accidental emissions of BWR-3 and BWR-4 reactors on the environment in the settlements neighboring the Fukushima-1 nuclear power plant were summed up. The methods of conducting research are described, starting with a sampling of environmental objects in the above prefectures (atmospheric air, soil, vegetation, agricultural products, sea and ocean water, and aquatic fauna) and sample preparation up to measurements and data processing to obtain results and their subsequent analysis. A comparison of the results of research with the volumes of emissions of radioactive substances due to the normal operation of various objects for the use of atomic energy was carried out. Conclusions are drawn about the medium-term (after 10 years) consequences of the nuclear accident at the Fukushima-1 nuclear power plant.*

*The maximum background radiation equal to 2  $\mu\text{Sv/h}$  observed at point No. 83, 20 km from the Fukushima-1 nuclear power plant, will not lead to irradiation of a person with doses exceeding the permissible limits if the time of his stay at the reference point is limited to 1.36 hours per day. The concentration of cesium-137 in the water of Japanese waters does not exceed the level of the radiation factor, upon reaching which protective intervention is necessary. The maximum specific activity of cesium-137, detected in soil and vegetation samples near the Fukushima-1 nuclear power plant, does not exceed the maximum specific activity (MZUA). In Japan as a whole, the specific activity of radionuclides in soils used for agricultural production and livestock grazing does not exceed the levels of global pollution*

*Keywords: radiation, environmental situation in Japan, nuclear power plant (NPP), Fukushima, environment, sampling*

# RADIATION ENVIRONMENTAL SITUATION IN JAPAN, 10 YEARS AFTER THE ACCIDENT AT THE NUCLEAR POWER PLANT "FUKUSHIMA-1"

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## 1. Introduction

The accident at the Fukushima-1 nuclear power plant was assigned a level 7 of complexity on the INES scale [1] – the same as that of the Chernobyl accident, although the amount of radioactivity released into the environment as a result of the accident at the Fukushima-1 nuclear power plant ( $770 \cdot 10^{15}$  Bq) is no more than 15 % of the emissions from the Chernobyl accident ( $5200 \cdot 10^{15}$  Bq). Comparison of the consequences of nuclear accidents by the total activity of volley ejection is not fully justified from the point of view of physics since the summation of the activities of alpha-, beta-, gamma- and neutron emitters is illogical, but, nevertheless, it is the only way to make a quantitative comparison of the scale of the incidents.

Therefore, studies on radiation monitoring in the area of the Fukushima-1 nuclear power plant 10 years after the accident are relevant, in order to determine the gamma radiation background of the area in the prefectures over which the

plumes of radioactive clouds spread, as well as residual levels of radioactive contamination of environmental objects and agricultural products.

## 2. Literature review and problem statement

Four units at the Fukushima-1 nuclear power plant were seriously damaged due to the loss of external electrical load and the failure of backup generators flooded by a 14-meter tsunami wave. As a result, there was an increase in temperature in the core, melting of fuel and core, steam-zirconium reaction [1], which led to the leakage of hydrogen and its explosion and, as a result, the release of large volumes of radioactive aerosols into the environment.

Paper [1] proposes a tree of failures that initiate and accompany a nuclear accident at a reactor. The developed fault tree allows for a thorough analysis of emergencies at nuclear reactors, the results of which can be used in the development

of organizational and technical measures to prevent nuclear and radiation accidents at various stages of their development.

Work [2] shows the activity of emergency emissions from the reactors BWR-3 (power unit 1 of the Fukushima-1 NPP), BWR-4 (power units 2 and 3 of the Fukushima-1 NPP), and RBMK-1000 (power unit 4 of the Chernobyl nuclear power plant). The main melting of the fuel occurred at an early stage at all three units, although the fuel remained mostly contained, with the exception of some volatile fission products that were discarded early or released from unit 2 in mid-March, as well as some soluble products that flowed along with the water, especially from unit 2, where the protective shell is clearly broken. Access was gained to all three reactor buildings but the dose rate inside remained high. Tepco announced a “cold shutdown” in mid-December 2011 when emissions of radioactive substances fell to a minimum level.

