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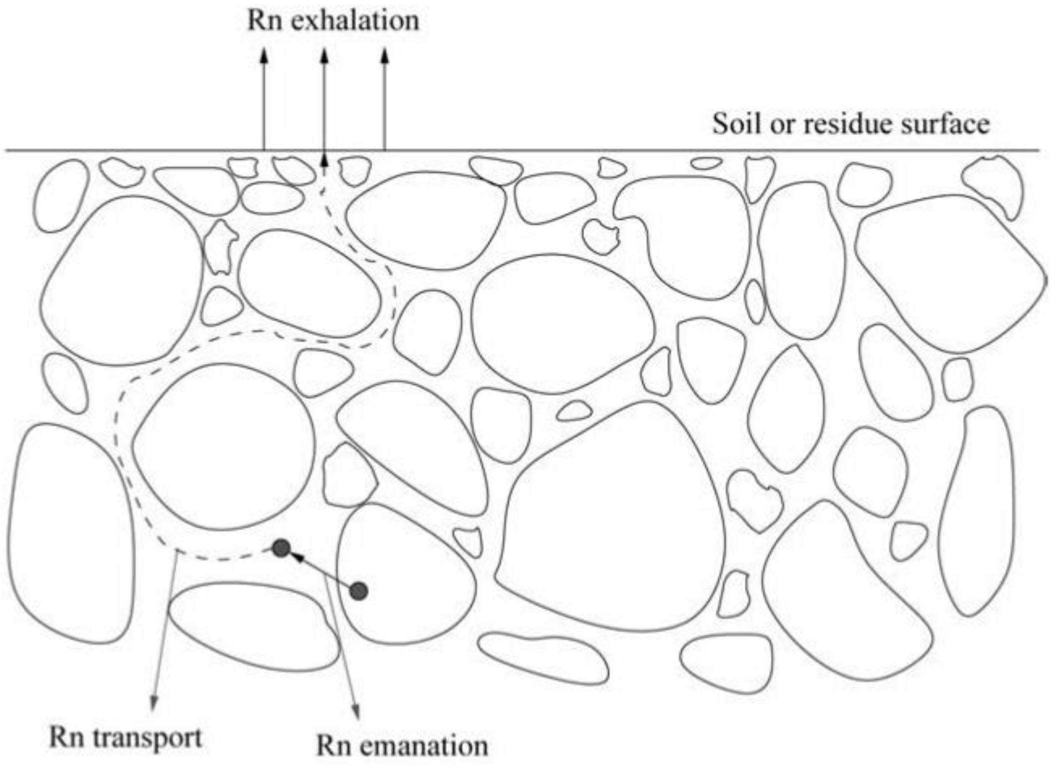
Radon in Soil Gas Measurements by RAD7 Active Device and Its Possible Correlation with Soil Grain Size and Composition

Sofija Forkapić[1], Robert Lakatoš[2], Selena Samardžić[2], Peter Jovanovič[3], Kristina Bikit-Schroeder[1], Dušan Mrđa[1]

[1]Faculty of Sciences, University of Novi Sad, Trg Dositeja Obradovića 3, Novi Sad, Serbia
[2]Faculty of Technical Sciences, University of Novi Sad, Trg Dositeja Obradovića 6, Novi Sad, Serbia
[3]Zavod za varstvo pri delu, Chengdujska cesta 25, Ljubljana, Slovenia

- *Soil is the most important factor affecting the radon level in the human living environments. It depends not only on uranium and radium contents but also on the physical and chemical properties of the soil.*
- *In this paper, the measurements of radon concentrations in soil gas determined by active radon monitor RAD7 in Vojvodina region, Northern Province of Serbia, are presented.*
- *^{226}Ra and ^{238}U activity concentrations in soil samples were measured by gamma ray spectrometry, while the grain size of soil samples has been determined by Malvern Mastersizer 2000 laser diffraction particle size analyzer.*
- *Soil gas radon concentration ranges between 5 and 46 kBq/m^3 indicating a low natural radiation area. The results of radionuclide content in soils were found to vary from 17,6 Bq/kg to 39,4 Bq/kg for radium and from 15 Bq/kg to 51 Bq/kg for uranium, respectively.*
- *As a result of standard regression analysis between the measured values, model that includes soil grain composition, radium and uranium content as predictors with 63.7% affects the radon concentration in soil.*



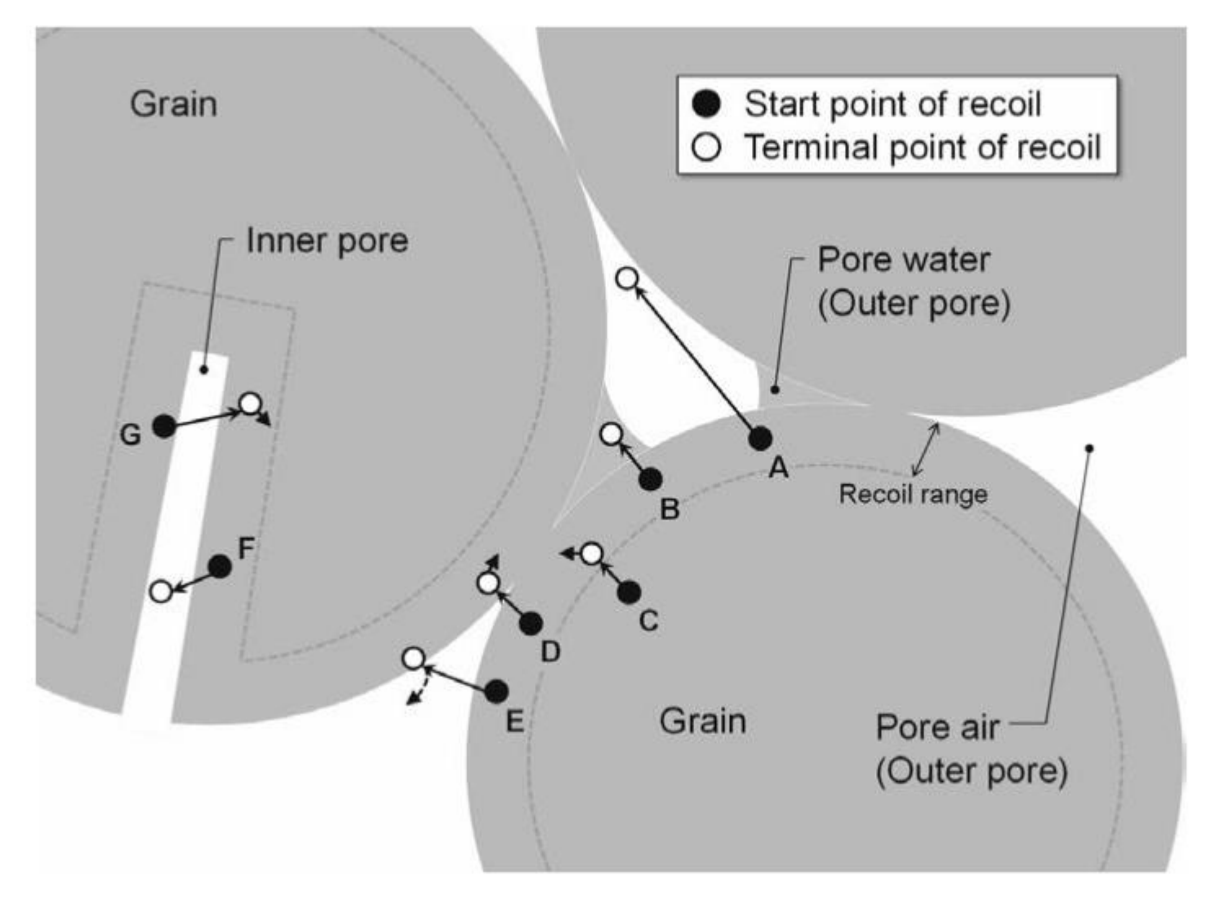
IAEA TRS 474: *Measurement and Calculation of Radon Releases from NORM Residues*, Y. Ishimori, K. Lange,P. Martin, Y.S. Mayya, M.Phaneuf, Vienna 2013

