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On the lognormality of radionuclide deposition

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ABSTRACT

The influence of the variation of soil density and the uncertainty of activity measurements on the statistical distribution of radionuclide concentrations on a site is considered. It is demonstrated that the influence of these factors adequately explains the observed deviation of radionuclide empirical probability distribution functions (empirical PDFs) from lognormal. In all probability lognormality of activity density distributions is the consequence of the atmospheric fallout process, as observed for deposition from Chernobyl and Fukushima. The results obtained are in no way specific to radioactive contaminants, and are consequently applicable for depositions of non-radioactive pollutants as well.

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1. Introduction

It is common knowledge that spatial structure of radioactive deposition represents a random geometrical multifractal field (Raes et al., 1991). In case of an old radioactive deposition, for example Chernobyl fallout, the smallest soil sites with multifractal structure have surface areas equal approximately to 1 m², with radionuclide distribution on such sites described by a lognormal distribution (Grubich, 2012). As a consequence, radioactive deposition can be represented as a mosaic formed by small sites, each having a lognormal distribution of activity. As a result, on any site formed by an arbitrary totality of such small sites, radionuclide distribution is best described by a lognormal distribution (Grubich, 2014). The above-said is valid both for activity density (Bq/m²) and activity concentration (Bq/kg).

These properties of radioactive deposition are probably caused by the atmospheric fallout process. Still, there are also other physical factors which, in principal, may influence the shape of empirical (observed) distributions of radionuclides:

- Transport of radionuclides in soil causing gradual change of radionuclide deposition.
- Distribution of soil density on site.
- Uncertainty (random error) of activity measurement, etc see (ITRC, 2006).

The thing in common for the above-listed factors is that they all can be called "secondary", as actually they do not influence the initial distribution of radionuclides of thin radioactive "film" formed by atmospheric fallout on the soil surface.

The main objective of this article is to study the influence exerted on the shape of radionuclide empirical distribution by two secondary factors: distribution of soil density on the site and uncertainty of activity measurements. As far as I know, no previous quantitative analysis has been carried out where the influence of these factors on the shape of empirical distributions of density and/ or activity concentration was specifically considered.

The results obtained in this article explain deviations, observed in a number of cases, of empirical distribution shape from lognormal (see, in particular, plots in Fig. 6b and c in Grubich et al. (2013)). The deviations were also exemplified in the review (Daniels and Higgins, 2002). The analysis of the impact of secondary factors on the empirical distribution function shape is of both theoretical and practical interest, as it makes possible determined a "true distribution" for thin radioactive film formed by atmospheric fallout.

2. Data

This article uses data for 90 Sr and 241 Am activity density and activity concentration on site P4, described earlier in Grubich et al. (2013) – see datasets No 27, 28 and 57, 58 in Table 1 of the said work. Here, only the information about site P4 and its radioactive contamination that was not published earlier is given. It should also

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Tabl	e 1	
Test	statistic	

Dataset	Nuclide	Value	Distribution	Test		
				CvM	AD	KS
27	90Sr	x (Bq/m ²)	$\Lambda(\mu, \sigma^2)$	0.072	0.646	0.624
			F _φ	0.040	0.438	0.531
57		y (Bq/kg)	$\Lambda(\mu, \sigma^2)$	0.127	0.863	0.701
28	²⁴¹ Am	x (Bq/m ²)	Λ(μ, σ ²)	0.046	0.293	0.590
58		y (Bq/kg)	Λ(μ, σ ²)	0.409	2.47	1.23
			Fω	0.145	0.835	1.04

be pointed out that in this article, unlike in Grubich et al. (2013), different notations for activity density and activity concentration are used: x - activity density, y - activity concentration.

2.1. Soil density

Site P4 with surface area 34 km² had areas with different types of soils (from peaty soils to soddy-podzolic-sandy soils). Due to this circumstance soil dry bulk density varied in wide range of values from 0.2 kg/L to 1.85 kg/L (a metal sampler that gave 20-cm-deep cores was used for sampling). The mean and coefficient variation of soil density were $\rho_0 = 1.19$ kg/L and CV $\rho = 0.320$ correspondingly. The histogram of soil density on site P4 is shown in Fig. 1. Axis of ordinates in Fig. 1 (and Fig. 2) is value of frequency, v, divided by bin width of histogram. Fig. 1 also shows results of fitting to the histogram of probability density functions $g(\rho)$ for three types of distributions: normal, beta and Gumbel distributions. As can be seen, soil density distribution on site P4 is best described by Gumbel distribution. Further on, instead of soil volumetric density variable ρ , we shall use soil surface density variable, corresponding to it,

$$\mathbf{z} = \boldsymbol{\rho} \cdot \mathbf{h},\tag{1}$$

where h = 20 cm - thickness of soil surface layer from which an increment was sampled.

2.2. Radionuclide distribution

In Grubich et al. (2013) it was shown that datasets considered in this article are best described by lognormal distribution $\Lambda(\mu, \sigma^2)$. Numeric values of the test statistic obtained for lognormal distributions are given in Table 1. Calculations were made for several types of tests: the Cramér–von Mises test (CvM), the Anderson–Darling test (AD) and the Kolmogorov–Smirnov test (KS).

Fig. 2 shows histograms of ⁹⁰Sr activity density (dataset 27) and ²⁴¹Am activity concentration (dataset 58), as well as functions of



Fig. 1. Histogram of soil density on site P4.



Fig. 2. Histogram: (a) - ⁹⁰Sr activity density; (b) - ²⁴¹Am activity concentration.

lognormal distribution probability density, which best describe these datasets (continuous curve). Fig. 3a and c shows plots for probability

$$\Pr\{X > x\} = [1 - F(x)], \tag{2}$$

where F(x) – distribution function (cumulative distribution function) of activity density.

In Fig. 3a and c dots represent probability values (2) for empirical distribution function.

$$Fn(X(i)) = (i - 0.5)/n,$$
 (3)

where $x_{(i)}$ – order statistics ($x_{(1)} \le x_{(2)} \le ... \le x_{(n)}$), corresponding to activity density dataset with sample size n (i = 1, 2, ..., n).

Plot of probability $Pr\{X > x\}$ for lognormal distribution $\Lambda = \Lambda(x|\mu, \sigma^2)$, best describing the dataset, is shown in Fig. 3a and c by heavy gray curve.

Finally, Fig. 3b and d shows similar diagrams for probability

$$\Pr\{Y > y\} = [1 - F(y)], \tag{4}$$

where F(y) – distribution function of activity concentration.

3. Methods

3.1. The quotient of random values

The relation between activity density x (Bq/m^2) and activity concentration y (Bq/kg) is given by

$$\mathbf{y} = \mathbf{x}/\mathbf{z},\tag{5}$$

where z - soil surface density (1) in units of kg/m².

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