Paper [3] considered a malfunction in the reactor cooling system, the feed pumps stopped, and the access of feed water circulating along the second circuit to the steam generators stopped. The turbine generator of the nuclear power plant was automatically turned off, and the auxiliary system for supplying feed water to steam generators was turned on. However, although all three pumps of the core cooling system functioned normally, water was not supplied to the steam generators. It turned out that the output valves on these pumps were closed during routine repairs. Radioecological studies were carried out in the above prefectures of Japan since the spread of radioactive clouds as a result of the accident at the Fukushima-1 nuclear power plant occurred precisely over their territories [3]. A study of the operation of reactors showed that the shortcomings of sodium as a heat carrier associated with its chemical activity in relation to the oxygen of the air and to the water are compensated by systems that ensure its reliable operation. Work [4] sets out the provision of methods and conditions for conducting dosimetric control and hygienic assessment of the parameters of laser radiation at the workplaces of service personnel in order to determine the degree of radiation danger to the human body. The sensitivity of the environmental pollution monitoring technique carried out in the area of the Fukushima-1 nuclear power plant depends mainly on the sensitivity of the spectrometer. During the research, 2 types of spectrometers were used. By determining the spectrum of isotopes contained in a sample, the spectrometer automatically indicates the measurement error of the activities of each radionuclide in the counting sample [4]. This deviation did not significantly affect the final measurement result, given the long half-lives of  $^{134}\text{Cs}$  ( $T_{1/2}=2.09$  years) and  $^{137}\text{Cs}$  ( $T_{1/2}=30$  years). Paper [5] touches upon the issue of exposure of spent nuclear fuel (SNF) of unit No. 4 as a result of the steam-zirconium reaction, hydrogen began to be released, which interacted with atmospheric oxygen, which led to an explosion that seriously damaged the reactor buildings of units Nos. 3 and 4. Irradiated fuel in holding pools must be cooled by maintaining the water level and forced circulation by pumps, under continuous control of the water temperature, which should not exceed  $25\text{ }^{\circ}\text{C}$ . However, the water temperature in the exhaust nuclear fuel holding pool of unit No. 4 was  $84\text{ }^{\circ}\text{C}$ , and the fuel assemblies stored in it were exposed. SNF in the cooling basin of the third power unit posed the greatest danger since the spent rods of its irradiated fuel tanks contained mixed uranium-plutonium fuel ( $\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_2$ ). In the fuel stations stored in the basins of other power units, the less toxic uranium fuel  $\text{UO}_2$  was used. This problem needed to be fixed. Work [6] considers the requirements of the standards that do not apply to radiation sources that create under any conditions of handling them:

1) individual annual effective dose of not more than 10 microsieverts;

2) an individual annual equivalent dose in the skin of not more than 50 millisieverts and in the eyepiece not more than 15 mSv;

3) a collective effective annual dose of not more than 1 person-sievert (hereinafter referred to as per-Sv), or when at a collective dose of more than 1 person-Sv, an optimization assessment shows the inexpediency of reducing the collective dose.

Study [7] considers the same methods and standards and sanitary-epidemiological monitoring. Analysis, assessment, and prognosis, as well as the determining of cause-and-effect relationships between the state of public health and the impact of environmental factors. Paper [8] discusses various types and descriptions of the application of the first BN-350 fast neutron power reactor. Work [9] considers the scenario of the development of an accident at the most powerful power plant in the Soviet Union – the Chernobyl nuclear power plant, the accident at the fourth power unit occurred during the experiment on the operation of the reactor on the run-out – similar to the events that occurred on the Three Mile Island in the United States, but the scale of the consequences of these nuclear events is incomparable. In [10], the authors highlight the issues of thermal inertia, which is much more difficult to calculate since it is not limited to the heat capacity of the structure in the reactor. Even after the cessation of the chain reaction in the reactor, the production of fragmentation fission products continues. The experiment was possible only when the emergency shutdown system of the reactor was turned off since the emergency power supply is automatically turned on when the reactor power decreases below the established limit. Paper [11] formulated and recorded an increase in temperature in the storage pools of spent nuclear fuel (SNF) of units No. 5, 6, the water level in which decreased, and the upper parts of the fuel rods were exposed. Nevertheless, the depressurization of the fuel rods did not occur since the power of the diesel generator of unit No. 6, connected to the emergency power supply system, was enough to supply water to the pools. Thus, the restored emergency power supply of the residual heat removal system made it possible to reduce the temperature in the SNF holding pools of power units 5 and 6, which were put into cold stop mode. Study [12] considered the results of the accident at the Fukushima-1 nuclear power plant four years after its occurrence where the specific activity of individual agricultural products contaminated with  $^{133}\text{I}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and other radionuclides exceeded the maximum permissible value. Of the agricultural produce grown in the prefectures of Japan, the most common and available are rice and spinach, for determining the residual level of radioactive contamination of food.

Analysis of the literature suggests that it is expedient to conduct a study of the radiation environmental situation in Japan ten years after the accident at the Fukushima-1 nuclear power plant.

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### 3. The aim and objectives of the study

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The purpose of this study is to determine the gamma radiation background of the area at the Fukushima-1 nuclear power plant, in March 2011, in 16 prefectures of Japan, over which plumes of radioactive clouds, residual levels of radioactive contamination of environmental objects and agricultural products spread.

To accomplish the aim, the following tasks have been set:

- justification for the sampling of air, dust, and precipitation, samples of sea and ocean water;
- determining the concentrations of long-lived and short-lived radioactive aerosols in air samples;
- determining the residual level of radioactive contamination in precipitation and settling dust, sea and ocean water, soil, vegetation, and agricultural products.

