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Calibration and testing of a portable NaI(Tl) gamma-ray spectrometer–dosimeter for evaluation of terrestrial radionuclides and ^{137}Cs contributions to ambient dose equivalent rate outdoors

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A commercially available NaI(Tl) based spectrometer-dosimeter has been used to separate contributions of the terrestrial radionuclides (^{238}U series, ^{232}Th series and ^{40}K) and ^{137}Cs to the total ambient dose equivalent rate (ADER), $\dot{H}^*(10)$, outdoors. The device had been initially calibrated by the manufacturer to measure: 1) the total ADER; 2) activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and 'effective' activity concentration of the terrestrial radionuclides, AC_{eff}, in soil, agricultural raw materials, forestry products and construction materials; and 3) ground contamination density by ^{137}Cs. In order to derive a conversion coefficient from AC_{eff} to the ADER stipulated by the terrestrial radionuclides ($ADER_{TRN}$), two series of additional calibration measurements were performed. The calibration measurements were conducted at 27 outdoor locations in the center of St.-Petersburg and in the Leningradskaya region (background areas). The conversion coefficient from AC_{eff} to $ADER_{TRN}$ of 0.51 (nSv h^{-1})/(Bq kg^{-1}) has been obtained using a regression analysis of experimental data. The intrinsic noise of the spectrometer and its response to cosmic radiation at sea level has been estimated to 7 nSv h^{-1}. The calibration factors must be used with caution and only for detectors similar to the one employed in this study (assembly based on a NaI(Tl) single crystal, 63 mm in diameter and 63 mm in length). The spectrometer-dosimeter and experimentally derived calibration coefficients have been tested in field at seven sites in the south-western districts of the Bryansk region that had been heavily contaminated by Chernobyl fallout. The contribution of ^{137}Cs to the total ADER varies between 40% and 95%. The preliminary results of the measurements confirm the potential of in situ gamma-ray spectrometry for assaying natural and artificial components of the ambient dose equivalent rate outdoors.

Key words: *gamma-ray, spectrometer, NaI(Tl), dosimeter, ambient dose equivalent rate, terrestrial radionuclides, ^{40}K, ^{226}Ra, ^{232}Th, ^{137}Cs.*

1. Introduction

Direct measurement of gamma dose rate in air using a gamma-ray dosimeter is one of the common procedures to assess the external exposure of humans in the case of an environmental contamination with technogenic gamma-ray emitting radionuclides [(Generic procedures for monitoring in a nuclear or radiological emergency. IAEA-TECDOC-1092, IAEA – International Atomic Energy Agency, 1999); (Radiation monitoring of the exposure doses of population at the territories radioactively contaminated due to the accident at the Chernobyl NPP. Recommended Practices. Adopted 27.12.07, implemented 27.12.07. Federal Center of Hygiene and Epidemiology of Federal Service for Surveillance on Consumer Rights Protection and Human Well-Being, Moscow, 2007)]. The dosimeter reading in terms of ambient dose equivalent rate, $\dot{H}^*(10)$, ($ADER_{reading}$) is the sum of the following major components: 1) dose rate due to the terrestrial radionuclides of ^{238}U series, ^{232}Th series and ^{40}K ($ADER_{TRN}$); 2) dose rate due to the directly ionizing and photon component of cosmic radiation; 3) intrinsic noise of the dosimeter; 4) dose rate due to technogenic radionuclides [1, 2, 3]. Additional small contribution to $ADER_{reading}$ can be associated with some other terrestrial radionuclides, including those of the ^{235}U series and ^{138}La, the so called "cosmogenic" natural gamma-emitting radionuclides (^{7}Be and ^{22}Na) and Rn progenies in the atmosphere [3, 4, 5, 6]. For correct estimation of external exposure from artificial sources, it is necessary to subtract all other components from the dosimeter reading.

Several approaches for separation of the $ADER_{reading}$ components have been proposed and practically implemented. Specifically, for assessing the sum of

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contributions of cosmic radiation and intrinsic noise of a dosimeter, additional measurements with the dosimeter can be carried out on a surface of a large water body (e.g., lake or sea) [(Radiation monitoring..., 2007), 2, 7, 8]. The contribution from terrestrial radionuclides to the total ADER may be evaluated by determination of the ^{238}U, ^{232}Th and ^{40}K activity concentrations in soil using stationary or/and portable gamma-ray spectrometers. It can be done with: 1) soil sampling and subsequent laboratory analysis or/and 2) direct measurements in field (*in situ* measurements), e.g., [2, 5, 9, 10, 11].

In the 1960–70s, Beck et al. [5, 12] developed the theoretical principles of *in situ* gamma-ray spectrometry and practically demonstrated that *in situ* measurements of soil activity could provide more representative data for evaluation of gamma dose rate in air than the data obtained by soil sample collection and laboratory analysis. Since these pioneering studies the field has rapidly expanded (for review see [13, 14, 15]). The assessment of the external dose components was one of the applications of *in situ* gamma-ray spectrometry after the Chernobyl accident (1986). In particular, a portable germanium detector was used to differentiate absorbed dose rates in air due to uranium series, thorium series, ^{40}K and ^{137}Cs during an extended survey of indoor and outdoor terrestrial gamma radiation in Greece [16].

The present investigation is a study on the application of a commercially available portable gamma-ray spectrometry system for *in situ* analysis of the activity concentration in soil and ADER in the remote period after the Chernobyl accident. We have used a NaI(Tl) based spectrometer-dosimeter from ATOMTEX (Belarus). The AT6101D spectrometer [17] has the pattern approval certificates of the Republic of Belarus, the Russian Federation, Ukraine and Kazakhstan. The device is calibrated by the manufacturer to determine: 1) activity concentrations (Bq kg^{-1}) of the terrestrial radionuclides of ^{226}Ra, ^{232}Th and ^{40}K in soil, agricultural raw materials, forestry products and construction materials; 2) ground contamination density by ^{137}Cs, A_{Cs} (Bq m^{-2}); 3) total ambient dose equivalent rate, ADER (nSv h^{-1}).

Potentially, the device can be used to separate contributions of terrestrial radionuclides and ^{137}Cs to the total ADER. Unfortunately, the spectrometer was not factory calibrated in such a manner. Therefore, the main aim of the present study was to deduce the calibration coefficient for conversion of the measured AC_{eff} (Bq kg^{-1}) to the terrestrial component of ambient dose equivalent rate, $ADER_{TRN}$ (nSv h^{-1}). It has allowed quantifying the dose rate associated with terrestrial radionuclides in the presence of ^{137}Cs contamination. Another aim was to test the spectrometry system in field at the radioactively contaminated areas of the Bryansk region (Russia).

The laboratory and field measurements were carried out in the period 2014–2016.

