An overview of time integrated measurements of indoor radon and thoron concentrations in Republic of Macedonia

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Introduction

- Radon, thoron and their decay products contribute more than half to the effective dose which the general population receives from natural sources.
- Both gasses are subject on large temporal and spatial variability;

Indoor radon

- The main source of indoor radon is ^{226}Ra contained in building materials as well soil;
- Transport pathways are the porous environment of the soil and the building materials, as well as cracks, faults, etc.
- The radon transport mechanisms: diffusion and advection cased by the concentration and pressure gradients , respectively.

Indoor radon

- The factors affecting the indoor radon concentration can be assigned into three groups:

1. Radon potential –characterize the building sub-surface (in terms of radon generation in soil and condition for transport until its exaltation of the surface
2. Building characteristics
3. Building user life stile and habits

- Additionally, all three factors are affected by the meteorological conditions, resulting with high temporal variability

Introduction

- The main source of indoor thoron concentration (Tn) is ^{232}Th contained in building material.
- As a consequence of Tn short half life (55.6 seconds):
- Tn cannot migrate over long distances,
- Indoor spatial concentration gradient

Rn and Tn surveys in Republic of Macedonia

- During the last decade, considerable attention has been payed to the indoor radon and thoron surveys in the Republic of Macedonia

Radon survey performed (year):	No of observation	Period of exposure (months)	Annual radon concentration
2008/2009	437	3 x 4 seasons	Arithmetic mean of all seasons
2012	73	3 x winter season	Seasonally corrected
2013/2014	76	12	measured
2013	43	3 x winter season	seasonally corrected

Thoron survey performed (year):	No of observation	Period of exposure (months)	Annual thoron concentration
2008/2009	300	3 x 4 seasons	Arithmetic mean of all seasons/seasonally corrected
2012	30	3 x winter season	seasonally corrected

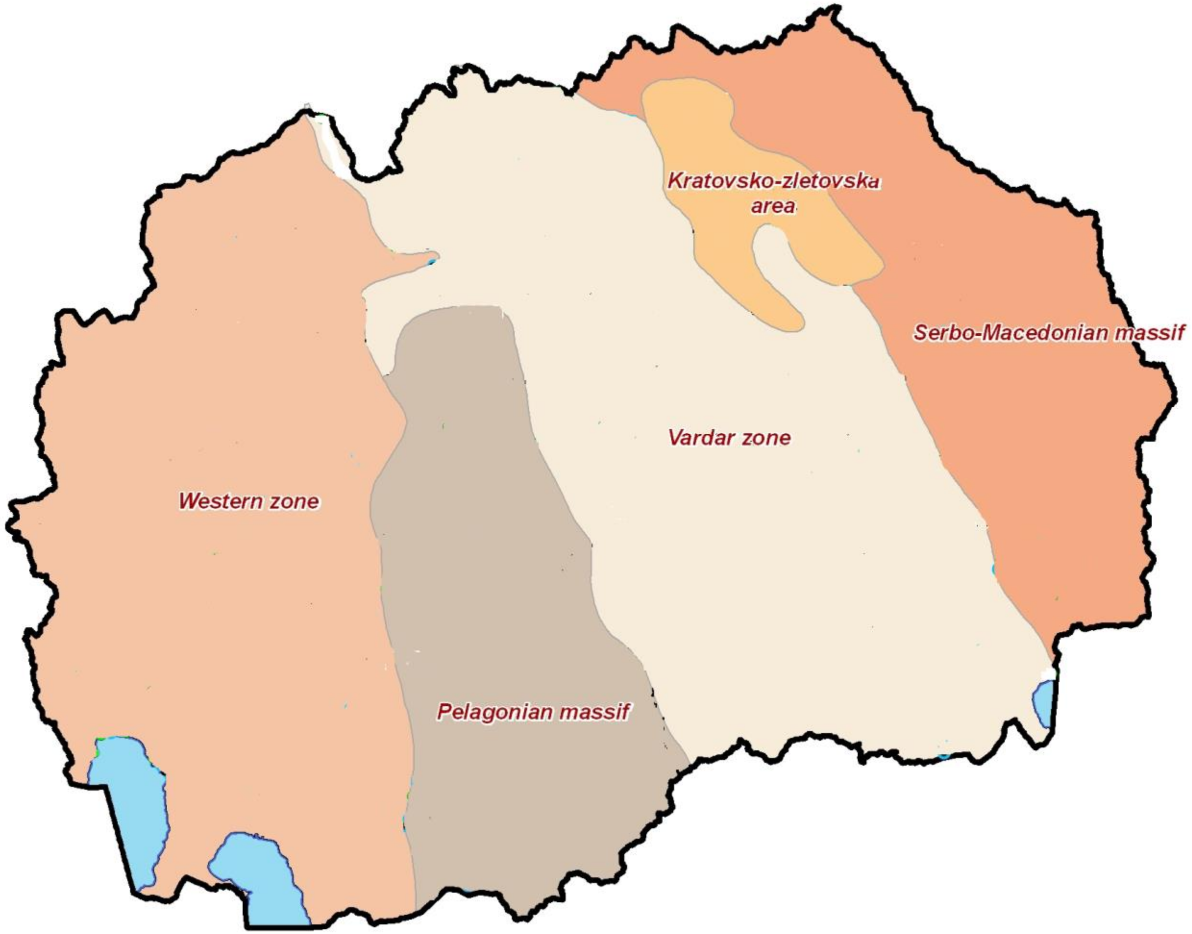
Rn and Tn surveys in Republic of Macedonia

