

The influence of soil density and the character of radioactive ^{134}Cs and ^{137}Cs pollution's distribution on *in situ* measurements

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Abstract

Periodic radiation monitoring of soils today is a priority task not only for Belarus, but also for Japan, suffered by Fukushima nuclear power plant accident. Use of portable and light spectrometers with ability to perform *in situ* measurements makes it possible to quickly estimate specific activity of measured radionuclides with required accuracy in particular soil site. Basic information of a gamma radiation source (radionuclides content, effective radius of measurement area and thickness of contaminated layer) can be obtained directly during measurement. The purpose of this research is to test the feasibility of using algorithms for determination of specific activity and thickness of contaminated layer under conditions of soil measurement with variable density parameters and radiocesium distribution in soil.

Monte-Carlo simulating allowed to estimate the degree of deviation of the shape of simulated spectra obtained with the use of Monte-Carlo soil model with uniformly distributed radionuclide in it, and for the case when the radionuclide distribution by soil profile can be described by an exponential function. For these cases of natural distribution of radiocesium, the pulse-amplitude spectrum is formed by an effective thickness of the contaminated site, which contains more than 90 % of radionuclides.

The developed Monte-Carlo model of a probe and contaminated soil site allows to estimate the effect of the variability of soil density on the total count rate of the pulse-amplitude spectrum. As a result of theoretical estimations, the relationship between the effective radius of contaminated site is determined as a function of soil density.

Analysis of the influence of radial zones of the cylindrical gamma source on *in situ* gamma-spectrometer showed that the main contribution to the total count rate of the pulse-amplitude spectrum is made by the radial zone with radius of up to 40 cm from the center of the probe, regardless of the thickness of the contaminated layer in geometry «Probe is located on the soil surface». A small site facilitates the selection of measurement area of land with a sufficiently flat surface, which is desirable during surveying the territories, especially with complex terrain.

Keywords: *in situ* gamma-spectrometer, contaminated soil layer, non-uniform distribution, effective thickness, density of soil.

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Влияние плотности почвы и характера распределения радионуклидов ^{134}Cs и ^{137}Cs по профилю при *in situ* измерениях

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Проведение периодического радиационного мониторинга почв на сегодняшний день является одной из приоритетных задач для обеспечения радиационной безопасности не только в Беларуси, пострадавшей от Чернобыльской катастрофы, но и в Японии, территории которой подверглись радиоактивному загрязнению в результате аварии на АЭС Фукусима. Цель настоящей работы заключалась в проверке возможности применения разработанных на основе упрощенной («равномерной») модели алгоритмов определения активности и толщины загрязненного слоя в условиях радиометрии почвенного покрова с вариативными параметрами плотности и распределения радиоцезия по глубине.

Использование портативных *in situ* спектрометров позволяет оперативно оценить удельную активность контролируемых радионуклидов и плотность загрязнения почвы прямо на месте измерения и с необходимой точностью. Основная информация об источнике гамма-излучения (присутствующие радионуклиды, эффективный радиус участка и толщина загрязненного слоя) может быть получена непосредственно по результатам измерения аппаратурного спектра в сравнении с теоретическими (калибровочными) спектрами. Теоретические спектры рассчитывают путем имитационного моделирования процесса *in situ* измерений активности радионуклидов ^{134}Cs и ^{137}Cs , равномерно распределенных в однородной почвенной среде постоянной плотности. На практике следует учитывать приближенность принятой модели измерений в отношении реального профиля заглубления радиоактивных загрязнений и изменчивости плотности исследуемых почв.

Анализ влияния радиальных зон цилиндрического источника на интегральную скорость счета спектрометра показал, что основной вклад вносят радиальные зоны в радиусе до 40 см от центра устройства детектирования при расположении его на поверхности почвы вне зависимости от толщины загрязненного слоя. Небольшая по площади зона влияния облегчает выбор контролируемых участков земли с достаточно плоской поверхностью, что желательно при обследовании территорий, особенно со сложным рельефом.