2. Materials and methods

2.1. Instruments and the factory calibration procedure

The portable spectrometer-dosimeter MKS AT 6101D [17] consists of two blocks: detection and processing units that are connected by a water-proof cable (Fig. 1). The detection unit is a NaI(Tl) cylindrical scintillation detector (63 mm in diameter and 63 mm in length). The energy resolution (FWHM at 662 keV of ^{137}Cs) of the detector is 7.3%. The detector is placed in a temperature-and-shock-resistant dust-proof and moisture-proof container (121 mm in diameter and 477 mm in length). The weight of the unit is 4 kg. The available geometries for measurements are 2π (on a surface) and 4π (in a well). Spectrometric information from the detection unit is transferred into a processing unit (mass = 0.8 kg) and is displayed on LCD screen. The multichannel analyzer is adjusted to 512 channels at the energy range from 40 keV to 3 MeV. Up to 300 spectra can be stored in the energy independent memory of the spectrometer. The instrumental spectra processing algorithm allows data display in the form of activity concentration of the terrestrial radionuclides, ground contamination density by ^{137}Cs and its activity concentration.

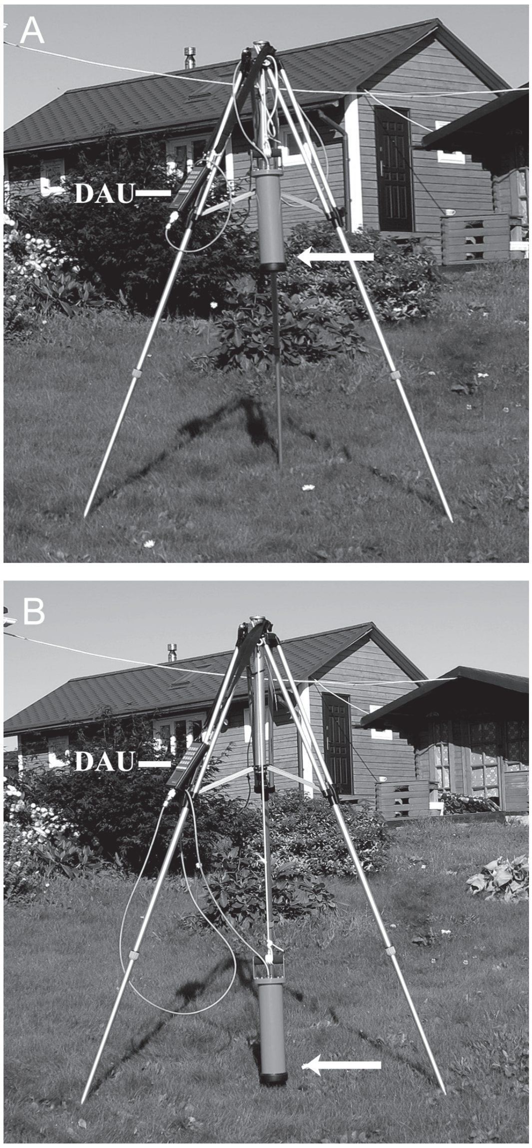

Fig. 1. *In situ* gamma-ray spectroscopic measurements with MKS AT6101D at a kitchengarden in Khittolovo (plot Hit-2), the Leningradskaya region in 2015. The detection unit and the data analysis unit (DAU) are set up on an aluminum tripod. The white arrows indicate approximate position of the effective center of the detector: at a height of about 1 m (the top panel A) and 0.1 m (the lower panel B) above the ground

For energy calibration of the spectrometer, point sources of ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{54}Mn, ^{22}Na, ^{60}Co, ^{152}Eu, ^{137}Cs, ^{228}Th and ^{88}Y were used. The 3 window matrix method (see e.g., [18]) was used to calculate the ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K activities of the soil samples. The energy windows were centered on the 1461 keV (^{40}K), 1764 keV (^{214}Bi) and 2615 keV (^{208}Tl) full-energy peaks (Fig. 2) for the estimation of ^{40}K, ^{238}U, and ^{232}Th activity concentrations, respectively. The uranium ^{238}U and thorium ^{232}Th content was calculated under the assumption that secular equilibrium exists between all of the radionuclides within the decay series. High volume standards containing ^{40}K, ^{226}Ra and ^{232}Th have been used to calibrate the spectrometer. The field spectrum processing includes subtraction of the so called “background spectrum” recorded by the manufacturer inside a 10 cm thick lead shield. Intrinsic relative uncertainty of the monitored radionuclide concentration measurements is ±20% for the terrestrial radionuclides and ±30% for ^{137}Cs (maximum).

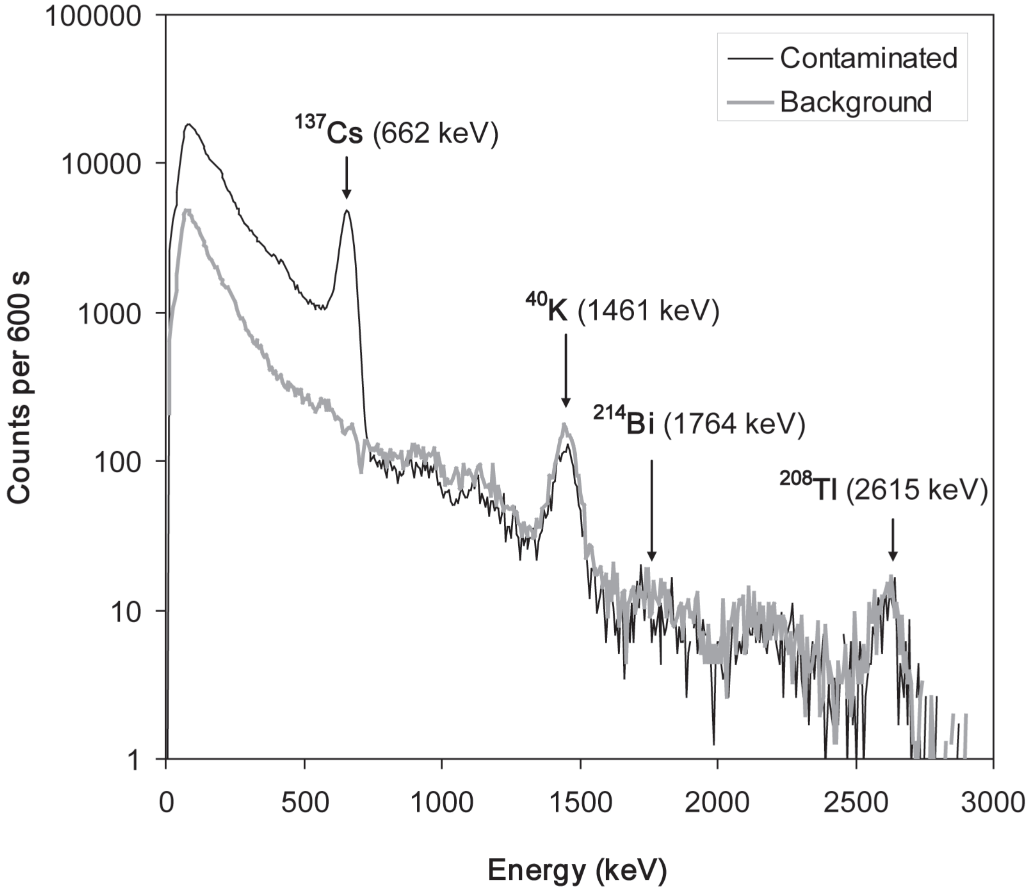

Fig. 2. *In situ* NaI(Tl) gamma-ray spectra recorded with MKS AT6101D at the background (plot Hit-2, Khittolovo, the Leningradskaya region) and contaminated (plot Les-13kit, Novozybkov, the Bryansk region) kitchengardens in 2015. Positions of peaks from the natural radionuclides ^{214}Bi (^{238}U series), ^{208}Tl (^{232}Th series) and ^{40}K, and man-made radionuclide ^{137}Cs are indicated by arrows. Counting time is 925 s for the background plot and it is 695 s for the contaminated plot

