ISSN 1063-7788, Physics of Atomic Nuclei, 2021, Vol. 84, No. 11, pp. 1929–1934. © Pleiades Publishing, Ltd., 2021.

RADIATION SAFETY

Methods for Measuring Daughter Products of Radon Decay in the Surface Atmospheric Layer of the Earth

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Received October 31, 2020; revised November 9, 2020; accepted November 10, 2020

Abstract—At present, an urgent problem is the study of temporal variations of various types of nuclear radiation from the lithosphere in foothill and desert landscapes. This refers to the fluxes of neutrons, gamma quanta, beta particles, and emanations of heavy chemical elements. The problem of studying such distributions in the surface atmospheric layer of the Earth remains relevant, owing to the fact that they are concentrated in the human habitat and have a direct impact on the health of the population. This work is devoted to the study of beta spectra of the natural radiation background in the surface atmospheric layer of the Earth. The origin of the measured beta spectra is associated with the daughter products of the decay of radon isotopes ^{219}Rn, ^{220}Rn, and ^{222}Rn in three natural radioactive series. Measurements of the spectra were performed from October 2018 to October 2019 in the foothills of the Zailiysky Alatau of the Tien Shan in Almaty. The frequency of measurements, on average, was about 10 measurements per day with an exposure of at least 2000 s. A database of daily, seasonal, and annual variations in beta spectra has been accumulated. For the analysis of the data obtained, a special software program Analyzer of the Beta Spectra Array was developed, which made it possible to process the standard output files of the Sputnik spectrometric installation and to integrate each spectrum in a given time interval. Time variation over the measured period was described using a standard mathematical computer package for wavelet analysis. The wavelet spectra obtained as a result of integration are used to identify daily, seasonal, and annual effects in variations of beta emanations. Along with this, similar mathematical processing was performed to predict the impact of external factors in the temporal variations of beta particles.

Keywords: daughter products of radon decay, beta spectra, natural radionuclides, wavelet analysis, variations in natural background radiation

DOI: 10.1134/S1063778821090118

1. INTRODUCTION

During the decay of uranium and thorium isotopes in radioactive natural chains, radon isotopes are formed, which from the depths of the lithosphere, by coagulation and further diffusion, enter the surface atmospheric layer. Despite numerous studies of radon emanation [1–4], the problem of studying the distributions of radon concentrations in the Earth's atmosphere remains relevant, owing to the fact that the radioactive gas radon is concentrated in the human environment and has a direct impact on health of the population in the form of an increased risk of cancer. The contribution of radon and daughter products of radon decay to the total background radiation is large and amounts to more than 50%. According to the International Commission on Radiological Protection (ICRP, publications no. 50 and no. 65), the bulk of oncological diseases of the lungs and bronchi are caused precisely by radon isotopes and, in particular, by daughter products of radon decay [5, 6]. Radon is classified as a Group I carcinogen by the World Health Organization (WHO) and the International Agency for Research on Cancer (IARC) [7]. Radon exposure is the second most important cause of lung cancer in smokers and the first for nonsmokers in the world [8, 9].

As a result of decays of radon isotopes in the surface atmospheric layer, radon daughters are formed. Most of the experimental methods have been developed to determine the alpha radioactive background generated by the radon daughters [10–15]. However, among the radionuclides that make the greatest contribution to the dose rate of internal radiation, the short-lived decay products of ^{222}Rn (about 60%) are in first place: they are mainly composed of beta-radioactive isotopes, such as ^{214}Pb (26.8 min), ^{214}Bi (19.9 min), ^{210}Tl (1.3 min), ^{210}Bi (5.013 days), ^{210}Pb (22.3 years), and

