Journal of Environmental Radioactivity 172 (2017) 1–9

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

# Radioactive contamination of the soil–plant cover at certain locations of Primorsky Krai, Sakhalin Island and Kamchatka Peninsula: Assessment of the Fukushima fallout

L.N. Mikhailovskaya [a], I.V. Molchanova [a, 1], V.N. Pozolotina [b, *], Yu.N. Zhuravlev [c], Ya.O. Timofeeva [d], M.L. Burdukovsky [d]

[a] *Laboratory of Common Radioecology, Institute of Plant and Animal Ecology, Ural Branch, Russian Academy of Sciences, Vos'mogo Marta Str. 202, Ekaterinburg, 620144, Russian Federation*
[b] *Laboratory of Population Radiobiology, Institute of Plant and Animal Ecology, Ural Branch, Russian Academy of Sciences, Vos'mogo Marta Str. 202, Ekaterinburg, 620144, Russian Federation*
[c] *Laboratory of Biotechnology, Institute of Biology and Soil Science, Far East Branch, Russian Academy of Sciences, pr. 100-letiya Vladivostoka 159, Vladivostok, 690022, Russian Federation*
[d] *Biogeochemistry Sector, Institute of Biology and Soil Science, Far East Branch, Russian Academy of Sciences, pr. 100-letiya Vladivostoka 159, Vladivostok, 690022, Russian Federation*

A R T I C L E I N F O

*Article history:*
Received 30 August 2016
Received in revised form
8 February 2017
Accepted 10 February 2017

*Keywords:*
Radionuclides
Global fallout
Accident at the “Fukushima” NPP
Primorsky Krai
Sakhalin Island
Kamchatka Peninsula

A B S T R A C T

The contamination densities of soil–plant cover at certain locations within the Primorsky Krai, Sakhalin Island and Kamchatka Peninsula attributable to $^{90}$Sr, $^{137}$Cs and $^{239,240}$Pu were 500–1390 Bq m$^{-2}$, 980–2300 Bq m$^{-2}$ and 37–74 Bq m$^{-2}$, respectively. These values do not exceed average global background levels, typical for mid-latitudes of the Northern Hemisphere. The spatial distribution of radionuclides depends on the climatic conditions of the region. A positive dependence of the $^{90}$Sr and $^{137}$Cs contamination densities, as well as additional $^{137}$Cs from NPP “Fukushima” in the soil, was determined based on the sum of annual atmospheric precipitation within the study areas. No trends in the spatial distribution of Pu isotopes were observed. The $^{137}$Cs contribution from the “Fukushima” NPP constitutes 11–300 Bq m$^{-2}$ in the Primorsky Krai, Sakhalin Island and at the Kamchatka peninsula, i.e., 1–22% of the total amount of radionuclides in the soil. The contribution of this radionuclide to the contamination of moss–lichen vegetation ranged from 7 to 42%.



## 1. Introduction

Nuclear weapons tests and accidents at nuclear fuel cycle enterprises have led to global radioactive contamination of the biosphere by long-lived artificial radionuclides: $^{90}$Sr, $^{137}$Cs and $^{238,239,240}$Pu. Study of the formation and incorporation of radionuclides into the biological cycling of matter is a topical problem within radioecology. Assessment of the radioecological situation in the Far Eastern region of Russia is important because of the area's proximity to numerous radioactive sources. These include 13 nuclear power plants (NPP) situated on the territories of neighbouring states, as well as a number of potentially dangerous objects under the administration of the Russian Federation's Ministry of Defense. We know of few data describing the levels of radioactive contamination of the natural ecosystems of the Primorsky Krai, Sakhalin Island, Kuril Islands and the Kamchatka Peninsula (Onischenko et al., 2011; Ramzaev et al., 2013; UNSCEAR, 2013). After the accident, which occurred in March 2011, fallout $^{134}$Cs, $^{137}$Cs and $^{131}$I were registered everywhere in the Northern Hemisphere (Bossew et al., 2012; EPA, 2011; Kirchner et al., 2012; Masson et al., 2011), including the different regions of Russia (Bolsunovsky and Dementyev, 2011; Bulgakov et al., 2011; Molchanova et al., 2013; Onischenko et al., 2011; Ramzaev et al., 2013). The objectives of this study were: 1) the study of the distribution of long-lived artificial radionuclides $^{137}$Cs, $^{90}$Sr, $^{239,240}$Pu in soil-vegetable cover at certain locations of the Primorsky Krai, Sakhalin Island and the Kamchatka Peninsula; 2) assessment of the $^{137}$Cs and $^{134}$Cs fallout from accidental NPP