- (a) Emanation — radon atoms formed from the decay of radium escape from the grains (mainly because of recoil) into the interstitial space between the grains.
- (b) Transport — diffusion and advective flow cause the movement of the emanated radon atoms through the residue or soil profile to the ground surface.

(c) Exhalation— radon atoms that have been transported to the ground surface and then exhaled to the atmosphere.

Factors affecting radon emanation

- Radium distribution, particle size and shape
- Moisture content
- Mineralogical aspects such as lattice structure, porosity, grain shape, and elemental composition



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soil probe method for the measurement of radon concentration at different depths using a continuous radon monitor

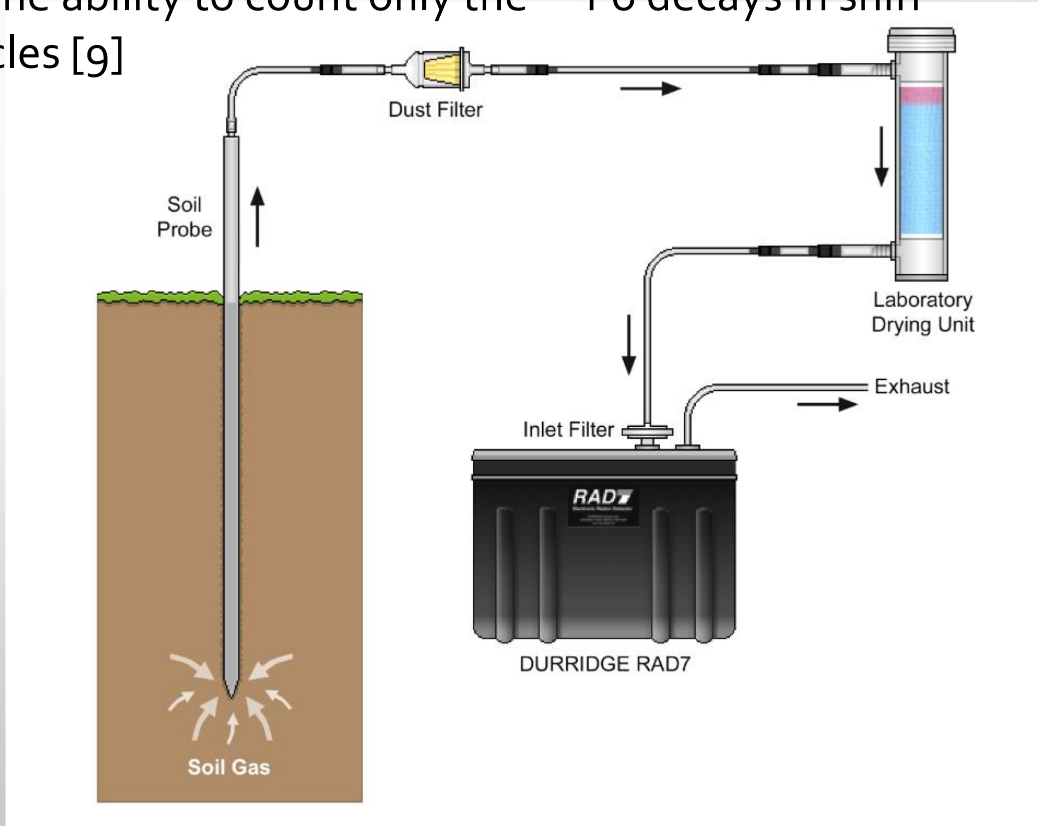
Soil gas radon activity concentrations were measured in situ by RAD7 alpha-spectrometer (DURRIDGE Company) with stainless soil gas probe using grab protocol. While pumping, the air flow rate is about 0.7 l/min and therefore 3.5 l of soil gas is extracted from the soil at the depth of 80 cm [8]. The ability to count only the ^{218}Po decays in sniff mode means that the measurements are unaffected by other particles [9]

$$C(z) = C_\infty\left(1 - \exp\left(-z/L_r\right)\right)$$

where

C_∞ is the radon concentration at great depth (Bq/m^3);
z is the thickness of the material (m);

and L_r is the radon diffusion length (m).