Table 1. Scheme of decays of ^{222}Rn daughters

Radio-nuclide	Decay type	$T_{1/2}$	E, MeV	Radon daughter
^{222}Rn	α	3.8235 d	5.590	^{218}Po
^{218}Po	α 99.98% β^- 0.02%	3.10 min	6.115 0.265	^{214}Pb ^{218}At
^{218}At	α 99.90% β^- 0.10%	1.5 s	6.874 2.883	^{214}Bi ^{218}Rn
^{218}Rn	α	35 ms	7.263	^{214}Po
^{214}Pb	β^-	26.8 min	1.019	^{214}Bi
^{214}Bi	β^- 99.98% α 0.02%	19.9 min	3.272 5.617	^{214}Po ^{210}Tl
^{214}Po	α	0.1643 ms	7.883	^{210}Pb
^{210}Tl	β^-	1.30 min	5.484	^{210}Pb
^{210}Pb	β^-	22.3 yr	0.064	^{210}Bi
^{210}Bi	β^- 99.99987% α 0.00013%	5.013 d	1.161 5.036	^{210}Po ^{206}Tl
^{210}Po	α	138.376 d	5.407	^{206}Pb
^{206}Tl	β^-	4.202 min	1.532	^{206}Pb
^{206}Pb		stable		

^{206}Tl (4.202 min) (Table 1). Next come natural radionuclides: ^{40}K and ^{87}Rb.

Measurements of radon emanation are rather complicated, because they depend on various accompanying natural and anthropogenic factors. Time variations of radon emanation studied by the authors [16, 17] also showed a strong dynamics of concentration not only from daily and seasonal variations. In addition to temporal variations of radon and radon daughters, their concentrations depend on the height inside buildings and in the free atmosphere, the geological landscape, temperature, pressure, and other complex factors [18]. Variations in radon emanation also depend on the localization of tectonic faults in the Earth's crust, the composition and porosity of the soil, and the presence of near-surface groundwater, as well as other geological properties of the area.

The main dose received by inhalation of radon is formed as a result of saturation of body cells and intercellular space with radon daughters [19]. Thus, it seems relevant to measure the activity of the distributions of beta-active radionuclides of radon daughters in the surface layer of the atmosphere at various moments of time. This is primarily due to the study of the risks of oncological morbidity caused by radionuclides entering the human body through the respiratory tract and the digestive system. To assess the accumulation factor in the human body of beta radionuclides of radon daughters, year-round measurements of beta spectra were carried out in the residential foothill zone of the Tien Shan in the spurs of the Zailiyskiy Alatau.

2. MEASUREMENT PROCEDURE AND RESULTS

The technique for measuring the temporal variations of natural beta radiation is based on the registration of spectra in the range up to 5.5 MeV. When measuring the activity of radionuclides in the soil samples under study, a scintillation beta spectrometer of SKS-99 Sputnik type was used. The spectra of natural beta-active radionuclides were measured in the period from October 2018 to October 2019. The frequency of year-round measurements averaged about 10 measurements per day with an exposure of at least 2000 s. During the year of measurements, a large array of experimental data on spectra was accumulated. For the analysis of the spectra, the authors developed a special software program Analyzer of the Beta Spectra Array. This mathematical support software made it possible to process the standard output files of the Sputnik spectrometric installation by integrating each spectrum in specified intervals. Figure 1 shows the interface of this mathematical software, which allows one to visualize the results of the analysis and select the optimal range of integration.

During the measurement period, more than 1600 files with beta spectra were accumulated. The developed software, after integrating each spectrum, formed the time variation (Fig. 2) averaged over a certain period. In this case, the averaging was performed over 2 d. It can be seen that, against the background of daily fluctuations, a seasonal variation of natural beta-active radionuclides is definitely traced.

Time variation over the measured period was mathematically analyzed using wavelet analysis. Wavelets are special functions in the form of short waves (bursts) with a zero integral value and with localization along the axis of the independent variable (t or x), capable of shifting along this axis and scaling (stretching/compressing). Any of the most commonly used wavelet types generates a complete orthogonal system of functions. In the case of wavelet analysis (decomposition) of a process (signal), owing to a change in scale, wavelets are able to reveal anomalies in the characteristics of the process on different scales, and by means of a shift, it is possible to analyze the properties of the process at different points throughout the studied interval. It is precisely due to the completeness property of this system that it is possible to restore (reconstruct or synthesize) the process by means of the inverse wavelet transformation. Owing to this, knowing the calculated wavelet analysis coefficients, one can predict the flux of beta particles of the natural background radiation in the near future.