#### 4. The study materials and methods

The hypothesis of this study was the possibility of determining the gamma radiation background, the measurement of which was carried out from the windward (Aomori and Miyagi) and leeward (Niigata, Ishikawa, Ibaraki, Kanagawa, Shizuoka, Osaka, Kyoto, Okayama, Tottore, Shimane, Kagoshima, Nagasaki, and Ehime) sides from the Fukushima-1 nuclear power plant. With the help of portable dosimeters ICS-331B and ICS-323C made by the Japanese company “Hitachi Aloka Medical, Ltd.” (6-22-1, Mure, Mitaka-shi, Tokyo, 181-8622, Japan), capable of recording an equivalent dose rate of X-ray, beta and gamma radiation ranging from 0.03  $\mu\text{mSv/h}$  to 10  $\text{mSv/h}$ . Data on dose capacities were noted on a schematic map of the area in the cities of the prefectures along the path of propagation of radioactive clouds [3, 4].

To determine the concentration of long-lived and short-lived radioactive aerosols in air samples taken in prefectures adjacent to the Fukushima-1 nuclear power plant. For sampling, the portable aspiration device RADeCO, model H-810-2, was used.

Air samples were taken in prefectures adjacent to the Fukushima-1 nuclear power plant in 2021 for 2 hours: the air was passed at a speed of 35 CFM (0.98  $\text{m}^3$  per minute) through an HD-2061 filter with a diameter of 0.025 m [4]. The HD-2061 filters through which air was passed were insulated, and the ash residue was subjected to spectrometric analysis.

The residual level does not exceed the permissible values of radioactive contamination in precipitation and settling dust, sea and ocean water, soil, vegetation and agricultural products, samples of which were taken in the same prefectures of Japan where gamma background levels were measured and air intake were taken.

Samples of dust and precipitation were collected in vinyl containers with dimensions of 0.5 $\times$ 0.5 m, with a height of the sides of 10 cm, strengthened at a height of 1.5–2 m from the ground surface. The dimensions of the containers provide the necessary accountability of the sample at a low level of radioactive deposition [4].

Containers with accumulated samples of settling dust were treated with a cotton swab soaked in alcohol, which was subsequently insulated at a temperature of 450  $^{\circ}\text{C}$  and subjected to spectrometric analysis. Liquid sediments were poured from the containers into quartz cups and evaporated on an electric stove; the walls of the cups were wiped with a cotton swab soaked in a 10 percent solution of sulfuric or nitric acid, which was also measured on a spectrometer after ashing.

Soil samples were taken by quartering at a depth of 50–100 mm, dried, and sieved through a sieve with a cell size of 1–1.5  $\text{mm}^2$ . Vegetation samples weighing 1–2 kg were taken at the same points as the soil, dried, and insulated. Measurements of soil activity in the dry air state and ash vegetation were carried out on the third to the fifteenth day after selection.

#### 5. Results of environmental monitoring and sampling

##### 5.1. Justification for sampling

The volumes of sea and ocean water samples were 2–6 liters, which were evaporated in quartz cups before the formation of a dry residue on the walls. The dry residue was removed with a cotton swab soaked in alcohol or in a solution of nitric acid and was ashed at a temperature of 450  $^{\circ}\text{C}$ . Then a spectrometric analysis of counting samples was carried out.

In the same waters in which water samples were taken, fish of various varieties were caught. The caught fish were delivered to the laboratory where counting samples were prepared from fish heads with gills, bodies with ridges, and from the liver. Ashing of the fish also occurred at a temperature of 450  $^{\circ}\text{C}$ , and the ash residue was subjected to spectrometric analysis. Since fishing for the most popular fish varieties in Japan is carried out by bottom trawls at a depth of 40–60 m (yellow-tailed lacedra, golden mackerel) to 120–150 m (halibut), counting samples were made from products purchased in harbors and at fairs near them.

##### 5.2. Determining the presence of radionuclides

To determine the presence of radionuclides in agricultural products of terrestrial origin, spectrometric analysis of rice and spinach collected in areas over which a radioactive cloud from the epicenter of the accident at the Fukushima-1 nuclear power plant spread was carried out. The collected rice and spinach were dried first at room temperature, then in a drying oven at a temperature of 120  $^{\circ}\text{C}$ , after which they were settled in a muffle furnace at a temperature of 450  $^{\circ}\text{C}$ . The specific activity of man-made radionuclides was determined in ash residues of spinach and rice using a spectrometer.

Comparative data on the total activities released into the environment as a result of accidents at the Fukushima-1 nuclear power plant and at the Chernobyl nuclear power plant are given in Table 1 [2].

Table 1  
The activity of emergency emissions from the reactors BWR-3 (power unit 1 of the Fukushima-1 nuclear power plant), BWR-4 (power units 2 and 3 of the Fukushima-1 nuclear power plant), and RBMK-1000 (power unit 4 of the Chernobyl nuclear power plant).