Ключевые слова: *in situ* гамма-спектрометр, загрязненный слой почвы, неравномерное распределение радионуклида, эффективная толщина, плотность почвы

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Introduction

The soil, together with atmosphere air and natural waters, is a main subject for radiation and environmental monitoring [1, 2]. Portable gamma spectrometers with the probe, located above the soil (usually at 1 m height) [3–6] or on its surface, are being developed for the purposes of immediate survey of soil radioactive contamination [3, 7]. An important condition for the reliability and validity of *in situ* techniques of radionuclides activity determination in soil is to take into account the effect of its density and thickness of contaminated layer, as well as the distribution profile of radioactive substances in soil [3, 7, 8].

The described in [7] principles of the analysis of experimental spectra allows to simultaneously obtain information on the content and thickness layer with gamma-emitting ^{134}Cs and ^{137}Cs nuclides, expecting their homogeneous distribution over the contaminated layer. The algorithm for determination of specific activity and thickness of the contaminated layer is based on comparative analysis of experimental and simulated spectra of cesium isotopes obtained for a scintillation gamma spectrometer [7]. Simulated spectra were calculated using the Monte-Carlo (MC) method for the NaI(Tl) detector ($\varnothing 63 \times \varnothing 63$ mm) in the model of radiometric measurements of the contaminated site of the soil, represented as a cylindrical source with uniformly distributed ^{134}Cs and ^{137}Cs isotopes. The elemental composition (the effective atomic number $Z_{\text{eff}} \approx 10$) and the density (1.5 g/cm^3) of the model soil are chosen with maximum correspondence to average parameters of soils of Tohoku region (Japan) and Gomel region (Belarus) [2, 9–11]. For typical soils in areas contaminated by the Chernobyl and Fukushima nuclear accidents, the Z_{eff} values are almost the same in a wide range of densities from 1.0 to 1.8 g/cm^3 [2, 9, 10].

The purpose of this research is the verification of the application of the algorithm, proposed in [7], for determination of specific activity and thickness of the contaminated layer under conditions of soil radiometry with variable density parameters and radiocesium distribution by soil profile.

Materials and methods

The researches were based on simulation of the radiometric MC model [7], developed according to the purpose of the work and parameters of the probe. MCNP 4B software was used for simulating [12]. The model of the measurement soil vol-

ume, as in [7], is a cylindrical source located in a semi-infinite isotropic environment with the radioactively contaminated layer of the fixed thickness. Gamma radiation of contaminated soil site was registered by the detector located on the middle of the upper surface of a cylindrical source. The studied model, unlike the prototype, is not limited to one average value of the soil density and the requirement of uniform distribution of cesium radionuclides over a layer of fixed thickness and can be used to assess the uncertainty of measurement *in situ* with variable parameters.

The height of the cylindrical source d is equal to the thickness of contaminated layer. The field of-view of the probe characterizes the radius r . In MC modeling, the number of emitted particles with a given energy should be distributed uniformly over the cylinder. With an increase in the radius r , the fraction of the photons emitted from regions of measurement area located far from the detector (and also the simulation time) increases rapidly, but the chance of these photons to contribute to the energy distribution of the pulse-height spectrum (F8 tally) becomes less and less important. Therefore, when modeling an extended (including infinite) source, the size of the measurement area should be restricted by some effective radius r_{eff} , at which the energy distribution of the pulse-amplitude spectrum $I(E)$ will be close to experimental one for the real object.

To determine the effective radius is need to use acceptable relative deviation β . By definition, the quantity β is written in the form:

$$\beta = \frac{I_{FEP}(\infty) - I_{FEP}(r)}{I_{FEP}(\infty)}, \quad (1)$$

were $I_{FEP}(r)$ – the count rate the range $\pm 3\sigma$ of the full energy peak (FEP) for the source of given radius r ; $I_{FEP}(\infty)$ – the count rate in the same range for the source of infinite radius.

Value of r can be considered as the effective radius $r = r_{\text{eff}}$ of the source corresponding to the relative deviation β .

The areas behind the circle with radius $r > r_{\text{eff}}$ make an insignificant contribution to the value $I_{FEP}(r)$, since the probability of a gamma-quantum reaching from them to the detector, even after scattering in the soil and air, is negligible. At $\beta = 10\%$, a source with a radius r_{eff} provides 90 % of the total count rate in energy range from 50 to 3000 keV of simulated spectrum of contaminated with cesium ra-

dionuclides soil layer with thickness d in the definite geometry of measuring.