The intercomparison of continuous radon active monitors RAD7

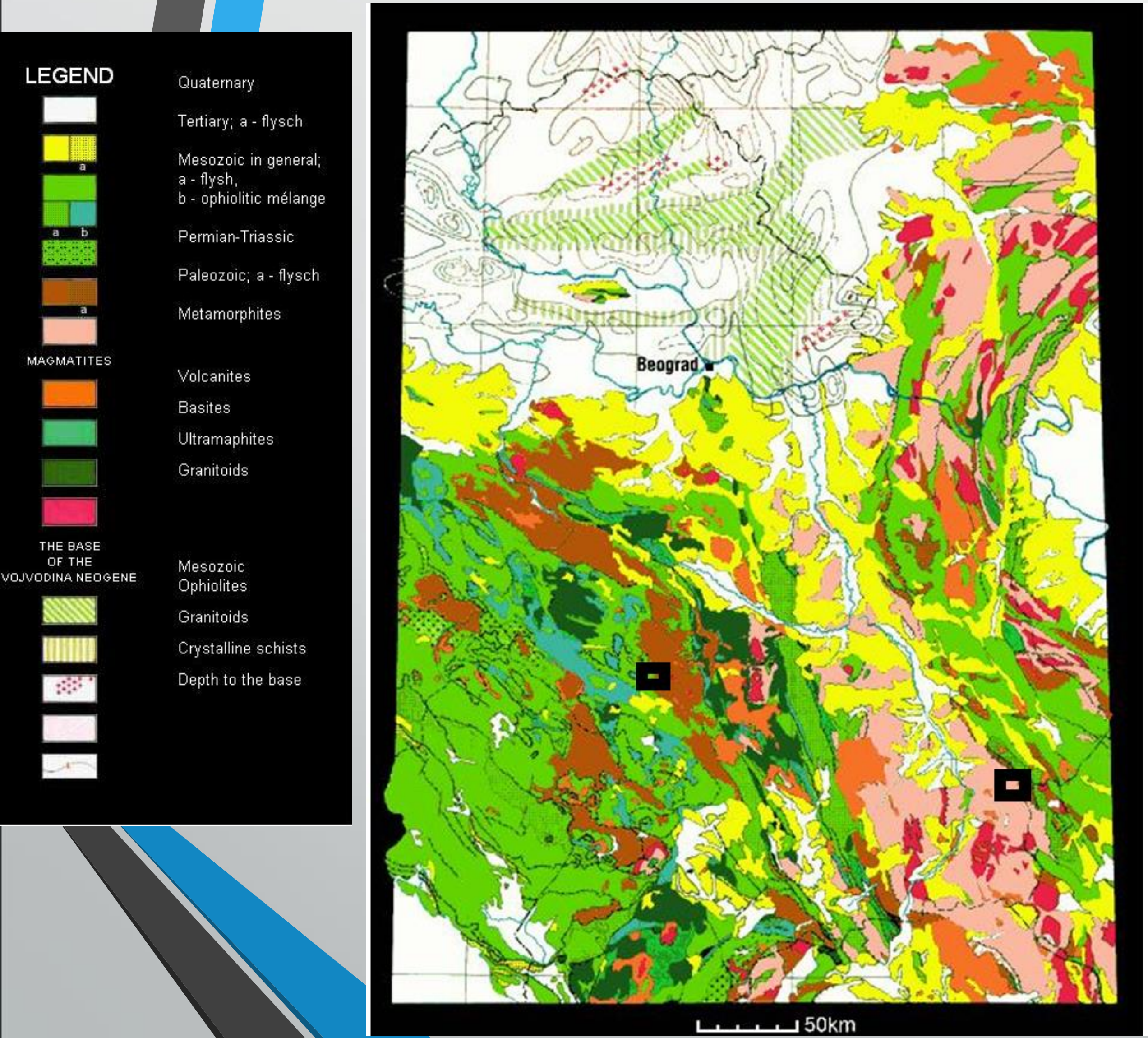
- 2017 in Ljubljana by two accredited laboratories for these measurements>
- – Zavod za varstvo pri delu – ZVD, Slovenia
- – Laboratory for radioactivity and dose measurements – PMF NS, Serbia.
- It was focused on investigation of consistency results of laboratories respect to same samples in real measurement conditions.
- grab protocol with sniff mode at the field near ZVD at two sites located at the distance of 78 cm from each other .
- The stainless soil gas probes were inserted in soil to the depth of 80 cm
- The last calibration of used devices was performed in radon chamber at the accredited trial metrological Lab. SUJCHBO Kamenna, Czech Republic in 2015. Calibration laboratory is traceable to PTB Braunschweig, Germany.

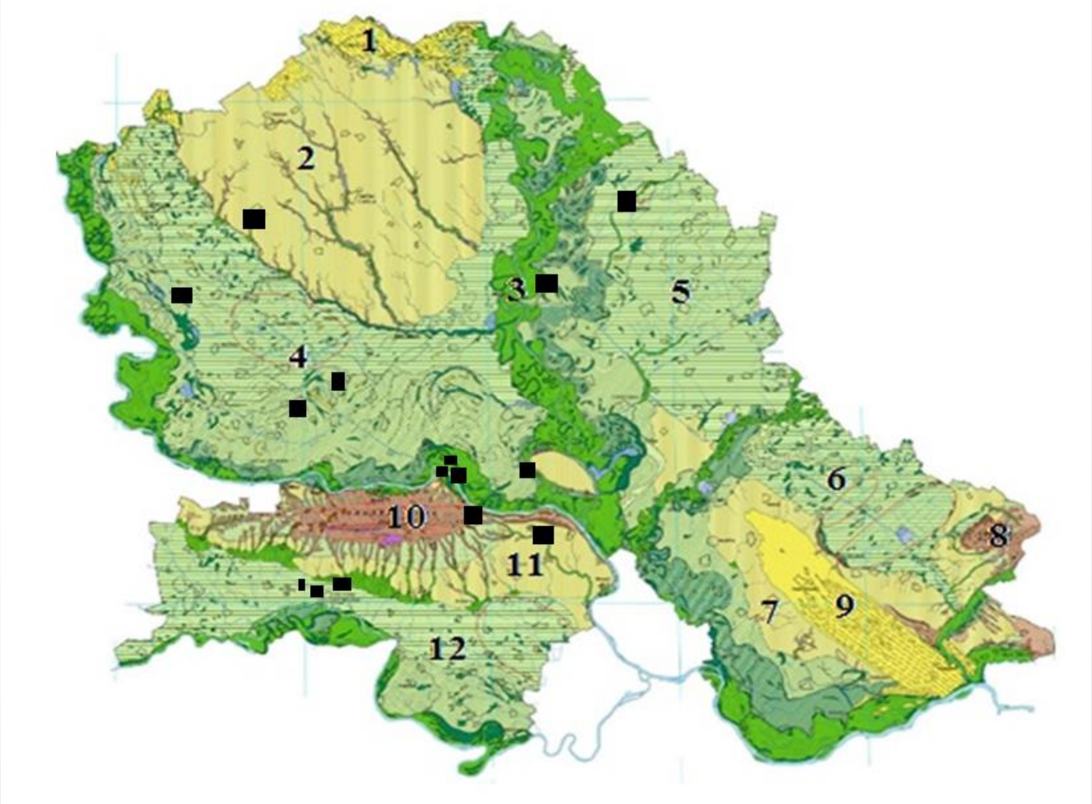
Soil gas measurements intercomparison results