Isotope	$^{133}\text{Xe}$	$^{134}\text{Cs}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{131}\text{I}$	$^{239}\text{Pu}$
Emission from BWR-3, Bq	$3.4 \times 10^{18}$	$7.1 \times 10^{14}$	$5.9 \times 10^{14}$	$6.1 \times 10^{12}$	$1.2 \times 10^{16}$	$8.6 \times 10^7$
Emission from BWR-4, Bq	$3.5 \times 10^{18}$	$1.6 \times 10^{16}$	$1.4 \times 10^{16}$	$4.8 \times 10^{13}$	$1.4 \times 10^{17}$	$3.1 \times 10^9$
Emission from BWR-4, Bq	$4.4 \times 10^{18}$	$8.2 \times 10^{14}$	$7.1 \times 10^{14}$	$8.5 \times 10^{13}$	$7.0 \times 10^{15}$	$4.0 \times 10^7$
Total emission from the Fukushima-1 nuclear power plant	$1.1 \times 10^{19}$	$1.8 \times 10^{16}$	$1.5 \times 10^{16}$	$1.4 \times 10^{14}$	$1.6 \times 10^{17}$	$3.2 \times 10^9$
Emission from RBMK-1000, Bq	$1.7 \times 10^{18}$	$3.8 \times 10^{16}$	$8.5 \times 10^{16}$	$1.4 \times 10^{17}$	$1.8 \times 10^{18}$	$0.7 \times 10^{14}$

The cold shutdown parameters (core temperature below 80 °C) of the emergency reactors of the Fukushima-1 nuclear power plant were achieved only by the end of October 2011; at the same time, the flow of radioactivity into the external environment stopped.

In monitoring radioactive contamination in the prefectures of Aomori, Miyagi, Fukushima, Niigata, Ishikawa, Ibaraki, Kanagawa, Shizuoka, Osaka, Kyoto, Okayama, Tottore, Shimane, Kagoshima, Nagasaki, and Ehime, a procedure consisting of sampling, preparation of counting samples, their spectrometric analysis, and processing of the results obtained was used. Radioecological studies were carried out in the above prefectures of Japan since the spread of radioactive clouds as a result of the accident at the Fukushima-1 nuclear power plant occurred precisely over their territories [3].

The accuracy of the environmental pollution monitoring methodology carried out in the area of the Fukushima-1 nuclear power plant depends mainly on the sensitivity of the spectrometer. During the research, 2 types of spectrometers were used: one – portable with a lower threshold of sensitivity of 6 Bq, and the other – stationary with a lower threshold of sensitivity of 1 Bq. By determining the spectrum of isotopes contained in a sample, the spectrometer automatically indicates the measurement error of the activities of each radionuclide in the counting sample. With a probability of measurement error of more than 40 %, its result is annulled [4].

Pedestrian gamma survey in cities and prefectures located in the direction of propagation of the radioactive cloud was carried out using the industrial devices DKS-AT1121 and DKS-AT1123, operating on the basis of solid-state scintillators NaI. The average values of the results of measurements of the gamma background of the area in 16 prefectures are given in Table 2.

**Radiation levels of the area in the prefectures of Japan in the direction of the spread of radioactive clouds 10 years after the accident at the nuclear power plant “Fukushima-1”**

City name (prefecture)	Dose rate (radiation background), μmSv/h	Measurement date	Radiation control device
Fukushima*	0.24	06.05.2021	DKS-AT1121
Mito (Ibaraki)	0.20	12.05.2021	DKS-AT1121
Yokohama (Kanagawa)	0.16	13.05.2021	DKS-AT1121
Shizuoka	0.17	14.05.2021	DKS-AT1121
Kyoto	0.11	16.05.2021	DKS-AT1121
Osaka	0.14	17.05.2021	DKS-AT1121
Okayama	0.15	19.05.2021	DKS-AT1121
Matsuyama (Ehime)	0.10	22.05.2021	DKS-AT1121
Kagoshima	0.10	24.05.2021	DKS-AT1121
Nagasaki	0.08	25.05.2021	DKS-AT1121
Matsue (Shimane)	0.07	26.05.2021	DKS-AT1121
Tottori	0.09	29.05.2021	DKS-AT1121
Kanazawa (Ishikawa)	0.11	31.05.2021	DKS-AT1123
Niigata	0.13	05.06.2021	DKS-AT1123
Sendai (Miyagi)	0.12	08.06.2021	DKS-AT1121
Aomori	0.11	10.06.2021	DKS-AT1123

Note: \* – Measurements of the gamma background at Fukushima were performed throughout the prefecture, with the exception of the area of the 20-kilometer zone surrounding the BWR-3 and BWR-4 emergency reactors. At reference control point No. 83 at the boundary of the 20-kilometer zone, the dose rate did not exceed 2 μmSv/h

The results of spectrometric analysis of filters ND-2061 Table 3 showed that in the prefectures of Japan in the direction of the spread of radioactive clouds after the accident at the Fukushima-1 nuclear power plant, the volumetric activity of atmospheric air does not exceed  $4.5 \times 10^{-3}$  Bq/m<sup>3</sup>.