The value of the effective radius for a uniformly contaminated soil layer is mainly determined by its density and energy gamma radiation of the measured radionuclide, and weakly depends on the elemental composition of the substance [13, 14]. For radionuclide with several gamma-ray energies, the value r_{eff} is set by the most intense one. For nearby energies of the main gamma-ray energies of ^{134}Cs and ^{137}Cs in the range 550–900 keV, the effective radius values will not differ significantly. In further analysis, an isotope ^{137}Cs with gamma-ray energy of 662 keV was used to simplify the simulation in order to obtain simulated spectra.

In cases, observed in practice, the depth of distribution of radioactive pollution into the soil does not exceed 20–25 cm [2, 15, 16]. During natural migration, the content of radionuclides usually decreases in depends on depth [2, 15–17]. The process of radionuclides redistribution by profile depends on many factors: soil type and density, time since radioactive contamination; agricultural activities, quantity of clay content, etc. Over the time, the maximum content of radionuclides along the soil layers may vary inland. Even within a relatively small area, cesium isotopes can be concentrated both in comparatively thin (2–3 cm) and thick (over 10 cm) layers [17–19]. To take into account a non-uniformly distribution of radiocesium by soil profile, it is necessary to introduce a concept «effective depth» or «effective thickness» of the contaminated soil layer.

The non-uniformly distribution of radionuclides in the soil and the effective thickness of the contaminated soil layer

For soil model with non-uniformly distributed ^{137}Cs isotope, only few cases from the set of possible radionuclide distributions along the profile were considered [11, 19]. Radionuclide content Q was distributed from the surface into the soil on depth up to 3 cm, up to 5 cm, up to 7 cm and up to 12 cm according with dependences presented in Table 1.

The situation when the layer contaminated with the radionuclide was located under a «clean» cover of the soil was also researched. In this case, a clean environment was modeled between probe and contaminated soil layer with a soil layer of a similar elemental composition 1, 2, and 3 cm thick, in which photon starts were not set.

Table 1
Relative radionuclide content Q in the top and lower layer of the soil profile

The thickness of the soil layer with a non-uniformly distributed radionuclide	The content of ^{137}Cs in the top layer, (0-1 cm), %	The content of ^{137}Cs in the lower layer, %	Exponential dependence of radionuclide content by soil profile
up to 3 cm	65.4	9.3	$Q=173.8e^{-0.977d}$
up to 5 cm	55.0	2.5	$Q=112.7e^{-0.757d}$
up to 7 cm	48.0	1.0	$Q=87.19e^{-0.630d}$
up to 12 cm	38.0	0.2	$Q=58.60e^{-0.464d}$

The radiocesium experimental spectrum from contaminated soil during natural migration of the nuclide into the soil will differ in shape from the «equivalent» spectrum with uniform distribution of the nuclide in the soil layer of the same thickness. The MC simulating results showed that for simulated spectrum, which calculated for a non-uniformly distribution of the radionuclide in the soil layer to a defined depth (hereinafter, the spectrum S_{un}), some equivalent soil thickness with uniform radionuclide distribution can be found, in which an identical simulated spectrum is formed (hereinafter, the «equivalent» spectrum S_{eq}) (Figure 1).

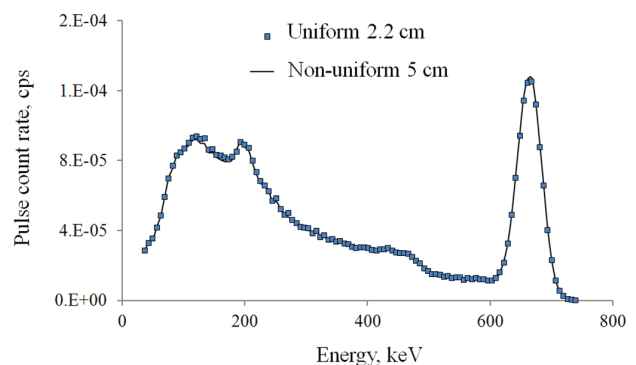


Figure 1 – Spectrum S_{un} with non-uniformly distributed ^{137}Cs and its «equivalent» spectrum S_{eq} with uniformly distributed ^{137}Cs

The deviation of the FEP heights with the gamma-ray energy spectrum of 662 keV S_{un} relatively to the «equivalent» spectrum S_{eq} , cited to mass unit considering the value of equivalent thickness of contaminated layer is presented in Table 2.