Soil, site 1 – lab.code 1			**Soil, site 2 – lab.code 1**		
Mode	Radon in soil [Bq/m3]	2σ Uncert. [Bq/m3]	Mode	Radon in soil [Bq/m3]	2σ Uncert. [Bq/m3]
Sniff	31600	2300	Sniff	26400	2100
Sniff	35600	2500	Sniff	29000	2200
Sniff	39000	2600	Sniff	30500	2300
Sniff	45000	5300	Sniff	32400	2400
Reported value	45000	5300	Reported value	32400	2400
Soil, site 1 – lab.code 2			**Soil, site 2 – lab.code 2**		
Mode	Radon in soil [Bq/m3]	2σ Uncert. [Bq/m3]	Mode	Radon in soil [Bq/m3]	2σ Uncert. [Bq/m3]
Sniff	41236	3292	Sniff	30899	2787
Sniff	44382	3674	Sniff	32584	3056
Sniff	43708	3674	Sniff	32135	3090
Sniff	42584	3685	Sniff	33596	3180
Reported value	44382	3674	Reported value	33596	3180
Bias %	**1.4 %**		**Bias %**	**4 %**	

- The study was conducted in urban settlements on 14 locations with high and moderate level of indoor radon. Criterion for measurement points selection was also to cover all geomorphological units in Vojvodina region [6]: two mountains, four loess plateaus, three loess terraces, four alluvial plains and two sandstone terrains.
- For results evaluation SPSS (Statistical Package for the Social Sciences) statistics were used to see whether there is a correlation between measured activities. In order to predict the radon concentrations in soil, standard regression analysis was performed with ^{238}U, ^{226}Ra, and the soil grain size and soil composition as predictors.
- The connection between radium and uranium content in soil and radon emanation in soil might be used in modeling the radon risk.
- The radon risk classification in many countries in Europe is based on the soil gas radon measurements and permeability determination according to geogenic radon potential concept [7]. The main advantage of this concept is given by the fact that the radon risk is independent on human influence and temporally constant over a geological timescale.

Geomorphologic units and locations of sampling





Horgoš sands (1), Bačka loess plateau (2), Alluvial plain (3), Bačka loess terrace (4), Banat loess terrace (5), Tamiš loess plateau (6), Deliblato loess plateau (7), Vršac mountains (8), Deliblato sands(9), Fruška Gora mountain (10), Fruška Gora loess plateau (11) and Srem loess terrace (12

Gamma spectrometry measurements of soil

On each measurement point soil sample was taken from the surface layer (5–30 cm) and after drying and homogenization packed in cylindrical geometry 62 mm x 67 mm. Typical mass of samples was 200-300 g and measurement time was 80 ks. Activity concentrations of radionuclides in soil were determined by low-level gamma spectrometry method on shielded HPGe detectors with maximal background reduction.

The gamma spectra were acquired and analyzed using the Canberra Genie 2000 software. A special procedure developed in the Novi Sad laboratory was used for the determination of the ^{238}U activity concentration from gamma-lines of the first progeny of this radionuclide, ^{234}Th .

MDA according to time of measurements for each radionuclide:

9 Bq/kg za ^{238}U,
2.5 Bq/kg za ^{226}Ra,
1.3 Bq/kg za ^{232}Th,
13.3 Bq/kg za ^{40}K i
1.2 Bq/kg za ^{137}Cs.

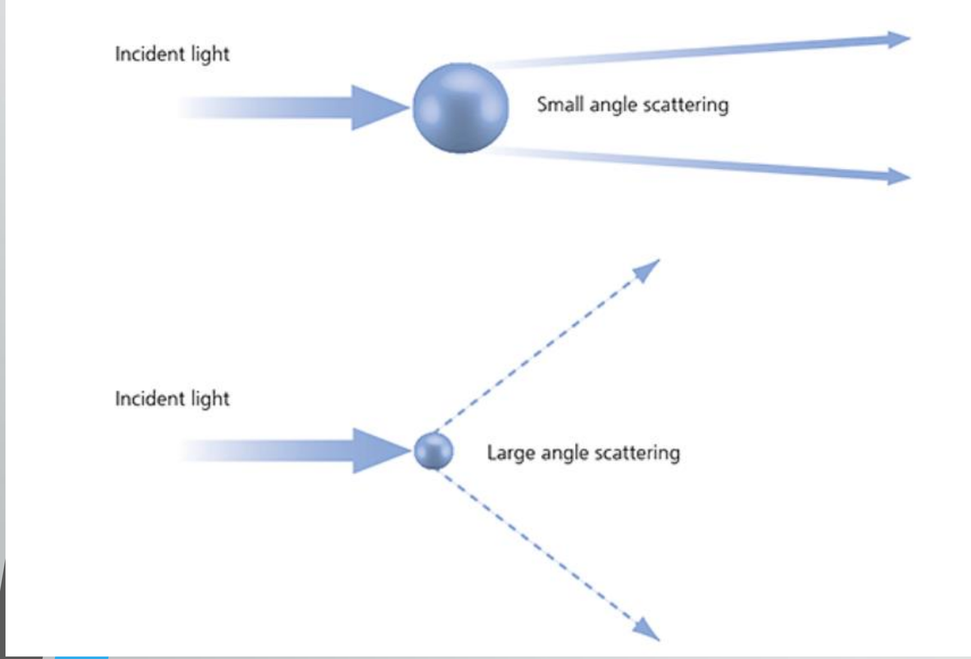
Particle size distribution

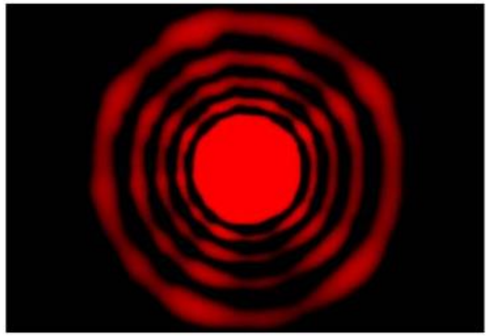
- Volume distribution of soil grain sizes was determined by laser diffraction dry method with particle size analyzer Mastersizer 2000, Malvern Instruments at the Faculty of Technical Sciences, Novi Sad

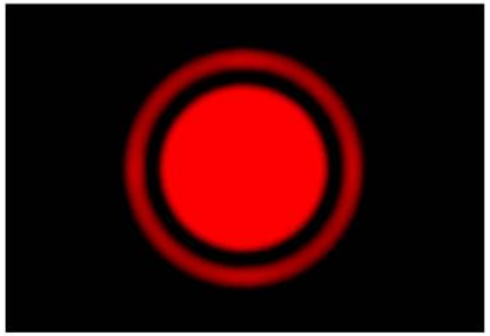
- A particle size distribution analysis was conducted using a Malvern Mastersizer 2000 Particle Size Analyzer capable of analyzing particles between 0.02 µm and 2000 µm. The Malvern Mastersizer 2000 records the light pattern scattered from a field of particles at different angles. The result of the analysis is the relative distribution of the volume or surface of spherically shaped particles in a range of size classes. Based on particle size analysis the following fractions were determined: coarse sand (500-2000 µm), medium sand (250-500 µm), fine sand (62.5-250 µm), silit (3.9-62.5 µm), and clay (<3.9 µm).