**Table 3**  
**The volumetric activity of atmospheric air samples taken in the prefectures of Japan in the direction of the spread of radioactive clouds 10 years after the accident at the nuclear power plant “Fukushima-1”**

City name (prefecture)	Volume activity, Bq/m <sup>3</sup>	Measurement date	The volume of the pumped air, m <sup>3</sup>
Fukushima**	$4.3 \times 10^{-3}$	06.05.2021	100.3
Mito (Ibaraki)	$2.6 \times 10^{-3}$	12.05.2021	99.2
Yokohama (Kanagawa)	$1.8 \times 10^{-3}$	13.05.2021	99.8
Shizuoka	$2.7 \times 10^{-3}$	14.05.2021	98.9
Kyoto	$2.1 \times 10^{-3}$	16.05.2021	97.5
Osaka	$1.9 \times 10^{-3}$	17.05.2021	101.1
Okayama	$1.5 \times 10^{-3}$	19.05.2021	100.0
Matsuyama (Ehime)	$1.1 \times 10^{-3}$	22.05.2021	99.3
Kagoshima	$0.8 \times 10^{-3}$	24.05.2021	98.6
Nagasaki	$0.9 \times 10^{-3}$	25.05.2021	98.3
Matsue (Shimane)	$0.7 \times 10^{-3}$	26.05.2021	100.4
Tottori	$0.7 \times 10^{-3}$	29.05.2021	100.3
Kanazawa (Ishikawa)	$1.1 \times 10^{-3}$	31.05.2021	100.0
Niigata	$1.1 \times 10^{-3}$	05.06.2021	99.5
Sendai (Miyagi)	$2.3 \times 10^{-3}$	08.06.2021	98.6
Aomori	$0.9 \times 10^{-3}$	10.06.2021	100.0

Note: \*\* – The air intake in Fukushima was carried out at a distance of 20 km from the emergency reactors BWR-3 and BWR-4; air sampling directly at the industrial site of the Fukushima-1 nuclear power plant was not carried out

**Table 2**

The volumetric activity of atmospheric air was determined by the results of filter spectrometry. The product of the time and speed of pumping determines the volume of air passed through the filter. Dividing the result of spectrometric analysis by the volume of pumped air made it possible to obtain volumetric activity.

The results of spectrometric analysis of the ashed filters HD-2061 of the RADeCO aspiration device, model H-810-2, suggest that the main technogenic radioisotope that determines the volumetric activity of counting atmospheric air samples is cesium-137 Table 4. The dose of internal irradiation will depend not only on the volumetric activity of the radionuclide but also on the dispersion, and on the path of entry into the body.

Data in Table 3 may indicate that with an annual volume of inhaled air of 8100 m<sup>3</sup> [6, 7], an adult living on the border of the 20-kilometer zone from the Fukushima-1 nuclear power plant will be exposed to internal irradiation with a dose formed by a mixture of isotopes with a total activity of 34.8 Bq.

**5. 3. Determining the activity of atmospheric air spectrometry of filters and the level of surface pollution of the bottoms**

The volumetric activity of atmospheric air was determined by the results of filter spectrometry. Using a cup anemometer, the speed of air suction with a sampler,

or the speed of pumping through the filter, was measured. The product of the time and speed of pumping determines the volume of air passed through the filter. Dividing the result of spectrometric analysis by the volume of pumped air made it possible to obtain volumetric activity.

To calculate the annual dose of internal irradiation of a person from inhalation of air with a volume activity of  $4.3 \times 10^{-3}$  Bq/m<sup>3</sup>, it is necessary, first of all, to determine which radioisotopes it is formed by. The results of spectrometric analysis of the ashed filters HD-2061 of the RADeCO aspiration device, model H-810-2, suggest that the main technogenic radioisotope that determines the volumetric activity of counting atmospheric air samples is cesium-137 are given in Table 4. The dose of internal irradiation will depend not only on the volumetric activity of the radionuclide but also on the dispersion, and on the path of entry into the body.

Table 4

Radioisotope composition of atmospheric air in the prefectures of Japan in the direction of the spread of radioactive clouds 10 years after the accident at the nuclear power plant "Fukushima-1"