The wavelet analysis technique was as follows [20]: the wavelet transform (WT) of a one-dimensional signal is its representation in the form of a generalized Fourier series or integral over the system of basis functions

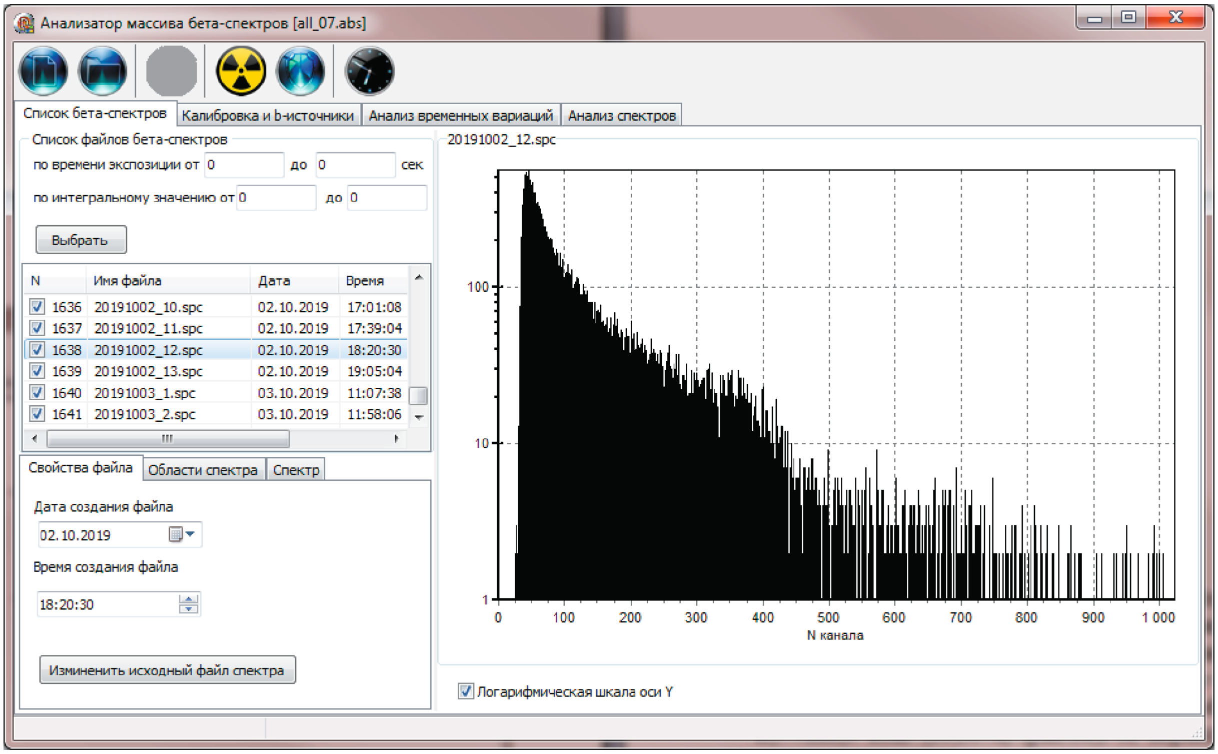

Fig. 1. View of spectrum from the software interface Analyzer of the Beta Spectra Array.