To represent the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th by a single quantity, which takes into account the total external exposure associated with them, ‘effective’ activity concentration of the natural radionuclides in soil, AC_{eff} (Bq kg^{-1}), is calculated automatically within the AT6101D dosimeter-spectrometer using the formula:

$$AC_{eff} = AC_{226Ra} + 1.31 \times AC_{232Th} + 0.085 \times AC_{40K} \quad (1)$$

where AC_{226Ra}, AC_{232Th} and AC_{40K} are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}), respectively [GOST 30108-94. Building materials and elements. Determination of specific activity of natural radioactive nuclides. Gosstroy Rossii; Moscow, 1995].

The definition of AC_{eff}, which is in use in the Russian Federation, is closely related to so called “radium equivalent activity, Ra_{eq}”, a commonly used radiological hazard index (e.g., [19, 20, 21]). For the calculation of Ra_{eq}, it is assumed that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same γ-ray dose rates. Therefore, the correction coefficients are 1.43 for ^{232}Th and 0.077 for ^{40}K. These are slightly (by about 10%) different from those values that have been used by the AT6101D producer for derivation of AC_{eff} (eq. 1).

The ambient gamma-radiation dose equivalent rate value in an inspection point is determined by the instrument spectrum analysis with the “spectrum-dose” operational functions. The measured pulse height distribution is converted automatically into the physical quantity of dose rate using the response functions that are recorded in the memory unit of the spectrometer-dosimeter. The correction functions (coefficients) are energy dependent. The validity of the spectrum-to-dose conversion function has been verified by the manufacturer using a strong standard ^{137}Cs source. For the reference dose rate values of 0.7, 7.00 and 70.0 µSv h^{-1}, the readings were 0.681, 7.03 and 70.8 µSv h^{-1}, respectively [AT6101D Spectrometer. Operation manual. ATOMTEX, Minsk, Belarus, 2014]. According to the manufacturer, the intrinsic relative uncertainty of the ADER measurements in field is within ±20% (maximum).

The manufacturer provides a reference KCl volume source (mass = 0.2 kg) for periodical energy calibration of the spectrometer *in situ* (the 1461 keV full-energy peak of ^{40}K). The energy calibration can also be done using a ^{137}Cs reference source (the 662 keV full-energy peak of ^{137}Cs–^{137m}Ba). Additionally, the spectrometer has a built-in automatic stabilization which is achieved by using light emitting diode. Its instability during a continuous survey is estimated as ±1.5% (maximum).

2.2. Additional in–house calibration and field testing

To estimate contribution of cosmic radiation and intrinsic noise of the device into the primary reading of the spectrometer, $ADER_{reading}$, three gamma-ray spectra were recorded on the frozen surface of the Finnish Gulf in the Leningradskaya region (Russia) in December 2016, shortly after formation of an ice cover. The measurements were performed at a distance of about 2.5 km from the shore-line. The site (60.113° N, 29.898° E) has an average water depth of 6 m.

To determine a relationship between AC_{eff} and ADER, spectrometric measurements were made outdoors at the territory of St.-Petersburg and at two settlements in the Leningradskaya region (Table 1). The areas can be considered as a “background” because they did not receive any substantial amount of Chernobyl fallout (Table 1). In these areas, the current ground contamination level by ^{137}Cs (from global fallout and the Chernobyl accident) is estimated below 5 kBq m^{-2}. A total of 27 sites (20 ground plots and 7 paved areas) were selected for the measurements (Table 2). The sites characteristics can be found in Tables 3 and 4. At 12 plots (Table 3), *in situ* measurements were performed with a downward facing detector at heights of 0.1 m and 1.0 m above the ground (or paved surface). The detection and processing units were mounted on an aluminum tripod (mass = 4.0 kg) (Fig. 1). Data from this series of measurement were used to evaluate: 1) a relationship between AC_{eff} and ADER for our spectrometric system and 2) a relationship between the results obtained at the two heights above the ground. The validity of the AC_{eff} to ADER conversion coefficient was checked in a second series of measurements carried out at 15 different plots at a height of 0.1 m above the ground (Table 4).

Table 1

List of background and contaminated settlements surveyed in 2014–2016

Region	Settlement	Latitude, °N	Longitude, °E	Altitude, m	^{137}Cs inventory (kBq m^{-2})[a]	Year of survey
			Background areas			
St.-Petersburg	St.-Petersburg	59.934	30.334	13	2–10 (<1.3)	2014, 2015
Leningradskaya region	Beloostrov	60.147	30.012	23	2–10 (<0.6)	2016
Leningradskaya region	Khittolovo	60.228	30.527	90	2–10 (<0.7)	2015, 2016
			Contaminated areas			
Bryansk region	Novozybkov	52.535	31.933	170	697 (3.1–447)	2015
Bryansk region	Zatishie	52.826	32.314	155	80 (44.6)	2016

[a] – the initial official ^{137}Cs inventory is given for 1986 according to [28, 29]. The values in brackets are the ^{137}Cs inventory determined in this study (on the year of the survey) with *in situ* measurements at a height of 0.1 m above the ground.

Table 2

Ambient dose equivalent rate ($ADER_{reading}$), surface ground contamination with ^{137}Cs (A_{Cs}), activity concentration of terrestrial radionuclides (AC_{40K}, AC_{226Ra}, AC_{232Th}) and ‘effective’ activity concentration of terrestrial radionuclides (AC_{eff}) determined with *in situ* measurements (AT1601D) at a height of 0.1 m above the ground at the background areas (St.-Petersburg and the Leningradskaya region) in 2014–2016

Parameter	$ADER_{reading}$ (nSv h^{-1})	A_{Cs} (kBq m^{-2})	AC_{40K} (Bq kg^{-1})	AC_{226Ra} (Bq kg^{-1})	AC_{232Th} (Bq kg^{-1})	AC_{eff} (Bq kg^{-1})
			Ground plots (n=20)			
Minimum	52	<0.73	458	11	22	80
Maximum	89	<0.73	947	29	45	155
Median	69	<0.73	589	18	31	111
Mean	70	<0.73	624	19	32	115
SD	9	n.e.	143	5	7	18

The end of table 2

Parameter	$ADER_{reading}$ ($nSv\ h^{-1}$)	A_{Cs} ($kBq\ m^{-2}$)	AC_{40K} ($Bq\ kg^{-1}$)	AC_{226Ra} ($Bq\ kg^{-1}$)	AC_{232Th} ($Bq\ kg^{-1}$)	AC_{eff} ($Bq\ kg^{-1}$)
			Plots paved with asphalt (n=4)			
Minimum	123	<0.87	1040	38	62	228
Maximum	149	<0.87	1570	62	75	268
Median	132	<0.87	1320	41	69	245
Mean	134	<0.87	1310	46	69	247
SD	11	n.e.	220	11	7	17
			Plots paved with granite (n=3)			
Minimum	189	<1.3	1240	54	138	356
Maximum	237	<1.3	1870	102	161	442
Median	210	<1.3	1410	89	150	420
Mean	212	<1.3	1510	82	150	406
SD	24	n.e.	330	25	12	45

SD – standard deviation; n.e. – not estimated.