City name (prefecture)	Volume activity, Bq/m <sup>3</sup>			
	<sup>131</sup> I	<sup>134</sup> Cs	<sup>137</sup> Cs	<sup>7</sup> Be
Fukushima	n/a	$0.04 \times 10^{-3}$	$2.2 \times 10^{-3}$	$1.8 \times 10^{-3}$
Mito (Ibaraki)	$0.01 \times 10^{-3}$	$0.02 \times 10^{-3}$	$1.3 \times 10^{-3}$	$0.9 \times 10^{-3}$
Yokohama (Kanagawa)	n/a	n/a	$0.9 \times 10^{-3}$	$0.5 \times 10^{-3}$
Shizuoka	n/a	$0.03 \times 10^{-3}$	$1.4 \times 10^{-3}$	$1.0 \times 10^{-3}$
Kyoto	n/a	$0.05 \times 10^{-3}$	$1.5 \times 10^{-3}$	$0.8 \times 10^{-3}$
Osaka	n/a	n/a	$1.1 \times 10^{-3}$	$0.5 \times 10^{-3}$
Okayama	n/a	n/a	$0.9 \times 10^{-3}$	$0.4 \times 10^{-3}$
Matsuyama (Ehime)	n/a	n/a	$0.8 \times 10^{-3}$	$0.5 \times 10^{-3}$
Kagoshima	n/a	n/a	$0.8 \times 10^{-3}$	$0.3 \times 10^{-3}$
Nagasaki	n/a	n/a	$0.7 \times 10^{-3}$	$0.3 \times 10^{-3}$
Matsue (Shimane)	n/a	n/a	$0.6 \times 10^{-3}$	$0.3 \times 10^{-3}$
Tottori	n/a	n/a	$0.6 \times 10^{-3}$	$0.4 \times 10^{-3}$
Kanazawa (Ishikawa)	$0.01 \times 10^{-3}$	n/a	$1.5 \times 10^{-3}$	$0.9 \times 10^{-3}$
Niigata	$0.01 \times 10^{-3}$	n/a	$0.7 \times 10^{-3}$	$0.6 \times 10^{-3}$
Sendai (Miyagi)	$0.01 \times 10^{-3}$	$0.06 \times 10^{-3}$	$1.1 \times 10^{-3}$	$0.8 \times 10^{-3}$
Aomori	n/a	n/a	$0.5 \times 10^{-3}$	$0.4 \times 10^{-3}$

Note: n/a value below the sensitivity threshold of the spectrometer

In samples of fish, rice, and spinach, taken in the direction of the spread of radioactive clouds as a result of the accident at the Fukushima-1 nuclear power plant, no radionuclides of man-made origin were found, which indicates the biological purity of food consumed by Japanese residents and exported to the Far East and Western Europe.

The maximum specific activity of cesium-137, detected in soil and vegetation samples near the Fukushima-1 nuclear power plant, does not exceed the minimum significant value (MZUA) equal to 10,000 Bq/kg [6].

The isotopic composition of the water of the Sea of Japan and the East China Sea and the Pacific Ocean, investigated 10 years after the accident at the Fukushima-1 nuclear power plant, is given in Table 7.

Table 5

Radioisotope composition of soil in the prefectures of Japan in the direction of the spread of radioactive clouds 10 years after the accident at the nuclear power plant "Fukushima-1"

City name (prefecture)	Specific activity of isotopes, Bq/kg						
	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>210</sup> Pb	<sup>228</sup> Th	<sup>226</sup> Ra	<sup>234</sup> Th	<sup>40</sup> K
Fukushima	1,500	1,100	85	44	37	30	453
Mito (Ibaraki)	130	81	n/a	50	36	26	510
Yokohama (Kanagawa)	127	73	100	40	35	22	512
Shizuoka	96	66	110	42	29	23	466
Kyoto	87	70	80	39	34	30	430
Osaka	110	75	90	45	30	25	679
Okayama	115	70	85	43	28	25	616
Matsuyama (Ehime)	69	53	n/a	38	28	29	700
Kagoshima	100	71	99	46	29	27	493
Nagasaki	98	72	98	45	30	28	492
Matsue (Shimane)	88	63	n/a	45	36	29	635
Tottori	130	74	n/a	45	36	22	501
Kanazawa (Ishikawa)	93	61	100	50	33	20	578
Niigata	101	89	107	52	30	25	480
Sendai (Miyagi)	107	99	83	41	30	25	599
Aomori	89	64	n/a	40	30	28	610

Note: n/a value below the sensitivity threshold of the spectrometer

Table 6

Radioisotope composition of vegetation in the prefectures of Japan in the direction of the spread of radioactive clouds 10 years after the accident at the nuclear power plant "Fukushima-1"

City name (prefecture)	Specific activity of isotopes, Bq/kg						
	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>210</sup> Pb	<sup>228</sup> Th	<sup>226</sup> Ra	<sup>234</sup> Th	<sup>40</sup> K
Fukushima	660	530	2.5	0.33	0.28	15	90
Mito (Ibaraki)	56	38	n/a	0.35	0.24	16	100
Yokohama (Kanagawa)	51	34	3.1	0.39	0.26	18	89
Shizuoka	42	27	2.4	0.40	0.23	15	80
Kyoto	45	28	1.9	0.42	0.27	20	78
Osaka	53	36	2.1	0.44	0.25	n/a	140
Okayama	49	36	1.8	0.47	0.28	13	142
Matsuyama (Ehime)	40	30	n/a	0.28	0.23	14	148
Kagoshima	48	33	1.8	0.34	0.24	n/a	99
Nagasaki	49	40	1.7	0.32	0.28	n/a	95
Matsue (Shimane)	47	32	n/a	0.29	0.29	19	110
Tottori	55	31	n/a	0.36	0.30	10	91
Kanazawa (Ishikawa)	50	39	2.2	0.29	0.31	10	100
Niigata	70	47	2.5	0.28	0.30	11	89
Sendai (Miyagi)	75	48	1.6	0.31	0.29	16	103
Aomori	49	36	n/a	0.39	0.28	17	106

Note: n/a value below the sensitivity threshold of the spectrometer

In the ash residues of cotton swabs, with the help of which dust samples were taken on the bottoms of vinyl containers, the presence of mainly cesium-137 was also detected. Levels of surface contamination of the bottoms of vinyl containers established in different prefectures range from  $0.1 \times 10^{-6}$  to  $1.5 \times 10^{-6}$  Bq/km<sup>2</sup>. Traces of other radionuclides (cadmium-109, lanthanum-140, cesium-134, etc.) in the isotopic composition

of dust samples are insignificant, or their volumetric (specific) activities are below the sensitivity level of the spectrometer.

