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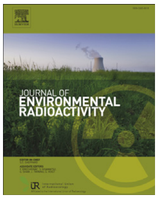

Distribution of artificial radionuclides in particle-size fractions of soil on fallout plumes of nuclear explosions

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ABSTRACT

In this paper are analyzed the artificial radionuclide distributions (^{137}Cs, ^{90}Sr, ^{241}Am, $^{239+240}$Pu) in particle-size fractions of soils from two radioactive fallout plumes at the Semipalatinsk Test Site. These plumes were generated by a low-yield surface nuclear test and a surface non-nuclear experiment with insignificant nuclear energy release, respectively, and their lengths are approximately 3 and 0,65 km. In contrast with the great majority of similar studies performed in areas affected mainly by global fallout where adsorbing radionuclides such as Pu are mainly associated with the finest soil fractions, in this study it was observed that along both analyzed plumes the highest activity concentrations are concentrated in the coarse soil fractions. At the plume generated by the surface nuclear test, the radionuclides are concentrated mainly in the 1000-500 μm soil fraction (enrichment factor values ranging from 1.2 to 3.8), while at the plume corresponding to the surface non-nuclear test is the 500-250 μm soil fraction the enriched one by technogenic radionuclides (enrichment factor values ranging from 1.1 to 5.1). In addition, the activity concentration distributions among the different soil size fractions are similar for all radionuclides in both plumes. All the obtained data are in agreement with the hypothesis indicating that enrichment observed in the coarse fractions is caused by the presence of radioactive particles resulted from the indicated nuclear tests.

1. Introduction

It is a well-known fact that formation of radioactive plumes due to nuclear tests is a consequence from production of radioactive particles, their transfer by air and their deposition to the earth surface at various distances from the explosion epicenter depending on test magnitude (Loborev, 1997). And one indicative parameter to assess the dispersion of local fallout generated by a nuclear test is the distribution of radionuclides among the different soil particle-size fractions. The interrelation between the distribution of radionuclides in the different sizes of the soil fractions and the dispersion of radioactive fallout particles have been evidenced in previous studies performed in areas affected by local fallout due to nuclear explosions.

In fact, in the epicenter of the "Trinity" nuclear explosion in New Mexico, US, the highest concentration of plutonium (0,2 Bq/g) in the topsoil was found associated to the sand size fractions (1–2 mm), while the concentrations for the same radionuclide in the clay fractions (<53 μm) were quite low (<0,004 Bq/g). Similar results were obtained analyzing soils collected at the epicenters of nuclear explosions at the Nevada test site, US, which can be explained by the deposition of larger plutonium particles in the vicinity of explosion epicenter (Hakonson, 2007). At more remote distances from the epicenter, for example at 45 km away from the epicenter of the «Trinity » explosion, is the fraction corresponding to <53 μm, the one significantly enriched with plutonium. This fraction contained over 70% of total plutonium content in the soil (Aleksahin, 1985).

Significant plutonium-bearing particles have been also observed in sites where low-yield, or safety non-nuclear tests have been performed, for example at the Nevada Test Site, US (Gilbert et al., 1988) and at the "safety-trials" tests at Maralinga Australia (Ikeda-Ohno et al., 2016; Johansen et al., 2014, 2016). In the case of non-nuclear tests, it has been observed that the Pu was deposited in fine particulate form and most radioactivity is associated to small soil fractions (to the 20–53 μm fraction at Nevada test site, to the

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<7 μm fraction at Maralinga test site).

The interrelation between the presence of radioactive particles and the distribution of radionuclides in the different grain-size soil fractions have been also observed in soils collected at the radioactive fallout plume generated by the surface thermonuclear explosion conducted in the 1953 at Semipalatinsk Test Site (the STS) (Shapiro, 2013). The obtained data shows that at a distance of 40 km from the explosion venue, the dimensions of radioactive particles ranged from 500 to 1200 μm while at a distance of about 100 km the soil fraction of 250–500 μm was the enriched one in radionuclides to a highest extent (Lukashenko, 2015).

Historically, the analysis of artificial radionuclide distribution in the different soil size fractions has been mostly devoted to nuclear tests with rather high yield, probably due to the fact that high yield explosions generate extensive radioactive contamination of territories. However, the analysis and characterization of the radioactive contamination from nuclear tests of low and ultra-low yield are equally topical for the Semipalatinsk Test Site (STS) from scientific and practical point of view. Studies of radionuclide distribution in particle-size fractions of soils in venues of surface nuclear and non-nuclear tests at STS will be of significant practical importance, especially under conditions of arid climate, for predicting radionuclide air contamination and inhalation hazard, and for evaluating wind transfer of radioactive substances, with the consequent implication in the application of remediation technologies at this territory. Also the data generated in this study can be related to the obtained ones in scenarios affected by nuclear weapons accident events such as Palomares (Spain) and Thule (Greenland).