In the process of MC simulating of spectra, more than 90 % of photon starts were distributed in the thickness of the source commensurate to the thickness of «equivalent» spectra. Thus, more than 90 % of the experimental spectrum will be formed due to contaminated layer of effective thickness equal to thickness of uniformly contaminated soil

layer, which ensures the formation of an «equivalent» spectrum S_{un} .

Table 2

Deviation of FEP height with gamma radiation energy of 662 keV spectra S_{un} relatively to «equivalent» spectra S_{eq}

The thickness of the soil layer with a non-uniformly distributed ^{137}Cs along the profile (Table 1), cm	Thickness «clean» soil layer above the contaminated layer, cm	The thickness of the soil layer with an uniform distribution ^{137}Cs , cm	The ratio of the FEP height, %
3	0	2	-5.2
	1	4	2.3
	2	6	10.1
5	0	2	3.1
	1	5	-1.7
	2	7	0.7
7	0	4	-9.1
	1	6	0.9
	2	8	3.3
12	0	4	-5.2
	1	6	5.0
	2	10	1.2

The influence of natural distribution of radionuclides into the soil on shape and intensity of experimental spectrum can be considered as unissued component of the systematic error in measuring by the methodological properties. According the data in the Table 2, for *in situ* measurements in geometry «Probe is located on the soil surface» the indicated error will not exceed 10 %.

The effect of soil density on *in situ* measurements

The results presented above, as well as the results of the MC simulation in [7], were obtained with an average homogeneous medium density of

1.3 g/cm³. In real conditions, the typical range of soil density is in the range from 1.1 g/cm³ to 1.8 g/cm³ with a tendency for increase by depth [2, 16].

In order to evaluate the effect of changes in soil density in the range from 1.3 g/cm³ to 1.8 g/cm³ on the count rate I_{FEP} in FEP region ($\pm 3\sigma$), simulated spectra of the ^{137}Cs radionuclide for different thicknesses of the contaminated layer were estimated. Figure 2 shows the results of MC simulations showing the inverse dependence of the effective radius r_{eff} on the soil density for equal values of the parameter β .

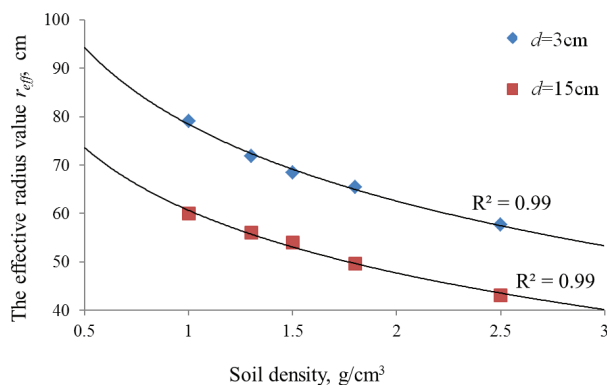


Figure 2 – Dependence effective radius vs. soil density

The component of the error of *in situ* measurements caused by the variability of the soil density, was estimated from simulated spectra calculated using the effective radius dependencies on the density in the range from 1.3 g/cm³ to 1.8 g/cm³ (see Figure 2). Table 3 shows the calculated data reflecting the nature of the change in the effective radius and count rate in FEP range $\pm 3\sigma$ depending on the soil density.

It was found that in the range of soil densities from 1.3 g/cm³ to 1.8 g/cm³, the count rate I_{FEP} in the FEP region $\pm 3\sigma$ do not change by more than $\pm 5\%$ compared to I_{FEP} obtained with an average density of 1.5 g/cm³.