Table 7

Radioisotope composition of water in the waters surrounding the islands of Honshu, Kyushu, and Shikoku, 10 years after the salvo release of activity as a result of the accident at the Fukushima-1 nuclear power plant

City name (prefecture)	Volume activity of isotopes, Bq/m <sup>3</sup>	Cs <sup>137</sup>	Cs <sup>134</sup>	Co <sup>60</sup>	Co <sup>57</sup>	Zn <sup>65</sup>
Fukushima		5.6	4.1	n/a	n/a	n/a
Mito (Ibaraki)		4.2	2.8	n/a	n/a	n/a
Yokohama (Kanagawa)		3.6	2.5	n/a	0,10	n/a
Shizuoka		3.7	3.0	n/a	n/a	n/a
Kyoto		4.0	2.8	n/a	0,15	n/a
Osaka		3.9	2.5	n/a	n/a	n/a
Okayama		3.7	2.8	n/a	n/a	n/a
Matsuyama (Ehime)		4.1	3.2	0.11	n/a	n/a
Kagoshima		3.9	3.1	n/a	n/a	n/a
Nagasaki		3.7	2.9	0.06	n/a	n/a
Matsue (Shimane)		3.5	2.6	n/a	n/a	n/a
Tottori		3.7	2.4	n/a	n/a	n/a
Kanazawa (Ishikawa)		3.7	2.7	n/a	0,16	n/a
Niigata		4.2	3.2	n/a	n/a	n/a
Sendai (Miyagi)		4.1	2.8	n/a	n/a	n/a
Aomori		3.6	2.8	n/a	n/a	n/a

Note: n/a value below the sensitivity threshold of the spectrometer

The effective dose of human exposure to radioactively contaminated surfaces ( $D_s$ ) can be found from the formula given in [9]:

$$D_s = K(i) \times A(i) \times X \times V \times T, \tag{1}$$

where  $K(i)$  is the dose factor depending on the distance from the place of release of the radionuclide into the atmosphere. At a distance of 20 km from the emergency reactors of the Fukushima-1 nuclear power plant,  $K(i) = 0.052 \times 10^{-13}$  Sv/Bq [9];

$A(i)$  is the volumetric activity of cesium-137 in atmospheric air (Table 4);

$X = 8.1 \times 10^{-6}$  s/m<sup>3</sup> – dilution coefficient of radioactive impurity in the atmosphere with Pasquille stability classes A, B, and F [10];

$V = 2.0 \times 10^{-2}$  m<sup>3</sup>/s – the rate of deposition of radionuclides to the earth from the atmosphere with Pasquille stability classes A, B, and F [10];

$T$  is the area of the contaminated surface.

However, the result obtained as a result of the application of equation (1) will have a significant error, if only because the density of contamination of the earth’s surface will not be equal to the surface contamination of vinyl plates installed at a height of 2 m. The results of studies of countable samples of soil and vegetation are given in Tables 5, 6.

**6. Discussion of results of the methodology and monitoring of radioactive contamination of the environment in the prefectures of Japan**

The main elements that entered the atmosphere during the accident were iodine-131 and cesium-137, their amount

was approximately 10 % of the emissions of the Chernobyl nuclear power plant.

Table 1 gives comparative data on the total capacities of emergency emissions from the reactors BWR-3 (power unit 1 of the Fukushima-1 NPP), BWR-4 (power units 2 and 3 of the Fukushima-1 NPP), and RBMK-1000 (power unit 4 of the Chernobyl nuclear power plant).

Table 2 gives the average values of the results of measurements of the gamma background of the area in 16 prefectures of Japan 10 years after the accident at the Fukushima-1 nuclear power plant.

Table 3 shows the results of spectrometric analysis of ND-2061 filters removed from the RAdECo sampler, model H-810-2, in the prefectures of Japan in the direction of propagation of a radioactive cloud after the accident at the Fukushima-1 nuclear power plant, the volumetric activity of atmospheric air does not exceed  $4.5 \times 10^{-3}$  Bq/m<sup>3</sup>.

Table 4 demonstrates that the dose of internal irradiation will depend not only on the volumetric activity of the radionuclide but also on the dispersion, and on the path of entry into the body.

Tables 5, 6 give the results of studies of countable soil and vegetation samples of radioisotope composition, samples of which were taken in the exclusion zone at a distance of 10–12 km from the Fukushima-1 NPP, contaminated mainly with cesium-137 and cesium-134.