About 20 out of 30 nuclear tests conducted at the “Experimental Field” technical site at Semipalatinsk had a low energy release (less than 1.5 kt in TNT equivalent). Moreover, at this site 40 surface non-nuclear (hydronuclear/hydrodynamic experiments) experiments with nuclear energy release comparable with the energy of high explosives were conducted. The experiments of this type have resulted in radioactive contamination of soil with transuranium radionuclides (Logachev, 2002; Moshkov et al., 2011). The work compiled in this paper form part of a research program that has as a main objective to analyze the mentioned contamination by studying the radionuclide distribution among a range of soil size fractions in the radioactive fallout plumes from low- and ultra-low yield surface tests conducted at the STS. The information gained in this work has a considerable practical importance because venues of surface nuclear explosions at the «Experimental Field » will have a priority in rehabilitation of the STS lands.

2. Material and methods

2.1. Objects of study

Two short plumes formed as the result of surface nuclear tests at the «Experimental Field » site of Semipalatinsk Test Site (the STS) were chosen to perform this study. Contours and geographical location of the selected plumes are clearly observed in the radioactive contamination map of the site (Nazarbaev et al., 2016). Selected objects are conditionally called “B-1” and “P” (Figs. 1 and 2).

The object “B-1” was formed as the result of a presumably low-yield surface nuclear explosion (1.2 kt) conducted on 30.10.1962. There is a crater in the epicenter of explosion, and the plume is extended in the south-eastern direction with a length of 3 km.

The object “P” was formed as the result of a presumably surface non-nuclear experiment. The approximate length of the generated plume is 650 m, and the exact date of this experiment is currently unknown. All the non-nuclear experiments at STS were conducted during the period between 13.03.1958 and 15.10.1963.

2.2. Research methodology

The work was performed in different steps: a) sampling and preparation of soil samples, b) particle-size fractionation of the soil samples and c) radionuclide activity concentration determinations in bulk soils and associated fractions.

2.2.1. Collection and preparation of samples

Samples were collected in the vicinity of the epicenter of the explosions and along the central line of the plumes with increasing distance from the epicenter. At the «B-1» object, in the epicenter area, two samples were taken (from the northern and the south-western side of the crater) (Fig. 1).

In each sampling point a small spot of 1–2 m^2 was selected from where the topsoil had been sampled until a depth of 5 cm. In each spot, 5 points located in the angles and in the center (“envelope” method) were used for the top soil collection, being the collected material combined into one general sample. Sampling was made in each of the 5 points of the spots by using a rectangular steel sampling scoop specifically made for the purpose. Scoop size were 5 cm high and 10 cm long and wide (5 × 10 × 10 cm).

The average weight of the combined sampled soil at each sampling point was 3–3.5 kg. After collection, the soil samples were dried in an oven at the temperature not exceeding 60 °C, and the coarse gravels and vegetative inclusions were removed from the dried sample. The dried samples were then sieved via sieve with 1 mm mesh size. The stony fraction of >1 mm was excluded from further research, while the fraction of <1 mm was exposed to further particle-size fractionation.

2.2.2. Particle size separation of soil

The techniques of «wet » sieving and sedimentation in a suspension were used for separation of soil into particle-size fractions.

The “wet” sieving technique was applied using 500, 250, 100, 63 and 40 μm mesh sieves stacked in a column with progressively larger openings towards the top. The column was placed in a mechanical shaker. The sample was placed in portions on the top sieve and washed carefully. After the sieving is complete the soil fraction on each sieve was collected in a separate porcelain cup, dried and weighed. Therefore, by using the «wet » sieving method the 1000-500; 500-250; 250-100; 100-63 and 63-40 μm fractions were separated. The fractions with particle size of less than 40 μm were subsequently separated by applying the sedimentation method.

The soil suspension collected after sieving was placed into several measuring cylinders of 1l volume (water column level was 352 cm), which were shaken until complete spreading of sediments from the bottom. The suspended material was then allowed to settle during the necessary period of time. The sedimentation times were 17 min, 1 h 10 min and up to 20 h 28 min and corrected depending on temperature of suspension according to a standard technique (GOST, 2015). Once sedimentation time was reached, the supernatant suspension was drained by decantation, and the sediment from the bottom was collected into a cup, dried and weighted. The decanted suspension was then carried back in the measuring cylinders, the water level was recovered and the abovementioned operations were repeated. The last fraction was separated by centrifugation.

The particle-size fractions of soil separated by sedimentation were analyzed via optical microscope in order to determine the size of particles. Size of the particles was measured using a micron level net. Optical microscope images of the analyzed fraction particles are shown in Fig. 3. From the obtained results, we can indicate that using the sedimentation method, the fractions 40-8 μm, 8-5 μm, 5-1 μm and <1 μm were separated.

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