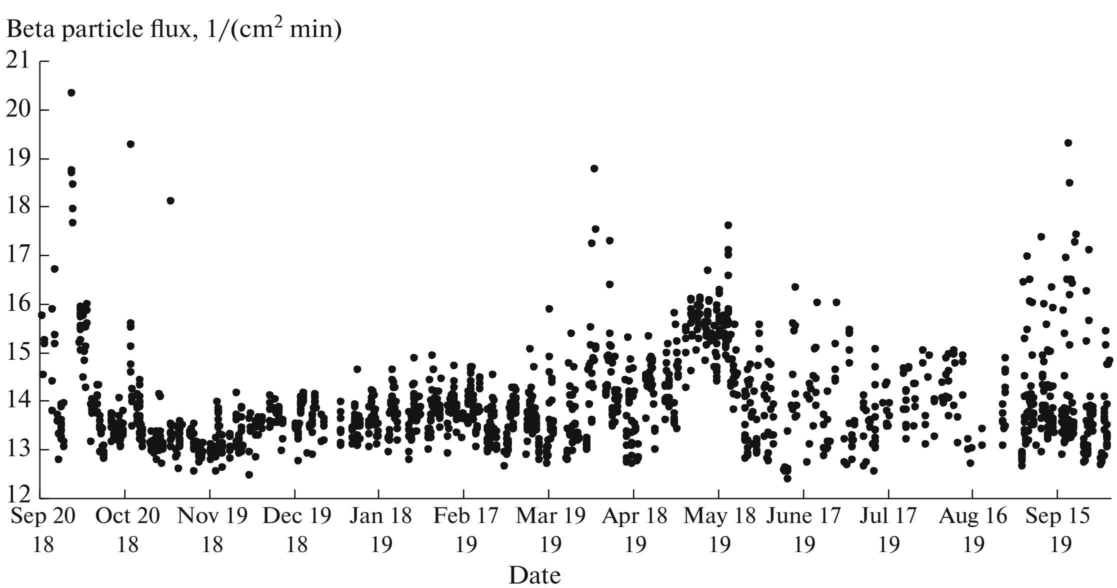

Fig. 2. Time variations of the radiation beta background.

$$\psi_{ab}(t) = \frac{1}{\sqrt{a}}\psi\left(\frac{t-b}{a}\right), \qquad (1)$$

constructed from the parent (initial) wavelet $\psi(t)$, which has certain properties owing to the operations of shifting in time (b) and changing the time scale (a). All designations are generally accepted. Then the continuous wavelet transformation will take the following form for the direct

$$W_S(a,b) = S(t)\psi_{ab}(t) = \frac{1}{\sqrt{a}}\int_{-\infty}^{\infty} S(t)\psi\left(\frac{t-b}{a}\right)dt \qquad (2)$$