* Corresponding author.
*E-mail address:* pozolotina@ipae.uran.ru (V.N. Pozolotina).
[1] Dr. Inna Molchanova has died.

http://dx.doi.org/10.1016/j.jenvrad.2017.02.006

"Fukushima" in contamination of the basic components of terrestrial ecosystems in these areas.

## 2. Materials and methods

### 2.1. The study site

Investigations were carried out during the period from 2012 to 2013 in the Primorsky Krai territory, which includes the Muravyov–Amursky Peninsula and Popov Island, as well as Sakhalin Island and the Kamchatka Peninsula. The position of the sites is shown in Fig. 1. The length of the study area in the latitudinal direction was more than 2000 km (42°–56° N, 131°–162° E). The predominant forms of relief at the Primorsky Krai are the remnant low hills, at the foot of which, as a rule, dealluvial shelves have formed. Mountain-valley terrain of the Sakhalin Island causes a high diversity in natural conditions. The main feature of the Kamchatka Peninsula is the presence of 160 volcanoes, including 28 active volcanoes. Plains and lowlands are situated here only in the river valleys and occupy small areas (Ivlev, 1977; Vyatkina et al., 2014).

The climate of the south-eastern region of Russia is monsoon. In the Primorsky Krai (Vladivostok City) annual amount of precipitation is estimated at 600–900 mm. At the Sakhalin Island, these values range from 500 to 700 mm in the northern part to 800–1200 mm in the southern part. At the Kamchatka, the amount of precipitation is also reduced in the latitudinal direction from 1200 mm in the south-east to 350 mm in the north-west of the peninsula. The average daytime temperatures increase in the direction from north to south (3.9° C in Petropavlovsk-Kamchatsky; 6.4 °C in Yuzhno-Sakhalinsk; 8.5 °C in Vladivostok; see http://meteoinfo.ru/.

Broad-leaved and coniferous–deciduous forests dominate in the Primorsky Krai. The prevailing type of vegetation in most parts of the Sakhalin Island is dark coniferous spruce-fir taiga. Kamchatka has well expressed high-altitude zones of vegetation, the spruce and larch forests, stone-birch forests, deciduous forests with admixture of alder and dwarf Siberian pine are widespread (Seledets, 2011; Vyatkina et al., 2014).

In the southern part of the Far Eastern region under deciduous forests, highly humus brown soils (Udepts, Udalfs) have formed with a powerful undifferentiated profile. The soils have a slightly acidic and neutral pH. Burozems (Udepts) are widely-spread also within the southern and central parts of the Sakhalin Island. Poor humidity, acidic podzolic soils (Orthods) predominate in the northern part of the island on the elevated landforms of coastal areas, on sandy and sandy loam deposits. They have a sharply differentiated profile, the depth is from 40 to 60 cm (Ivlev, 1977). In the low-lying areas of the sampling sites on the territory of southern part of the Russian Far East and the Sakhalin Island slightly acidic soils were formed with increased accumulation of clay fractions in the middle parts of the soil profiles. Gley features (olive and whitish mottles) are present in the soil profile, which indicates long-term soil water-logging. These soils are classified as Aquepts according to U.S. Soil Taxonomy (Staff, 1999). Coastal marine lowlands of the eastern coast of the Kamchatka are under influence of moderate ash falls. The ochre and layered volcanic ochre soils (Cryands) dominate here. Soils have polygenetic profiles with layers of coarse volcanic sand, characterized by high humus content (up to 10%). The proluvial meadow-sod soils formed in the valleys under forb meadows. The buried humus horizons are found often in soil profiles, they were formed as a result of water flows. These soils are characterized by slightly acidic reaction; the humus content varies from 5 to 8.5% (Vyatkina et al., 2014). Upper Cretaceous effusive rocks dominate in the studied areas. Their derivatives (such as granites, sandstones, siltstones, gravelites, conglomerates, lenses siliceous-clayey shales et al.) serve as soil-forming material.