The data in Table 7 indicate that the level of contamination of Japanese waters with cesium-137 does not exceed 0.006 Bq/l while the level of intervention for this radioisotope is 11 Bq/kg [6]. The sum of the ratios of activity of each of the radionuclides contained in sea and ocean water to the corresponding level of intervention does not exceed 1, which satisfies the requirements even for drinking water [6].

For the specific use of the results of the study aimed at obtaining a rational solution, it is necessary to perform the following stages:

- with an annual volume of inhaled air of 8100 m<sup>3</sup> [6, 7], an adult living on the border of the 20-kilometer zone from the Fukushima-1 nuclear power plant will be exposed to internal irradiation with a dose formed by a mixture of isotopes with a total activity of 34.8 Bq. Off the coasts of Miyagi, Fukushima, Ibaraki, Yokohama, Shizuoka, Osaka, Okayama, and Ehime prefectures, the Pacific Ocean water was sampled. Water samples from the East China Sea were taken at a distance of 500 m from Kagoshima, Nagasaki; and from the harbors of Shimane, Tottori, Ishikawa, Niigata, and Aomori boats were dispatched to sample the waters of the Sea of Japan. The samples of sea and ocean water were examined to determine the degree of dilution of activity ejected in the waters surrounding the islands of Honshu, Kyushu, and Shikoku. As a result of the accident at the Fukushima-1 nuclear power plant, radioactive substances with a total activity of about 5 PBq ( $5 \times 10^{15}$  Bq) [2] entered the waters of the Pacific Ocean, as well as the Sea of Japan and the East China Sea, which is 62.5 times higher than the total activity that was discharged into the Caspian Sea during the operation of the BN-350 reactor ( $0.08 \cdot 10^{15}$  Bq) [8];

- technogenic radioisotopes that settle on the surface of the earth are concentrated in the upper layer of the soil (about 5 cm). Within a year after the accident at the Fukushima-1 nuclear power plant in the prefectures of Japan, deep plowing was carried out, which made it possible to transfer part of cesium-137 and 134 to the lower layers of the soil, to which the root systems of plants do not reach.

Research continues to improve alternative decontamination methods [11]. It was found that 66 % of radioactive cesium dissolves in a concentrated solution of potassium chloride in 244 hours. At room temperature with gamma radiation power from contaminated soil can be reduced by more than 97 % if covered with a layer of unpolluted soil 20 cm thick;

– measurement of the gamma radiation background of the area showed that no radionuclides of technogenic origin were found in samples of fish, rice, and spinach selected in the direction of the propagation of radioactive clouds as a result of the accident at the Fukushima-1 nuclear power plant. In samples of the fish caught at sea and ocean water sampling sites, man-made radionuclides are contained in quantities well below the sensitivity level of the spectrometer, which is close to zero.

Further work on the implementation of the results of our seminal study could make it possible to use food products of biological purity, eaten by the inhabitants of Japan and exported to the Far East and Western Europe, as well as improve the accuracy of the measured results.

## 7. Conclusions

1. Permissible values of radioactive contamination do not exceed the residual level in precipitation and settling dust, sea and ocean water, soil, vegetation, and agricultural products, samples of which were taken in the same prefectures of Japan where gamma background and air intake levels were measured.

2. The highest volumetric activity of atmospheric air ( $4.3 \times 10^{-3}$  Bq/m<sup>3</sup>) and the level of surface pollution by

dust and precipitation ( $1.5 \times 10^{-6}$  Bq/km<sup>2</sup>), containing mainly cesium-137, are also registered in Fukushima Prefecture, therefore, when conducting individual dosimetry control of Japanese residents, it is necessary to take into consideration internal irradiation with traces of cesium present in the atmospheric air and settling dust after the accident at the Fukushima-1 nuclear power plant.

3. In Japan as a whole, the specific activity of radionuclides in soils used for agricultural production and livestock grazing does not exceed the levels of global pollution. The maximum specific activity of cesium-137, found in soil and vegetation samples near the Fukushima-1 nuclear power plant, no longer exceeds MZUA [6].

The concentration of cesium-137 in the water of Japanese waters does not exceed the level of the radiation factor, upon reaching which protective intervention is necessary. A significant drawback in the study of water samples is associated with an omission in determining their salinity, however, knowing that the spread of radioactive substances in the liquid does not occur fractally, but is described by the laws of diffusion, it can be reasonably assumed that the consequences of the accident at the Fukushima-1 nuclear power plant did not affect the biological purity of the Pacific Ocean, the Sea of Japan, and the East China Sea.

Thus, the nuclear accident at the Fukushima-1 nuclear power plant, which occurred in March 2011, according to experts, created a very difficult radiation situation, however, thanks to the highly organized liquidation and localization of its consequences, the environmental situation in Japan 10 years after the disaster is quite safe, including the territory of the 20-kilometer exclusion zone, where work continues on radiation monitoring of environmental objects.

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