Table 3

Values of ambient dose equivalent rate ($ADER_{reading}$) and 'effective' activity concentration of terrestrial radionuclides (AC_{eff}) determined at heights of 0.1 m and 1 m above the ground at 12 background plots in 2014–2016

Settlement	Code of plot	Location	Material	$ADER_{reading}$ ($nSv\ h^{-1}$)[a]			AC_{eff} ($Bq\ kg^{-1}$)[b]		
				0.1 m	1 m	1 m/0.1 m ratio	0.1 m	1 m	1 m/0.1 m ratio
Khittolovo	Hit-1	grassland	disturbed soil	66	65	0.98	109 (8)	103 (9)	0.94
Khittolovo	Hit-2	kitchendarden	cultivated soil	68	67	0.99	111 (9)	110 (10)	0.99
Beloostrov	Bel-1	kitchendarden	cultivated soil	67	65	0.97	117 (10)	105 (10)	0.90
Beloostrov	Bel-2	kitchendarden	cultivated soil	61	59	0.97	104 (13)	96.9 (14)	0.93
Khittolovo	Hit-4	kitchendarden	cultivated soil	80	76	0.95	140 (10)	128 (11)	0.91
Khittolovo	Hit-5	kitchendarden	cultivated soil	89	87	0.98	152 (11)	152 (13)	1.00
Khittolovo	Hit-6	kitchendarden	cultivated soil	74	75	1.01	117 (13)	124 (12)	1.06
Khittolovo	Hit-7	kitchendarden	cultivated soil	75	72	0.96	125 (11)	116 (12)	0.93
Khittolovo	Hit-3	forest	soil	52	50	0.96	79.7 (11)	76.9 (11)	0.96
St-Petersburg	IRH-1	paved area	asphalt	123	119	0.97	228 (11)	211 (11)	0.93
St-Petersburg	IRH-2	paved area	asphalt	149	138	0.93	268 (9)	236 (9)	0.88
St-Petersburg	IRH-3	paved area	asphalt	128	122	0.95	241 (11)	218 (10)	0.90
Minimum				52	50	0.93	80	77	0.88
Maximum				149	138	1.01	268	236	1.06
Median				75	74	0.97	121	120	0.93
Mean				86	83	0.97	149	140	0.95
SD				31	28	0.02	61	53	0.05

[a] – the primary reading of the AT1601D spectrometer-dosimeter; a statistical uncertainty (at the 2 sigma level) of the ADER measurements is below ±2%.
[b] – The statistical uncertainty (±%, at the 2 sigma level) for determination of AC_{eff} is given in brackets.
SD – standard deviation.

Table 4

A comparison between calculated (based on determination of the 'effective' activity concentration of terrestrial radionuclides, AC_{eff}) and measured values of ambient dose equivalent rate (ADER) at 15 background plots in 2014–2015

Settlement	Code of plot	Location	Material	AC_{eff} (Bq kg^{-1})[a]	ADER, calculated (nSv h^{-1})[b]	ADER, measured (nSv h^{-1})[c]	calculated/ measured ratio
St-Petersburg	PPF-2	grassland	disturbed soil	155 (13)	90.1	89.0	1.01
St-Petersburg	PPF-3	grassland	disturbed soil	125 (13)	74.8	73.0	1.02
St-Petersburg	PPF-4	grassland	disturbed soil	115 (11)	69.7	72.0	0.97
St-Petersburg	PPF-5	grassland	disturbed soil	99.2 (13)	61.6	63.0	0.98
St-Petersburg	SmC-2	grassland	disturbed soil	104 (12)	64.0	66.0	0.97
St-Petersburg	SmC-3	grassland	disturbed soil	99.2 (12)	61.6	61.0	1.01
St-Petersburg	AP-2	park	cultivated soil	103 (13)	63.5	69.0	0.92
St-Petersburg	AP-3	park	cultivated soil	115 (12)	69.7	74.0	0.94
St-Petersburg	AP-4	park	cultivated soil	104 (14)	64.0	65.0	0.99
St-Petersburg	AP-5	park	cultivated soil	106 (13)	65.1	67.0	0.97
St-Petersburg	AP-6	park	cultivated soil	110 (14)	67.1	69.0	0.97
St-Petersburg	SmC-1	paved area	asphalt	249 (11)	138	135	1.02
St-Petersburg	PPF-1	paved area	granite flagstone	442 (12)	236	237	1.00
St-Petersburg	AP-1	paved area	granite gravel	356 (14)	193	189	1.02
Khittolovo	Hit-8	paved area	granite gravel	420 (12)	225	210	1.07
Minimum				99.2	61.6	61.0	0.92
Maximum				442	236	237	1.07
Median				115	70	72	0.99
Mean				180	103	103	0.99
SD				124	63	60	0.04

[a] – a statistical uncertainty (±%, at the 2 sigma level) for determination of AC_{eff} is given in brackets.
[b] – the ADER is calculated using eq. (2) (ADER = 11 + AC_{eff} × 0.51).
[c] – the primary reading of the AT1601D spectrometer-dosimeter. A statistical uncertainty (at the 2 sigma level) of the ADER measurements is below ±2%.
SD – standard deviation.

The first practical testing of the spectrometer and empirically derived calibration factors was performed in Novozybkov, the Bryansk region. The town and its surroundings had been heavily contaminated following the Chernobyl accident (Table 1). The average residual ground contamination by ^{137}Cs was estimated as 396 kBq m^{-2} in 2014 [22]. The spectrometric measurements were performed at the typical locations [2]: grassland, kitchengarden, yard, street and forest (Table 5). For comparison, measurements were also made at a less contaminated site in the Bryansk region, Zatishie, where the average level of ground contamination in 2014 was only 41 kBq m^{-2} [22]. At each surveyed plot, gamma-ray spectra were recorded at a height of 0.1 m and 1.0 m, respectively, above the ground.