and for the inverse

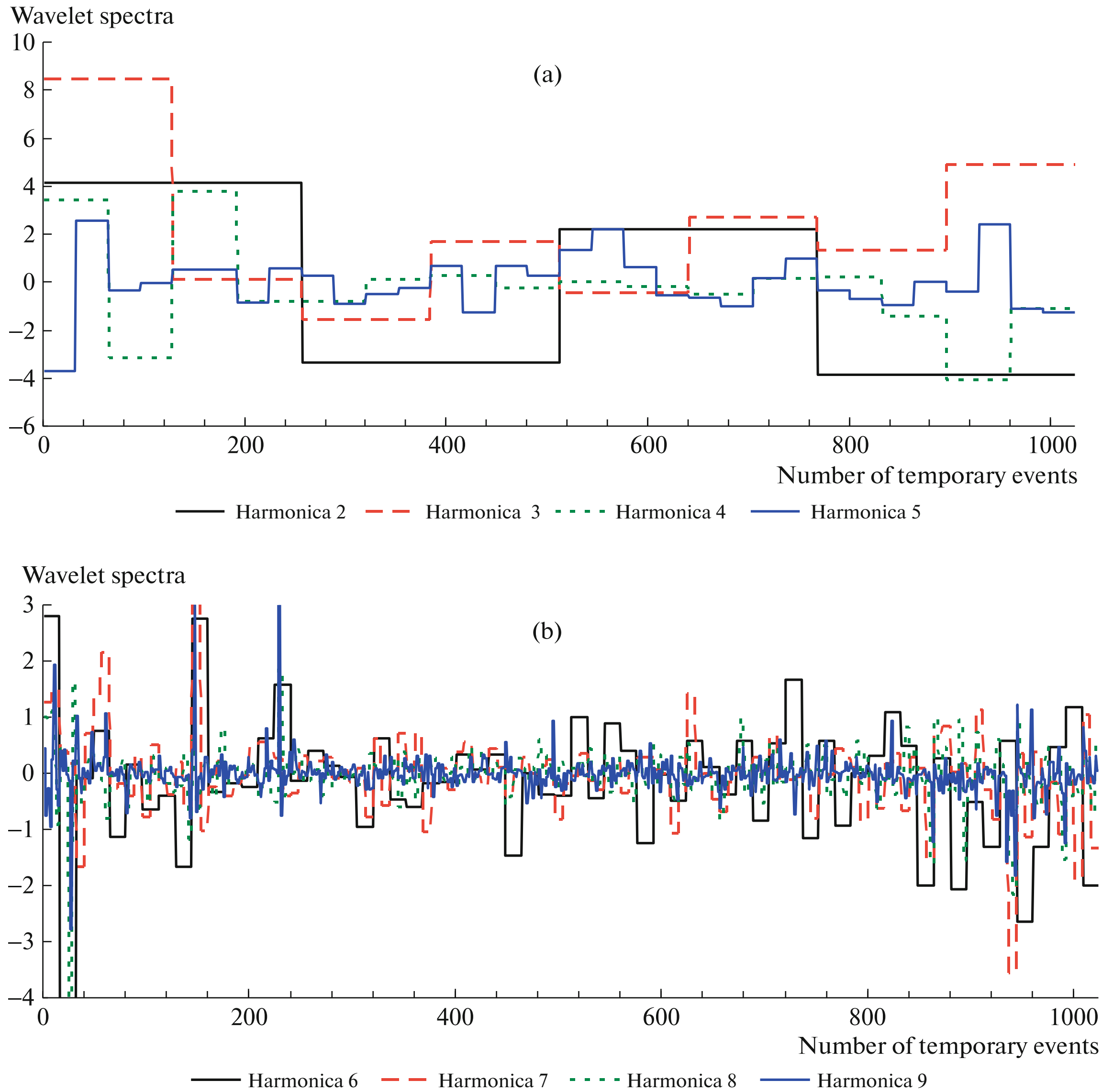

Fig. 3. Calculated wavelet spectra for the measured temporal beta background: (a) low frequencies; (b) high frequencies.

$$S(t) = \frac{1}{C_\psi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} W_S(a,b)\psi_{ab}(t) \frac{dadb}{a^2}, \tag{3}$$

where C_ψ is the normalizing coefficient

$$C_\psi = \int_{-\infty}^{\infty} |\psi(\omega)|^2 |\omega|^{-1} d\omega < \infty. \tag{4}$$

From (2) it follows that the wavelet spectrum $Ws(a, b)$ (scale-time spectrum), in contrast to the Fourier spectrum, is a function of two arguments: the first argument a (time scale) is similar to the oscillation period, that is, it is inverse to the frequency, and the second argument b is similar to the shift of the signal along the time axis. It should be noted that $Ws(b, a_0)$ characterizes the time dependence (at $a = a_0$), while the $Ws(a, b_0)$ dependences can be associated with the frequency dependence (at $b = b_0$).

As a result of the calculations of the wavelet transform, the wavelet spectra were obtained (Fig. 3) depending on the number of harmonics taken into account.

Figure 4 shows the full wavelet spectrum for the measured temporal beta background. Using the calculated wavelet spectra, it is possible to reconstruct the temporal variation of the beta background according to (3) using the required set of wavelet spectra to identify the time event of interest. Figures 5 and 6 show the reconstruction of temporal variation using six and eight wavelet spectra, respectively.