The density of the soil by profile ρ_{pr} is not constant and, as a rule, varies in the direction of increase, depending on the distribution [16]. Using MC simu-

Table 3

Effective radius values and changing of count rate in FEP range, depending on soil density

The density of the soil, g/cm ³	1	1.3	1.5	1.8	2.5
The effective radius values for the thickness of the contaminated soil layer $d = 3$ cm, cm	82	73	69	63	56
The deviation of the count rate I_{FEP} relatively to the ones at soil density of 1.5 g/cm ³ ($d = 3$ cm), %	-15.2	-4.0	0.0	3.5	22.3
The effective radius values for the thickness of the contaminated soil layer $d = 15$ cm, cm	62	57	54	49	46
The deviation of the count rate I_{FEP} relatively to the ones at soil density of 1.5 g/cm ³ ($d = 15$ cm), %	-9.0	-3.1	0.0	2.3	5.8

lation, the variation of the count rate I_{FEP} was evaluated as a function of the variety in soil density by profile. The soil density varied in the range from 1.1 g/cm³ to 1.8 g/cm³ in increments of 0.2 g/cm³ per one centimeter of the soil profile. The radionuclide was distributed non-uniformly to depth of up to 5 cm and up to 7 cm (Table 1) by soil profile, with «clean» soil layer 1 cm thick located between the probe and the contaminated soil layer (Figure 3).

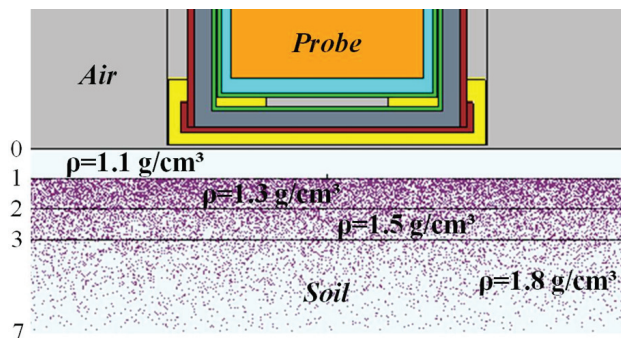


Figure 3 – Monte-Carlo model of the probe and the soil with different density by profile. The «clean» 0–1 cm layer of soil was located between the probe and contaminated layer 1–7 cm.

Figure 4 shows the simulated spectra of S_{ev} with the ¹³⁷Cs distribution depth up to 5 cm and up to 7 cm, their «equivalent» spectra S_{un} and simulated spectra obtained using MC soil model with a different density profile ρ_{pr} with «clean» top (0–1 cm) layer.

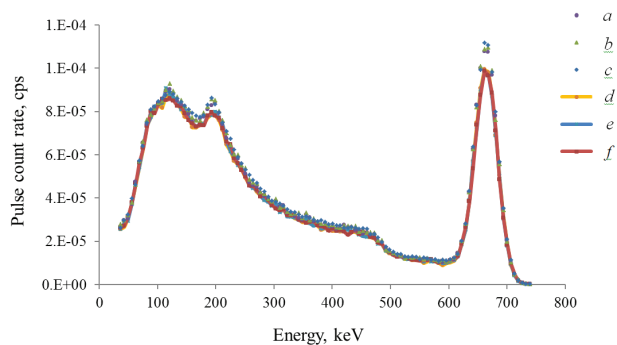


Figure 4 – Simulated spectra of the ¹³⁷Cs radionuclide in the soil: *a* – the spectrum S_{ev} ($d = 5$ cm) with «clean» soil layer, $\rho_{pr} \neq \text{const}$; *b* – the spectrum S_{ev} ($d = 5$ cm) with «clean» soil layer, $\rho_{pr} = \text{const}$; *c* – the «equivalent» spectrum S_{un} ($d = 5$ cm), $\rho_{pr} = \text{const}$; *d* – the spectrum S_{ev} ($d = 7$ cm) with «clean» soil layer, $\rho_{pr} = \text{const}$; *e* – the spectrum S_{ev} ($d = 7$ cm) with «clean» soil layer, $\rho_{pr} \neq \text{const}$; *f* – the «equivalent» spectrum S_{un} ($d = 6$ cm), $\rho_{pr} = \text{const}$

The heights deviations of the FEP for contaminated layers up to 5 cm and up to 7 cm with a different soil density compared to simulated spectra from a source with a density fixed at the profile of 1.5 g/cm³ are within $\pm 1\%$.