Laser diffraction is a widely used particle sizing technique for materials ranging from hundreds of nanometers up to several millimeters in size

Laser diffraction measures particle size distributions by measuring the angular variation in intensity of light scattered as a laser beam passes through a dispersed particulate sample.







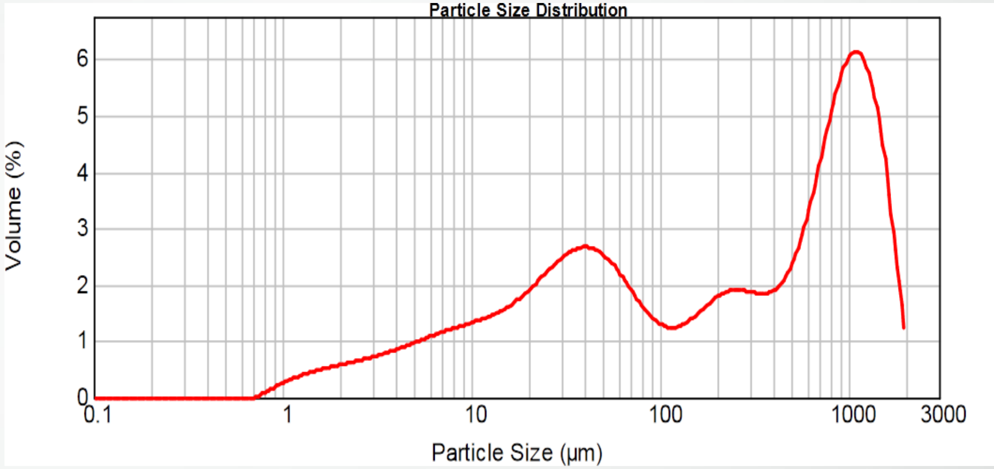


Figure 1. Particle size distribution moved to bigger particles

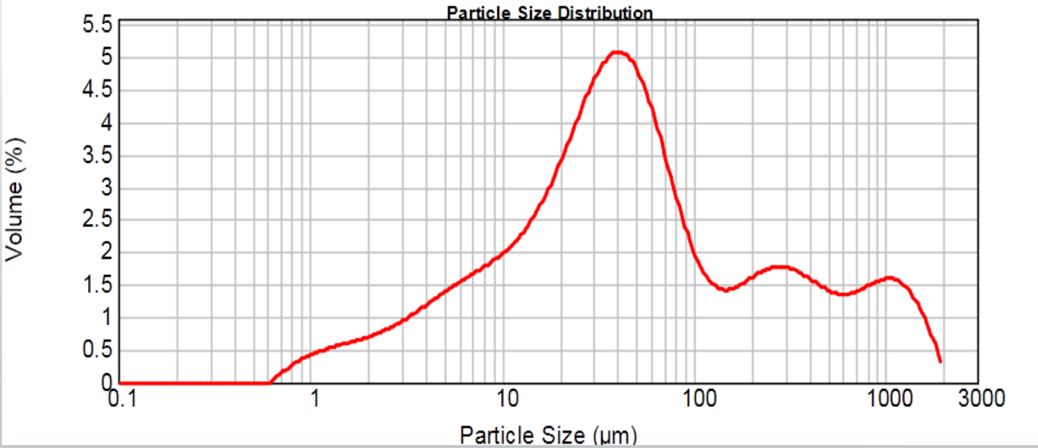
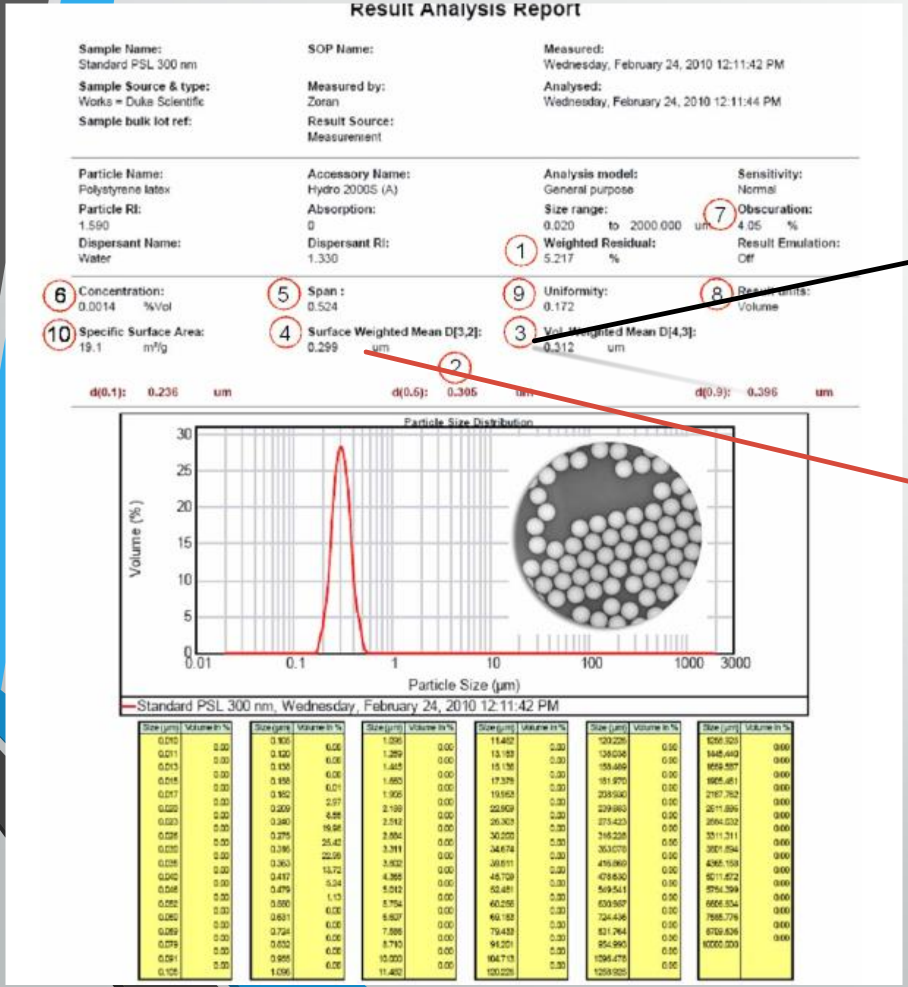