### 2.2. Sampling methods

Study plots were located on flat parts of the relief. They had a uniform topography and soil–plant cover. Brief descriptions of plots are given in Table 1. Soil profiles were located at the vertices of an equilateral triangle with a side length of 10 m. We collected from each cut the samples of litter, picking out the fresh litter (horizon L, sample area was 1.0 $m^2$), and the mineralized part (horizons F + H, sample areas were 0.4–1.0 $m^2$). Soil samples were taken as layers with 5 cm capacity to a depth of 20 cm (sample areas were 0.01–0.04 $m^2$). Total content of radionuclides in the 20-cm soil layer, was normalized to a unit area (i.e. 1 $m^2$), and is referred to as the contamination density. We averaged the samples of three individual profiles located at the vertices of the triangle to create a representative sample. In the vicinity of the profiles, the samples of above-ground mass of herbaceous plants, biomass of mosses and epiphytic lichens were taken. All samples were dried to a constant dry weight. Vegetable samples were crushed. Soils were triturated, riddled through a sieve (cell diameter was 1 mm). Then all samples were incinerated at t = 450° C.

### 2.3. Methods for determination of radionuclides

All analytical works were carried out in accredited laboratories IPAE UB RAS (accreditation certificate CAPK RU.0001.441492). The content of $^{137}$Cs and $^{134}$Cs in selected samples was determined by gamma-ray spectrometry with a germanium semiconductor detector "ORTEC" (USA) with a detection limit of 0.1 Bq. Gamma-ray peaks used in the measurements were 604 keV for $^{134}$ Cs and 662 keV for $^{137}$Cs. The counting time was 30000-170000 s (certified geometry: Petri dish of 80 $cm^3$, vessel Marinelli of 1000 $cm^3$ or the vessel Marinelli, filled with half).

The content of $^{90}$Sr was determined by radiochemical method (Tsvetaeva et al., 1984). The procedures based on the radionuclide leaching by 6 N HCl-solution from preliminary prepared samples. Precipitating of $^{90}$Sr in the oxalate form, separating of $^{90}$Sr kept in balance with the daughter product of decay – $^{90}$Y. Measurement of $^{90}$Y was carried out with the use of alpha-beta radiometer "UMF-2000" (Russia), detection limit of 0.2 Bq.

Plutonium content in samples was also determined by radiochemical method (Chen et al., 1993). This method includes leaching isotopes of plutonium by mixing concentrated acids ($HNO_3$+HCl) from preliminary prepared samples, precipitation on ion-exchange resin, then purification and stripping from the ion-exchange column by $H_2O$ and 0.5 N $HNO_3$ solution in succession, electrodepositing on disks made of stainless steel. Measurement of prepared specimen was carried out with the use of alpha-spectrometer "ORTEC" (USA). Lower limit of detection was 0.001 Bq. Procedural error of the methods did not exceed 20%. All results were calculated on air-dry weight of samples.

The contribution of $^{137}$Cs from Fukushima NPP accident to the pollution of the investigated areas was calculated by the content of $^{134}$Cs. The ratio $^{134}$Cs/$^{137}$Cs in gas-aerosol emissions at the time of the accident was 1.0 (Kirchner et al., 2012; Onischenko et al., 2011). All data are presented at the time of sampling.

We tested the statistical hypotheses using correlation analysis and non-parametric methods of Kruskal-Wallis ($H$) and Mann-Whitney ($z$) in the STATISTICA 8.0 software (StatSoft and Inc, 2007).