Contributions of radial source zones to the total count rate of the pulse-amplitude spectrum

The results presented above show that in during radiation monitoring of soils in the measurement geometry «Probe is located on the soil surface», the area of the cylindrical source, which provides the formation of 90 % of the total count rate of the pulse-amplitude spectrum, is limited to a circle with a radius of no more than 1 m regardless of soil density and nature of distribution of radiocesium along the profile.

In assessment of the effect of the sections of a cylindrical gamma-radiation source on the total count rate, the effective volume of the soil was considered as a set of radial zones – one central cylinder with a diameter of 10 cm and six coaxial cylinders, nested inside each other with an internal r_{in} from 10 to 60 cm and external $r_{out} = (r_{in} + \Delta)$ cm radii, where Δ is the width of the coaxial cylinder equal to 10 cm. The height of the cylinders was equal to the thickness of the contaminated layer d . When simulated spectra were calculated, photon starts were distributed only in the investigated radial zone, which subsequently moved from the center to the edge of the field-of-view of the probe limited by the effective radius r_{eff} . The spectra were calculated for the thickness of the contaminated layer $d = 5$ cm and $d = 15$ cm. The main contribution to the total count rate within a circle with a radius of r_{eff} regardless of the thickness of the contaminated source layer, provides the radial zones, located directly next to probe in a radius of 40 cm. Analysis of MC simulation results in the form of a distribution of the relative contributions of radial zones to the total count rate within the region limited by r_{eff} is shown in Figure 5.

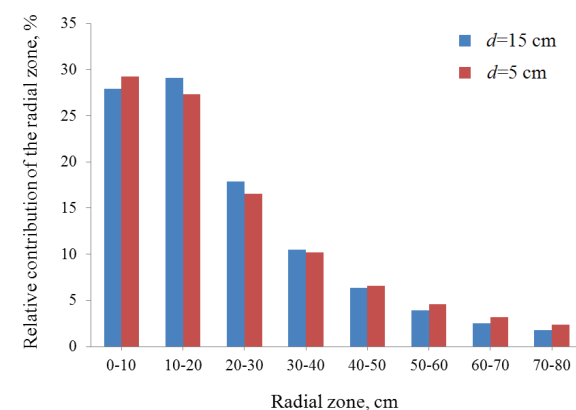


Figure 5 – Relative contribution of radial zone to the total count rate

For practical implementation of *in situ* soil measurements, it is not difficult to choose a soil

area whose surface will fully correspond to the adopted model of a cylindrical gamma-radiation source with a sufficiently flat surface. Wherein, there is no need to take into account the influence of trees, structures and other objects of the environment located at a distance of more than 2 meters. In addition, non-uniform ground at a distance of more than 40 cm from the center of the detector will not have a significant effect on the results of *in situ* measurements of the specific activity of radiocesium.

Summary

The results of MC simulating showed the efficiency of *in situ* measurement of soil with undefined density parameters and distribution of radiocesium in depth, using the algorithm of determine the thickness of the contaminated layer and the effective volume, which is based on comparative analysis of the experimental spectrum and simulated spectra.

The FEP count rate varies within $\pm 5\%$ for the soil density range from 1.3 g/cm^3 to 1.8 g/cm^3 relatively to the count rate at soil density of 1.5 g/cm^3 . This deviation ($\pm 5\%$) can be considered as the non-excluded systematic error of the *in situ* measurement in geometry «Probe is located on the soil surface».

According to results of MC simulating, the spectrum, obtained for ^{134}Cs and ^{137}Cs isotopes with arbitrary distribution in the soil, has coincidences in form and intensity simulated spectrum of the same radionuclides, uniformly distributed in a layer of some thickness. Formation of the experimental spectrum at *in situ* measurements of contaminated soils with ^{134}Cs and ^{137}Cs radionuclides with a natural distribution by profile is provided by an effective thickness layer, containing more than 90 % of radiocesium. The methodical error of determining the specific activity of ^{134}Cs and ^{137}Cs radionuclides in soil for the considered cases of natural distribution by profile is within $\pm 10\%$.

The presented data of MC simulating for determining of radial zone contribution to the total count rate of simulated spectrum demonstrate that the main contribution is made by the radial zone with radius of up to 40 cm from the center of the probe regardless of the thickness of the contaminated layer. Such small site facilitates the selection of measurement areas of soil with enough flat surface, which is desirable for surveying areas, especially with complex terrain.

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