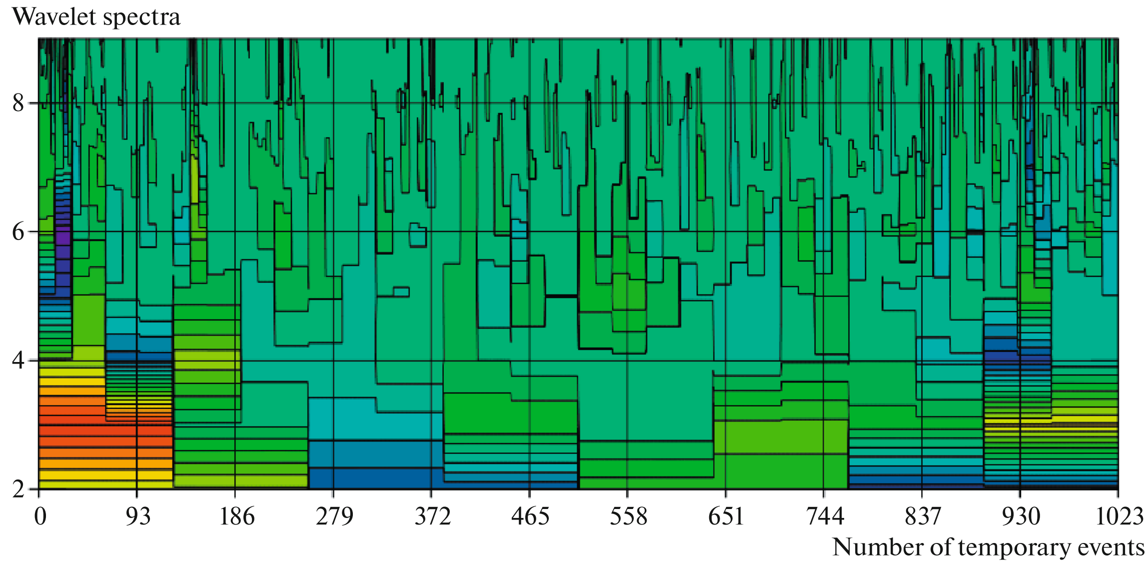

Fig. 4. Full wavelet spectrum for the measured temporal beta background.

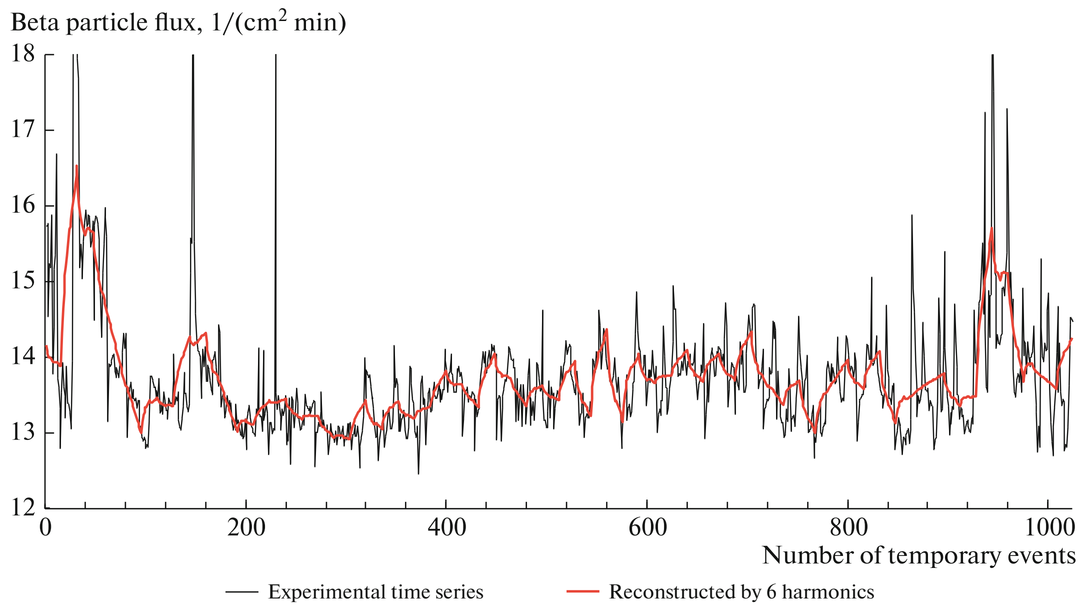

Fig. 5. Experimental and theoretical time series of the beta background for six harmonics.