Figure 2. Particle size distribution moved to smaller particles

Volume weighted mean and surface weighted mean



Result Analysis Report

Sample Name: Standard PSL 300 nm
SOP Name:
Measured: Wednesday, February 24, 2010 12:11:42 PM
Sample Source & type: Works = Duke Scientific
Measured by: Zoran
Analysed: Wednesday, February 24, 2010 12:11:44 PM
Sample bulk lot ref:
Result Source: Measurement

Particle Name: Polystyrene latex
Accessory Name: Hydro 2000S (A)
Analysis model: General purpose
Sensitivity: Normal
Particle RI: 1.590
Absorption: 0
Size range: 0.020 to 2000.000 um
7 Obscuration: 4.05 %
Dispersant Name: Water
Dispersant RI: 1.330
1 Weighted Residual: 5.217 %
Result Emulation: Off

6 Concentration: 0.0014 %Vol
5 Span : 0.524
9 Uniformity: 0.172
8 Result units: Volume
10 Specific Surface Area: 19.1 m²/g
4 Surface Weighted Mean D[3,2]: 0.299 um
3 Vol. Weighted Mean D[4,3]: 0.312 um
2
d(0.1): 0.236 um
d(0.5): 0.305 um
d(0.9): 0.396 um

$$d_{mm} = \frac{\sum m_1 d_i}{M} = \frac{(\pi\rho_p/6)\sum n_i d_i^3 d_i}{(\pi\rho_p/6)\sum n_i d_i^3} = \frac{\sum n_i d_i^4}{\sum n_i d_i^3}$$

$$d_{sm} = \frac{\sum s_1 d_i}{S} = \frac{\sum n_i d_i^3}{\sum n_i d_i^2}$$

Indoor radon gas concentrations, maximum ambient dose equivalent in contact to walls and floor

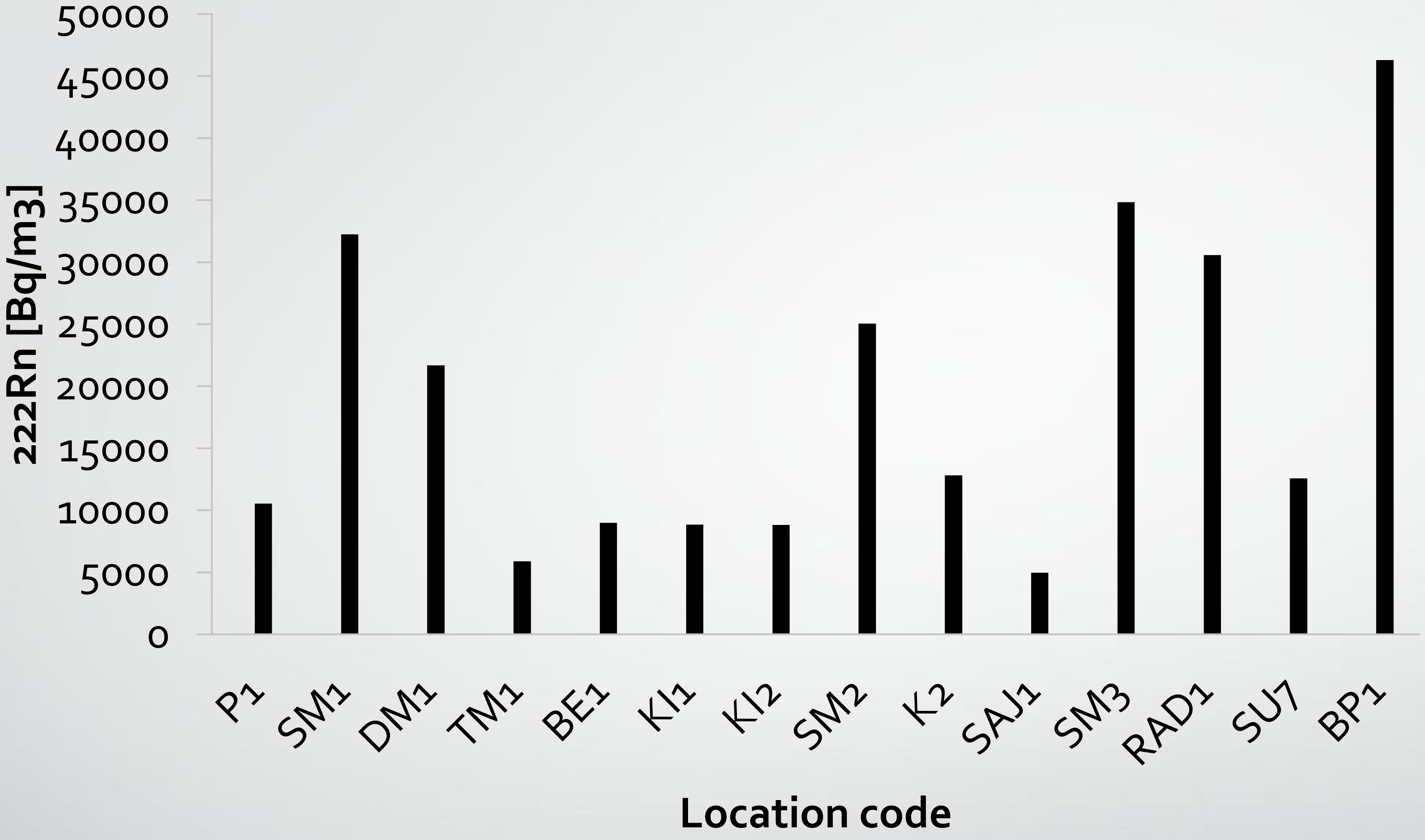
Location code	CR39 [Bq/m^3]	Charcoal canisters [Bq/m^3]	RAD7 ^{222}Rn [Bq/m^3]	H*(10) [μSv/h] walls	H*(10) [μSv/h] floor	BM walls	BM floor
SM1	188	46.2(2.1)	135	0.215	0.182	brick block	concrete
SM2	627	380(6)	75	0.221	0.201	brick	concrete
SM3	338	128(3)	133	0.257	0.225	brick, adobe	wood and soil
BP1	667	334(11)	109	-	-	brick	concrete
K2	481	144(6)	42	-	-	concrete. brick	concrete
P1	370	173(10)	140	0.154	0.139	brick, adobe	wood and soil
SAJ1	-	26(4)	16.5	0.233	0.209	brick and brick block	concrete
BE1	96	79(6)	36.2	0.197	0.251	brick, thermo insulation	concrete
DM1	126	110(7)	116	0.167	0.191	brick	concrete
TM1	-	90(6)	39.5	0.143	0.125	brick	concrete
KI1	28	54(6)	36.3	0.161	0.143	brick, concrete, brick block	concrete
KI2	-	161(7)	238	0.185	0.205	brick, brick block	wood and soil
SU7	315	262(9)	99	0.205	0.191	concrete, brick	concrete
SU8	455	270(9)	384	0.173	0.143	brick.5	concrete
RAD1	820	930(20)	638	0.167	0.175	stone. brick	stone
VR1	605	664(20)	524	0.155	0.173	Brick, brick block	concrete