Table 5

Ambient dose equivalent rate ($ADER_{reading}$), surface ground contamination with ^{137}Cs (A_{Cs}), activity concentration of terrestrial radionuclides and 'effective' activity concentration of terrestrial radionuclides (AC_{eff}) determined with *in situ* measurements (AT1601D) at a height of 0.1 m above the ground at contaminated areas of the Bryansk region in 2015–2016

Settlement	Plot	Location	Material	$ADER_{reading}$ (nSv h^{-1})[c]	A_{Cs} (kBq m^{-2})	AC_{40K} (Bq kg^{-1})	AC_{226Ra} (Bq kg^{-1})	AC_{232Th} (Bq kg^{-1})	AC_{eff} (Bq kg^{-1})
Novozybkov	Les-13str	street	asphalt	100	3.1 (22)	582 (7.4)	29.5 (26)	30.8 (16)	119 (12)
Novozybkov	Les-13ya	yard	disturbed soil	247	116 (<1)	468 (7.9)	23.1 (28)	22.6 (19)	92.6 (13)
Novozybkov	Les-13kit	kitchengarden	cultivated soil	252	121 (<1)	474 (6.1)	20.2 (25)	23.4 (14)	91.3 (11)
Novozybkov	Les-13gr[a]	grassland	disturbed soil	263	135 (<1)	490 (8.8)	17.4 (43)	29.0 (18)	97.0 (14)
Novozybkov	Fil-gr[b]	grassland	undisturbed soil	602	414 (<1)	184 (11)	9.9 (37)	10.3 (23)	39.1 (15)

The end of table 5

Settlement	Plot	Location	Material	$ADER_{reading}$ (nSv h^{-1})[c]	A_{Cs} (kBq m^{-2})	AC_{40K} (Bq kg^{-1})	AC_{226Ra} (Bq kg^{-1})	AC_{232Th} (Bq kg^{-1})	AC_{eff} (Bq kg^{-1})
Novozybkov	Fil-fo	forest	undisturbed soil	617	447 (<1)	222 (10)	11.6 (34)	11.9 (22)	45.9 (14)
Zatishie	Zat-fo	forest	undisturbed soil	78.9	44.6 (2.2)	156 (12)	6.7 (49)	8.9 (25)	31.6 (17)

[a]– the plot is adjacent to the plot Les-13str (street) located inside settlement.
[b]– the plot is located outside settlement.
[c]– the primary reading of the AT1601D spectrometer-dosimeter. A statistical uncertainty (at the 2 sigma level) of the ADER measurements is below ±2%.
Statistical uncertainties (±%, at the 2 sigma level) of determination of A_{Cs}, AC_{40K}, AC_{226Ra}, AC_{232Th} and AC_{eff} are given in brackets.

All outdoor measurements (excluding the measurements on ice) were conducted in dry weather in summer time. Counting times ranged from 300 s to 1800 s.

Statistical analysis of data was performed using Excel for MS Windows and the tools provided by free on-line calculators, Free Statistics Calculator [23] and Centr sovremennykh psykhotekhnologiy [24].

3. Results and discussion

3.1. Additional in–house calibration

Three repeated measurements performed on the frozen surface of the Finnish Gulf showed the same value of 7 nSv h^{-1}. This value includes response to cosmic radiation and contribution from intrinsic noise of the spectrometer. The measurements revealed a rather weak response of the spectrometer to the directly ionizing and photon component of cosmic radiation because the population-weighted average dose rate from this source at sea level corresponds to 31 nGy h^{-1} (absorbed dose in air) or 31 nSv h^{-1} (effective dose) [6]. One should note that the spectrometer-dosimeter AT6101D is not designed to measure the cosmic radiation.

A summary of the dose rate and activity concentration of radionuclides at 27 background plots is presented in Table 2. The measurements were performed close to the soil or paved surface i.e. in the position recommended by the AT6101D manufacturer for evaluation of radionuclides activity in the 2π geometry [AT6101D Spectrometer. Operation manual, 2014].

No 662 keV peak can be seen in the pulse height distributions recorded at any of the background plots. An example of a spectrum obtained at a background area is shown in Fig. 2. A spectrum recorded at a contaminated area is also shown for comparison to demonstrate a prominent 662 keV peak from ^{137}Cs–^{137m}Ba. The ground contamination density by ^{137}Cs at the background plots was found to be below the detection limit (1.3 kBq m^{-2}). This value should be taken with care because the detection limit, which is recommended by the manufacturer of the AT6101D spectrometer, is equal to 4 kBq m^{-2}. But in any case, the residual ^{137}Cs contamination at our background plots is rather low (less than 5 kBq m^{-2}).

Activity concentrations of terrestrial radionuclides were in the ranges: 458–1870 Bq kg^{-1} for ^{40}K, 11–102 Bq kg^{-1} for ^{226}Ra and 22–161 Bq kg^{-1} for ^{232}Th. Values of AC_{eff} varied from 80 Bq kg^{-1} to 442 Bq kg^{-1}. The minimum values were registered at ground plots while the highest activities were recorded at sites paved with granite. Asphalted plots occupied the intermediate position in terms of recorded activity. A similar pattern can be seen with respect to dose rates registered at plots covered by soil, asphalt and granite material (Table 2).

To derive the ADER to AC_{eff} ratio for the two heights 0.1 m and 1 m, a linear regression analysis was applied to the data obtained at 12 plots. The characteristics and radiometric data for the plots are presented in Table 3, while the constructed regression lines and regression equations are shown at Fig. 3 (the top and middle panels). The median dose rate at 1 m height is slightly smaller (by 3%) than that at a height of 0.1 m (Table 3). A similar difference (by 5% on average) between the two heights can be seen with respect to AC_{eff}. Although small, the differences between detector heights over ground for both radiometric parameters are statistically significant (the non-parametric Wilcoxon signed-rank test, $P < 0.01$, n=12). Beck and de Planque [4] calculated exposure rates in air from natural gamma emitters homogeneously distributed in soil. A 2% reduction in the exposure rates was found with increasing the height of measurement point from 0 m to 1 m above the ground (see Table 3 in [4]).

A strong positive correlation has been found between AC_{eff} and ADER (Fig. 3). Spearman's coefficient of correlation, R_{Sp}, is 0.988 ($P < 0.01$; n = 12) for 0.1 m height and 0.998 ($P < 0.01$; n = 12) for the 1 m height.

The slope coefficient of the regression line (Fig. 3) was calculated as 0.496 [s.e. = 0.013] nSv h^{-1} per Bq kg^{-1} for the 0.1 m height and 0.529 [s.e. = 0.011] nSv h^{-1} per Bq kg^{-1} for the 1 m height. The slope coefficients can be interpreted as conversion coefficients from AC_{eff} to the ambient dose equivalent rates due to terrestrial radionuclides of ^{238}U series, ^{232}Th series and ^{40}K ($ADER_{TRN}$). The Student's *t*-test showed that the two slopes did not significantly differ from each other (*t*-value = 1.938, $P > 0.05$). Therefore we have decided to pool data from the two heights together to derive a single coefficient for further analysis of experimental data. The scatter plot and regression equation for the pooled data are presented in the lower panel of Fig. 3. As one can see from this Figure, the unified $ADER_{TRN}$ to AC_{eff} ratio (conversion coefficient) is approximately equal to 0.51(rounded from 0.509) nSv h^{-1} per Bq kg^{-1}.