It can be seen from the results obtained that the temporal variations of natural beta activity are fairly accurately reproduced using wavelet analysis taking into account six or more harmonics. With an increase in the number of harmonics, the accuracy of coincidence between theory and experiment increases, which allows the obtained time scene to be extended to future time intervals in order, for example, to estimate the integral dose received by individual cohorts of the population (personnel, population).

3. CONCLUSIONS

Thus, in this work, long-term precision measurements of time variations of emanations of beta particles from the Earth's lithosphere have been performed. Particular attention has been paid to the possibility of using the results obtained to assess the dosimetric contribution to the total radiation received by personnel and the inhabitants as a result of exposure to natural beta radionuclides. A technique for reconstructing wavelet spectra by optimizing the val-

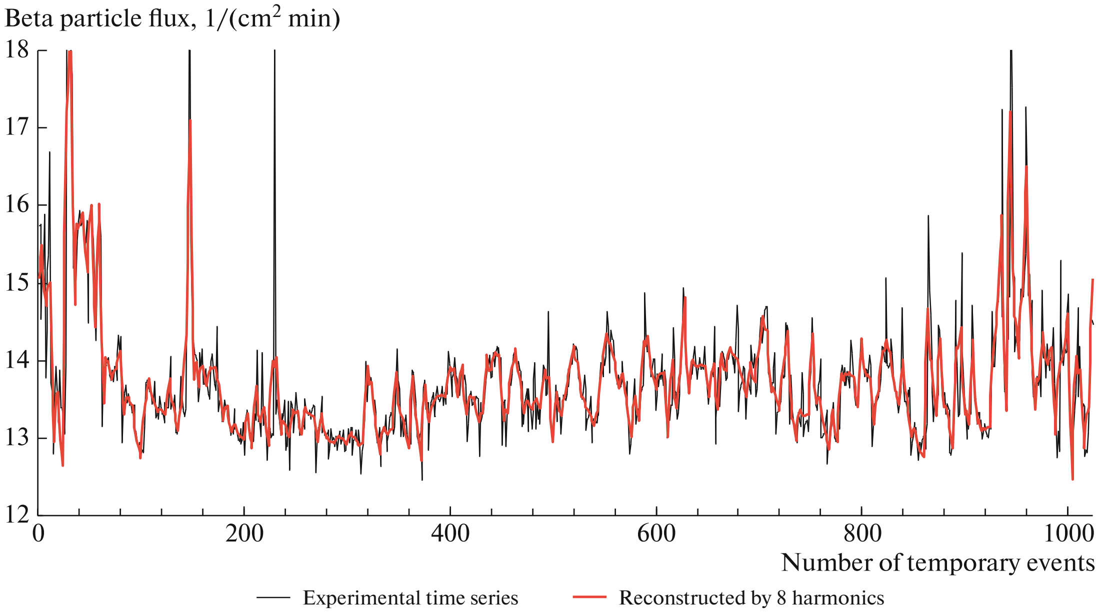

Fig. 6. Experimental and theoretical time series of the beta background for eight harmonics.

ues of wavelet coefficients obtained from standard wavelet analysis has been developed. It is shown that the accuracy of the obtained wavelet spectra substantially depends on the number of wavelet harmonics with an optimization of the wavelet coefficients of more than six. Such accuracy of reproduction of experimental data makes it possible to extend the actually measured variations of beta particle emanation to any time intervals in order to predict the doses obtained from the decay of radon and radon daughters and to assess the risks of oncological morbidity for both personnel and the general population.

FUNDING

This work was supported by the state grant funding for basic scientific research (projects no. IRN AP09058404, AP09258978, AP05131884).

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Translated by V. Selikhanovich