Gamma spectrometry analysis of soil and radon in soil gas

Location code	^{238}U [Bq/kg]	^{226}Ra [Bq/kg]	^{232}Th [Bq/kg]	^{40}K [Bq/kg]	^{137}Cs [Bq/kg]	^{222}Rn in soil [Bq/m^3]
SM1	28.9±1.4	29.8±1.4	44±3	515±28	6±0.7	32247(2787)
SM2	31±12	33±5	47.2±2.9	558±22	6.5±0.6	25056(2506)
SM3	39.3±1.9	39.4±1.9	46±10	530±30	9.2±1.1	34831(3034)
BP1	25±11	37±6	53±3	564±22	6±0.6	46292(3483)
K2	24±8	30.0±2.5	41.3±2.1	511±28	2±0.6	12809(1753)
P1	34±7	28.8±1.9	35±6	469±16	9.2±0.7	10528(1573)
SAJ1	51±13	33±3	34±6	540±30	3.1±1.1	4977(1213)
BE1	26±21	24.2±1.4	34±8	435±27	6.1±0.9	8989(1506)
DM1	15±8	23.0±2.2	28.8±2.6	454±25	4.4±0.6	21685(2270)
TM1	37±5	26.3±1.7	31.5±2.0	430±40	5.5±0.6	5888(1202)
KI1	17.5±2	17.6±1.4	27.4±2.0	499±26	1.9±0.3	8843(1472)
KI2	37±5	26.3±1.7	31.5±2.0	430±40	5.5±0.6	8820(1461)
SU7	28±9	33.8±2	32±10	463±23	3.3±0.6	12584(1730)
SU8	34±8	38±4	45±10	574±19	4.4±0.5	1103(561)
RAD1	36±5	22.7±0.8	31.2±2.3	530±80	45.7±1.7	30562(2764)
VR1	55±6	30.1±0.9	39±3	560±50	14.2±0.8	

Mechanical content and grain size of soil

Location code	Clay <3.9 µm [%]	Silit 3.9-62.5 µm [%]	Fine sand 62.5-250 µm [%]	Medium sand 250-500 µm [%]	Coarse sand 500-2000 µm [%]
SM1	4.18	37.75	25.14	17.24	15.75
SM2	6.62	38.99	16.63	11.45	26.27
SM3	7.41	38.58	18.47	16.92	18.61
BP1	9.03	50.46	16.08	10.41	14.02
K2	6.82	38.53	15.89	5.36	33.39
P1	3.75	38.31	26.50	19.61	11.83
SAJ1	7.22	43.92	24.52	11.67	12.73
BE1	4.89	35.39	26.35	13.51	19.86
DM1	4.40	25.08	49.21	4.90	16.41
TM1	4.52	38.29	44.52	7.45	5.22
KI1	5.92	29.35	27.93	29.21	7.56
KI2	5.96	32.48	14.14	9.13	38.28
SU7	8.67	57.58	13.54	4.49	15.72
SU8	8.13	56.49	17.19	7.24	10.95
RAD1	7.68	27.46	35.87	22.63	6.36
VR1	5.43	33.31	37.31	20.49	3.46