The intercept of the regression line of the pooled data (Fig. 3, the lower panel) was calculated as 11 nSv h^{-1} [s.e. = 2 nSv h^{-1}; 95% confidence interval = (8–14) nSv h^{-1}]. The intercept can be interpreted as the sum of contributions from intrinsic noise of the spectrometer, cosmic radiation, the natural radionuclides ^{7}Be and ^{22}Na, and a residual ^{137}Cs contamination. Cosmic radiation and intrinsic noise of the spectrometer are the major contributors to $ADER_{reading}$ at zero value of AC_{eff}. These sources give 7 nSv h^{-1}. The ^{7}Be and ^{22}Na contributions to the ambient gamma dose rate are very small, far below 1

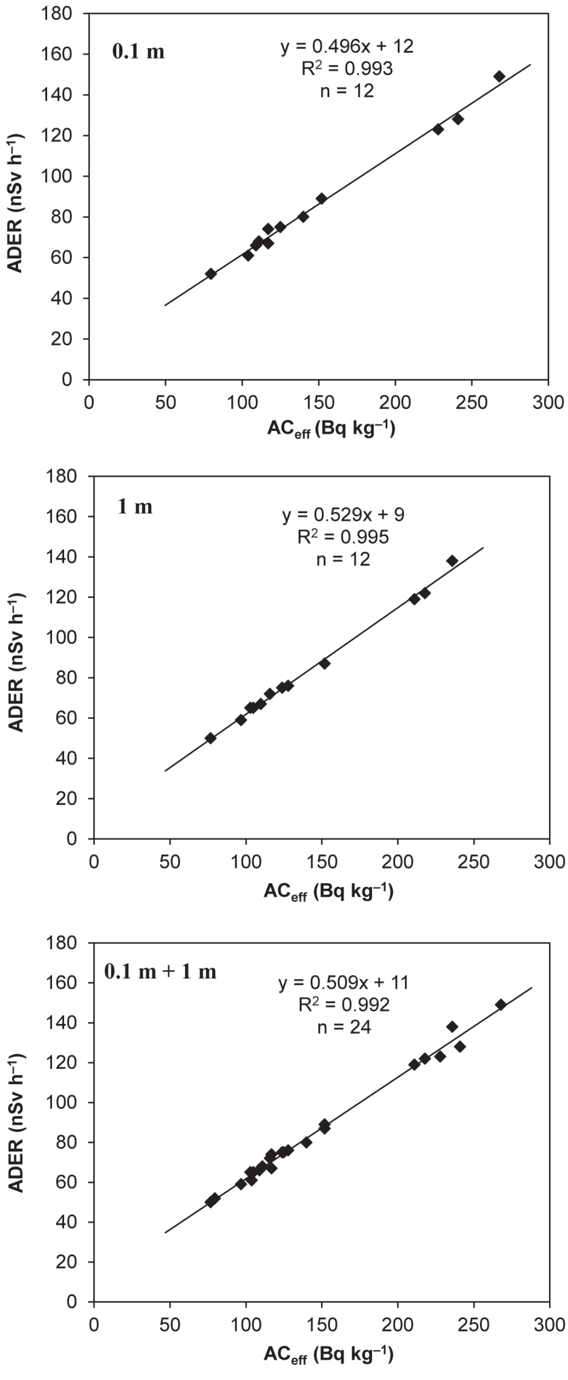

Fig. 3. Relationship between total ambient dose equivalent rate (ADER) and 'effective' activity concentration of terrestrial radionuclides (AC_{eff}) determined with *in situ* NaI(Tl) measurements (MKS AT1601D) at a height of 0.1 m and 1 m above the ground at background areas in St.-Petersburg and the Leningradskaya region in 2014–2016

nSv h^{-1} [3]. Using maximum expected level of the residual ^{137}Cs contamination of 4 kBq m^{-2} and the dose rate conversion factor DCF_{137Cs} = 1 nSv h^{-1} per 1 kBq m^{-2} [3], we can estimate the ^{137}Cs input to ADER at the background plots by a value of 4 nSv h^{-1}. The actual ^{137}Cs input may be substantially lower than the theoretical one because the DCF_{137Cs} value of 1 (nSv h^{-1})/(kBq m^{-2}) was deduced for undisturbed meadows before 2011. The soils at the majority of our plots had been tilled, which may reduce the normalized dose rate due to ^{137}Cs with a factor of ~2. For paved surfaces, the reduction factor for the DCF_{137Cs} may be even greater: ~ 6 [2].

3.2. Validation of the additional calibration

The validity of the derived relationship between $ADER_{TRN}$ and AC_{eff} was checked at 15 different plots (Table 4). Expected $ADER_{reading}$ (nSv h^{-1}) was calculated from measured AC_{eff} (Bq kg^{-1}) using the formula:

$$ADER_{reading} = DR_0 + CF \times AC_{eff}, \quad (2)$$

where DR_0 is the spectrometer-dosimeter reading at zero value of AC_{eff} (11 nSv h^{-1}) and CF is the conversion coefficient from AC_{eff} to $ADER_{TRN}$ [0.51 (nSv h^{-1})/(Bq kg^{-1})].

A very good consistency between the ADER calculated with AC_{eff} and the measured ADER has been found (the last column in Table 4). The deviation from unity for the ratio between calculated and measured values did not exceed 8%; the mean and median ratio was 0.99. To some extent, the good agreement is associated with the fact that the same spectrum was used for derivation of AC_{eff} and ADER, although different calibration procedures were used to derive values of AC_{eff} and ADER.

It is also interesting to compare the experimental value of 0.51 (nSv h^{-1})/(Bq kg^{-1}) for AC_{eff} to $ADER_{TRN}$ conversion coefficient with theoretical expectations. Bossew et al. [3] summarized available literature data and concluded that dose conversion coefficients differ remarkably between authors (eight references). For example, dose conversion coefficient for ^{238}U homogeneously distributed in soil varied from 0.357 to 0.463 (nGy h^{-1})/(Bq kg^{-1}). The Sv/Gy conversion coefficient for U series is estimated to be 1.253 (see [3] and references therein). Therefore, the theoretically expected conversion coefficient from AC_{eff} to $ADER_{TRN}$ may range from 0.45 (nSv h^{-1})/(Bq kg^{-1}) to 0.58 (nSv h^{-1})/(Bq kg^{-1}) with a mean value of 0.54 (nSv h^{-1})/(Bq kg^{-1}) and median value of 0.56 (nSv h^{-1})/(Bq kg^{-1}). Our experimental value of 0.51 (nSv h^{-1})/(Bq kg^{-1}) lies within the range and deviates less than 10% from the theoretical mean and median values.

Our calibration coefficients must be used with caution and only for the devices similar to the one employed in this study. We also suppose that each spectrometer-dosimeter should be additionally calibrated before using it for decomposition of ADER because systematic uncertainty of the AC_{eff} and ADER measurements is not negligible. It is declared by the manufacturer that this uncertainty may be as large as ±20% for determinations of activity concentration of terrestrial radionuclides and measurements of ADER [AT6101D Spectrometer. Operation manual, 2014]. Additional uncertainties of the ADER evaluation using the AC_{eff} data may be associated with variations in vertical distributions of radionuclides in soil or in its cover (e.g., asphalt, flagstones, granite gravel). It can be reasonably assumed that terrestrial radionuclides are distributed uniformly throughout a soil profile [2]. However, this assumption is hardly applicable for paved areas. The asphalted pavement has a complex structure consisting of asphalt layer, base and sub-base. Activity concentrations of natural radionuclides may vary between building materials used in construction of roads and pavements [25]. Nearby buildings may also influence the outdoor dose rate expected only from soil [10].

3.3. In field measurements in the Bryansk region

Table 5 shows the ambient dose equivalent rate, surface ground contamination with ^{137}Cs, activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th and their ‘effective’ activity concentrations determined at a height of 0.1 m above the ground at seven typical plots in contaminated areas in the Bryansk region, as measured in 2015–2016.

Activity concentrations of terrestrial radionuclides were in the ranges: 156–582 Bq kg^{-1} for ^{40}K, 10–30 Bq kg^{-1} for ^{226}Ra and 10–31 Bq kg^{-1} for ^{232}Th. Values of AC_{eff} varied from 32 Bq kg^{-1} to 119 Bq kg^{-1}. The lowest values were registered in the forest at Zatishie, while the highest activities were recorded at a street covered by asphalt. The median AC_{eff} in soils from the Bryansk region (67 Bq kg^{-1}, n = 6) was about 1.6 times lower in comparison with the level registered in soils from St.-Petersburg and the Leningradskaya region (111 Bq kg^{-1}, n = 20). The difference between the groups is statistically significant (the Mann–Whitney U test, $P < 0.01$).

Measured ground deposition densities (inventories) of ^{137}Cs (Table 5) ranged widely from 3.1 kBq m^{-2} to 447 kBq m^{-2}. The minimum inventory was found at a street in Novozybkov. The street had been covered with new asphalt several years after the Chernobyl accident. The low value of the ^{137}Cs inventory at the treated street shows that no significant re-contamination of the new asphalt surface has occurred. The maximum inventory was registered at the undisturbed grassland and forest sites located at the Novozybkov’s suburbs. The measured ground deposition density of ^{137}Cs at the forest and grassland (410–450 kBq m^{-2}) corresponds well to the contamination level officially reported for Novozybkov in 2014 (~400 kBq m^{-2}) [22]. The measured ground deposition density of ^{137}Cs in the forest at Zatishie (45 kBq m^{-2}) also correlates very well with the officially established level of ^{137}Cs contamination: 41 kBq m^{-2} [22]. The measured inventories of ^{137}Cs at the kitchengarden Les-13gar, yard Les-13ya and grassland Les-13gr in Novozybkov appeared to be 2–3 folds less than the expected ^{137}Cs inventory of ~400 kBq m^{-2}. The observed difference can be explained by a deep penetration of ^{137}Cs at these three plots. The ground areas had been dug by the owner many times after the Chernobyl accident. These observations indicate the necessity of further calibration and testing of AT6101D spectrometer with respect to the quantitative determination of ^{137}Cs in undisturbed and disturbed soils.

Table 6 shows measured ADER, ‘effective’ activity concentration of natural radionuclides and estimations of dose rates due to: 1) terrestrial radionuclides ($ADER_{TRN}$), and 2) ^{137}Cs ($ADER_{Cs\text{-}tot}$) for the height of 1 m above the ground at contaminated areas in the Bryansk region.

Table 6

Ambient dose equivalent rate ($ADER_{reading}$) and ‘effective’ activity concentration of terrestrial radionuclides (AC_{eff}) determined with AT1601D, and estimations of dose rates due to terrestrial radionuclides ($ADER_{TRN}$) and ^{137}Cs ($ADER_{Cs\text{-}tot}$) at a height of 1 m above the ground at contaminated areas in the Bryansk region in 2015–2016

Settlement	Code of plot	Location	$ADER_{reading}$ (nSv h^{-1})[c]	AC_{eff} (Bq kg^{-1})	$ADER_{TRN}$ (nSv h^{-1})	$ADER_{Cs\text{-}tot}$ (nSv h^{-1})	^{137}Cs input to ADER (%)
Novozybkov	Les-13str	street	107	101 (14)	51.5 (14)	47.5 (17)	44
Novozybkov	Les-13ya	yard	184	82.2 (13)	41.9 (14)	134 (4.9)	73
Novozybkov	Les-13kit	kitchengarden	241	84.4 (11)	43.0 (12)	190 (3.4)	79
Novozybkov	Les-13gr[a]	grassland	242	84.7 (14)	43.2 (15)	191 (4.0)	79

The end of table 6

Settlement	Code of plot	Location	$ADER_{reading}$ ($nSv\ h^{-1}$)[c]	AC_{eff} ($Bq\ kg^{-1}$)	$ADER_{TRN}$ ($nSv\ h^{-1}$)	$ADER_{Cs\text{-}tot}$ ($nSv\ h^{-1}$)	^{137}Cs input to ADER (%)
Novozybkov	Fil-gr[b]	grassland	546	40.4 (15)	20.6 (15)	517 (2.1)	95
Novozybkov	Fil-fo	forest	531	48.3 (13)	24.6 (14)	498 (2.1)	94
Zatishie	Zat-fo	forest	77.6	30.0 (16)	15.3 (17)	54.3 (5.4)	70

[a] – the plot is located inside settlement.
[b] – the plot is located outside settlement.
[c] – the primary reading of the AT1601D spectrometer-dosimeter. A statistical uncertainty (at the 2 sigma level) of the ADER measurements is less than ±2%.
Statistical uncertainties (±%, at the 2 sigma level) for determination of AC_{eff}, $ADER_{TRN}$ and $ADER_{Cs\text{-}tot}$ are given in brackets.

Ambient dose equivalent rate at a height of 1 m above the ground ranged from 78 $nSv\ h^{-1}$ to 546 $nSv\ h^{-1}$. The median value of ADER at the Bryansk region (241 $nSv\ h^{-1}$; n = 7) is about three times higher than the one in St.-Petersburg and the Leningradskaya region (74 $nSv\ h^{-1}$; n = 12). The difference between the contaminated and background regions is statistically significant (the Mann–Whitney U test, $P < 0.01$). No correlation was found between AC_{eff} and ADER at the contaminated plots. Spearman's coefficient of correlation is -0.143 ($P > 0.05$; n = 7). At the same time, ADER and ^{137}Cs ground deposition densities at the plots (n = 7) were strongly positively correlated; Spearman's rank correlation coefficient has been calculated as 0.929 ($P < 0.05$).