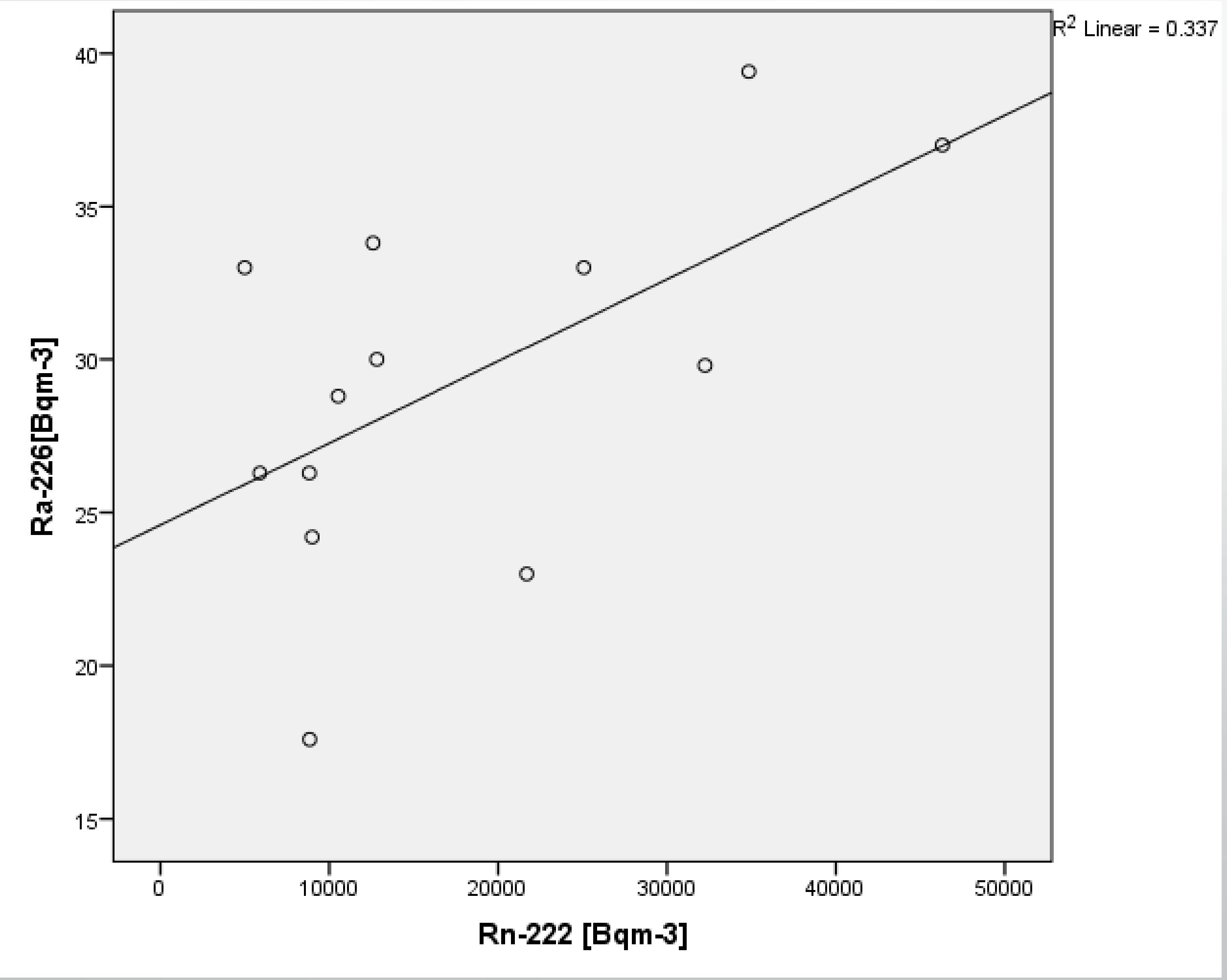


Soil gas radon concentrations arranged in ascending order of clay component content of soil samples (x-axis)

Location code	Surface Weighted Mean [µm]	Vol. Weighted Mean [µm]	^{238}U [Bq/kg]	^{226}Ra [Bq/kg]	^{222}Rn in soil [Bq/m^3]
SM1	23.367	281.480	28.9±1.4	29.8±1.4	32247(2787)
SM2	17.148	355.711	31±12	33±5	25056(2506)
SM3	15.900	282.726	39.3±1.9	39.4±1.9	34831(3034)
BP1	12.502	217.508	25±11	37±6	46292(3483)
K2	16.890	431.599	24±8	30.0±2.5	12809(1753)
P1	24.882	234.298	34±7	28.8±1.9	10528(1573)
SAJ1	15.627	221.245	51±13	33±3	4977(1213)
BE1	21.426	321.311	26±21	24.2±1.4	8989(1506)
DM1	25.761	274.794	15±8	23.0±2.2	21685(2270)
TM1	21.801	153.585	37±5	26.3±1.7	5888(1202)
KI1	20.347	219.475	17.5±2	17.6±1.4	8843(1472)
KI2	19.674	484.266	37±5	26.3±1.7	8820(1461)
SU7	12.469	224.978	28±9	33.8±2	12584(1730)
SU8	13.135	183.311	34±8	38±4	1103(561)
RAD1	16.834	206.388	36±5	22.7±0.8	30562(2764)
VR1	20.380	170.247	55±6	30.1±0.9	

Statistical analysis

- From linear correlation analysis between all measured parameters and radon in soil concentrations the Pearson correlation coefficient was calculated with two-tailed statistical significance of 0.05. It was only found statistically significant correlation between ^{226}Ra content and radon in soil concentrations



For regression analysis two groups of predictors must be considered because soil types and surface and volume weighted mean particle size are significant correlated which strongly affect regression model. The first model with ^{226}Ra, ^{238}U, fine sand, medium sand, silit and clay as predictors can be explained by 63.7% (adjusted R square =0.637, $p<0.05$).

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate	Change Statistics				
					R Square Change	F Change	df1	df2	Sig. F Change
1	.905[a]	.818	.637	7833.932	.818	4.507	6	6	.045

a. Predictors: (Constant), finesand, mediumsand, zemlja uranijum, silit, zemlja radijum, clay

ANOVA[a]

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	1659555156	6	276592526.1	4.507	.045[b]
	Residual	368222956.3	6	61370492.72		
	Total	2027778113	12			

a. Dependent Variable: zemlja radon

b. Predictors: (Constant), finesand, mediumsand, zemlja uranijum, silit, zemlja radijum, clay

The second one with ^{226}Ra, ^{238}U and surface weighted mean of soil particle sizes predicts radon in soil concentrations with probability of 61.4% (adjusted R square =0.614, $p<0.05$).

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate	Change Statistics				
					R Square Change	F Change	df1	df2	Sig. F Change
1	.843[a]	.711	.614	8073.174	.711	7.371	3	9	.009

a. Predictors: (Constant), surfaceweigth, zemlja uranijum, zemlja radijum

ANOVA[a]

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	1441192836	3	480397611.9	7.371	.009[b]
	Residual	586585277.0	9	65176141.89		
	Total	2027778113	12			

a. Dependent Variable: zemlja radon

b. Predictors: (Constant), surfaceweigth, zemlja uranijum, zemlja radijum

Coefficients[a]

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.	95.0% Confidence Interval for B		Correlations		
		B	Std. Error	Beta			Lower Bound	Upper Bound	Zero-order	Partial	Part
1	(Constant)	-44350.988	27136.583		-1.634	.137	-105738.204	17036.228			
	zemlja uranijum	-926.396	281.344	-.684	-3.293	.009	-1562.841	-289.952	-.206	-.739	-.590
	zemlja radijum	2460.932	594.484	1.134	4.140	.003	1116.115	3805.748	.581	.810	.742
	surfaceweigth	945.406	733.307	.318	1.289	.229	-713.450	2604.262	-.279	.395	.231

a. Dependent Variable: zemlja radon

Instead of conclusions

❑Obtained results of RAD7 intercomparison measurement showed good agreement confirming suitability of this radon active monitor for repeatable and sustainable detection of radon in soil gas.

❑Correlation of radon in soil gas concentrations and finest soil component (clay) that correspond to permeability of soil shows that other parameters, such as porosity, influence the emanation processes for grain aggregates. This fact becomes relevant for measurements in situ, where grains are present in aggregates [11].

❑It was found statistically significant correlation between ^{226}Ra content in soil and radon in soil gas concentrations. As a result of standard regression analysis between the measured values, model that includes soil grain composition, radium and uranium content as predictors with 63.7% affects the radon concentration in soil.

Thank you for your attention!

Hvala na pažnji!