Total dose rate due to ^{137}Cs, $ADER_{Cs\text{-}tot}$ ($nSv\ h^{-1}$), was calculated using the following formula:

$$ADER_{Cs-tot} = ADER_{reading} - DR_0 - CF \times AC_{eff} \quad (3)$$

where $ADER_{reading}$ is the dosimeter reading; DR_0 is the expected reading of the dosimeter at zero value of AC_{eff} and no ^{137}Cs contamination (8 $nSv\ h^{-1}$); AC_{eff} is 'effective' activity concentration of terrestrial radionuclides ($Bq\ kg^{-1}$); CF is the conversion coefficient from AC_{eff} to $ADER_{TRN}$ [0.51 ($nSv\ h^{-1}$)/($Bq\ kg^{-1}$)].

Note that the DR_0 value of 8 $nSv\ h^{-1}$ includes the value of 7 $nSv\ h^{-1}$ obtained during measurements above the surface of the Finish Gulf. Additionally, 1 $nSv\ h^{-1}$ may be attributed to: 1) contribution from natural radionuclides different from ^{238}U series, ^{232}Th series and ^{40}K, and 2) a small difference in altitudes between the surface of the Finnish Gulf (0 m a.s.l.) and the terrains in the Bryansk region (150–170 m a.s.l.).

Calculated values of $ADER_{Cs\text{-}tot}$ are shown in column 7 at Table 6, while the contribution of the man-made source to $ADER_{reading}$ is presented in column 8 at Table 6. In Novozybkov, the contribution of ^{137}Cs to the total ambient dose rate is over 90% at the undisturbed grassland and forest, about 75% at disturbed ground plots and about 40% at an asphalted street. This can be explained by the high residual ^{137}Cs contamination of soils in Novozybkov: ~ 400 $kBq\ m^{-2}$. The level of the ^{137}Cs contamination in Zatishie (~ 40 $kBq\ m^{-2}$) was about ten times lower than that in Novozybkov. Nevertheless, the artificial source dominated the ambient dose rate in the Zatishie's forest where the total measured ADER (78 $nSv\ h^{-1}$) was comparable with the average ADER (68 $nSv\ h^{-1}$) determined at nine ground plots from the Leningradskaya region. To a great extent, the strong contribution (70%) of ^{137}Cs to the total dose in the Zatishie's forest is attributable to the relatively low activity concentrations of natural radionuclides in soil (AC_{eff} = 31.6 $Bq\ kg^{-1}$; Table 5) and the relevant gamma dose rate in air at a height of 1 m above the ground ($ADER_{TRN}$ = 15 $nSv\ h^{-1}$; Table 6).

The tested spectrometer-dosimeter and technology to separate the natural and ^{137}Cs components of the ambient dose equivalent rate can be applied not only for the Chernobyl-affected territories but also for other sites where radioactive contamination is fully dominated by ^{137}Cs. For example, these may be areas contaminated as a result of the accident happened in Elektrostal (Russia) in April 2013, or sites of the peaceful underground nuclear explosions which were carried out in the USSR in the last century [26, 27]. Potentially, the territories contaminated by radiocaesium (^{137}Cs + ^{134}Cs) after Fukushima accident (2011) can be surveyed using the AT6101D device, although some correction for the presence of ^{134}Cs might be required.

4. Conclusions

The results of the measurements confirm the applicability of in situ gamma-ray spectrometry for decomposition of the ambient dose equivalent rate outdoors. After additional calibration it is possible to use a commercially available NaI(Tl) gamma-ray spectrometer-dosimeter to separate the natural and ^{137}Cs components of the ambient dose equivalent rate. The additional calibration requires performing *in situ* measurements at the environment which has negligible contamination by ^{137}Cs (at the level of a few $kBq\ m^{-2}$). The conversion coefficient from AC_{eff} to $ADER_{TRN}$ of 0.51 ($nSv\ h^{-1}$)/($Bq\ kg^{-1}$) has been obtained using a regression analysis of experimental data. The intrinsic noise of the spectrometer and its response to cosmic radiation at sea level has been estimated to 7 $nSv\ h^{-1}$. The dosimeter-spectrometer and experimentally derived calibration coefficients have been tested in field in the Bryansk region that had been heavily contaminated by Chernobyl fallout. The contribution of ^{137}Cs to the total ADER varies between 40% and 95%. Currently further studies are conducted to: 1) obtain new data from areas contaminated by ^{137}Cs, and 2) derive calibration coefficients for indoor locations and for backpack based radiation detection systems.

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Калибровка и опробование портативного NaI(Tl) гамма-спектрометра-дозиметра для оценки вкладов терригенных радионуклидов и ^{137}Cs в мощность амбиентного эквивалента дозы на открытом воздухе

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Коммерчески доступный сцинтилляционный NaI(Tl) спектрометр-дозиметр был использован для дифференциальной оценки вклада естественных радионуклидов (семейство ^{238}U, семейство ^{232}Th и ^{40}K) и ^{137}Cs в мощность амбиентного эквивалента дозы (МАЭД), $\dot{H}^*(10)$, определенную на открытом воздухе. Прибор был изначально откалиброван производителем для измерения: 1) МАЭД; 2) удельной активности ^{226}Ra, ^{232}Th, ^{40}K и удельной эффективной активности естественных радионуклидов ($А_{эфф}$) в почве, сельскохозяйственной продукции, продуктах лесного хозяйства и строительных материалах; 3) плотности поверхностного загрязнения ^{137}Cs. С целью определения значения коэффициента перехода от $А_{эфф}$ к МАЭД, обусловленной естественными радионуклидами ($МАЭД_{ест}$), мы провели две серии дополнительных калибровочных измерений. Эти измерения были выполнены на открытом воздухе на 27 площадках, расположенных в г. Санкт-Петербург и в Ленинградской области. Полученные результаты были обработаны с использованием метода линейного регрессионного анализа. Значение коэффициента перехода от $А_{эфф}$ к $МАЭД_{ест}$ оказалось равным 0,51 (нЗв/ч)/(Бк/кг). Этот коэффициент перехода рекомендуется применять с осторожностью и только для детекторов, похожих на тот, который был использован в настоящей работе (сборка на основе монокристалла NaI(Tl) 63 мм в диаметре и 63 мм в длину). Сумма собственного фона прибора и его отклика на космическое излучение на уровне моря была оценена величиной 7 нЗв/ч. Спектрометр-дозиметр и экспериментально полученные калибровочные факторы были опробованы в полевых условиях в Брянской области на семи площадках, сильно загрязненных чернобыльскими выпадениями. Вклад ^{137}Cs в общую МАЭД колебался от 40% до 95%. Предварительные результаты настоящего исследования подтвердили перспективность использования полевой гамма-спектрометрии для оценки вкладов природных и техногенных источников в мощность амбиентного эквивалента дозы на открытом воздухе.

Ключевые слова: *гамма-излучение, спектрометр, NaI(Tl), дозиметр, мощность амбиентного эквивалента дозы, естественные радионуклиды, ^{40}K, ^{226}Ra, ^{232}Th, ^{137}Cs.*

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