- All measurements were done using the nuclear track detectors.
- The detectors were set in the rooms with the highest occupancy time at a distance greater than 0.5 m from each wall and at a minimum of 20 cm from any other object.

- Republic of Macedonia is situated on Balkan Peninsula and covered 25 713 km^2; Population: 2 022 547
- Great diversity though mountains occupy nearly 80% and basins 20% of the country;

Geotectonical Zones in Republic of Macedonia

- According to the geotectonic, the territory is divided in four zones and an area:



Descriptive statistic of 40K, 226Ra and 232Th specific activities, measured in 213 soil samples by gamma spectrometry

A(Bq/kg)	No.	Min.	Max.	Med	AM	SD	CV(%)	GM	GSD
40K | KZA	8	378	783	665	652	122	18%	639	1.25
40K | PE	36	193	959	645	644	159	24%	622	1.34
40K | SMM	25	348	1390	684	664	239	35%	627	1.41
40K | VZ	90	80	1089	485	496	170	34%	463	1.51
40K | WMM	54	186	974	644	648	174	27%	621	1.37
226Ra | KZA	8	36	101	60	61	19	30%	**58**	**1.36**
226Ra | PE	36	36	123	52	57	20	34%	**55**	**1.35**
226Ra | SMM	25	23	99	41	42	17	40%	40	1.43
226Ra | VZ	90	9	87	30	31	12	39%	29	1.47
226Ra | WMM	54	18	86	39	41	13	32%	39	1.38
232Th | KZA	8	40	67	54	53	9	16%	**52**	**1.18**
232Th | PE	36	35	145	50	59	24	39%	**56**	**1.40**
232Th | SMM	25	23	83	40	43	18	41%	40	1.47
232Th | VZ	90	7	79	31	32	12	37%	30	1.52
232Th | WMM	54	17	69	41	42	12	29%	40	1.36

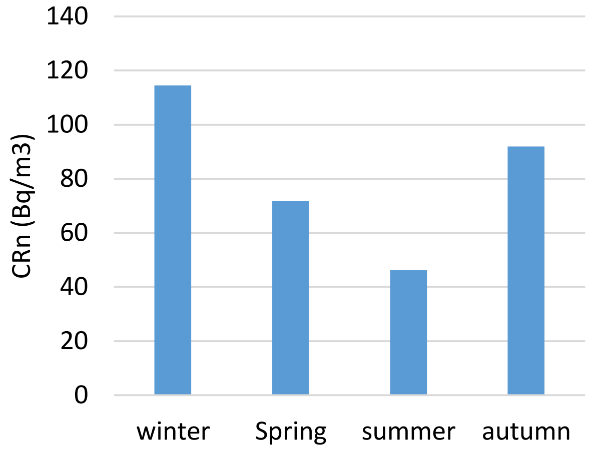
Variations of indoor radon concentrations in Republic of Macedonia

- Indoor radon seasonal variations
- Annual indoor radon data characterization
- Factor affecting spatial variations
- Spatial variability of building factors (6 building factors considered in analysis)

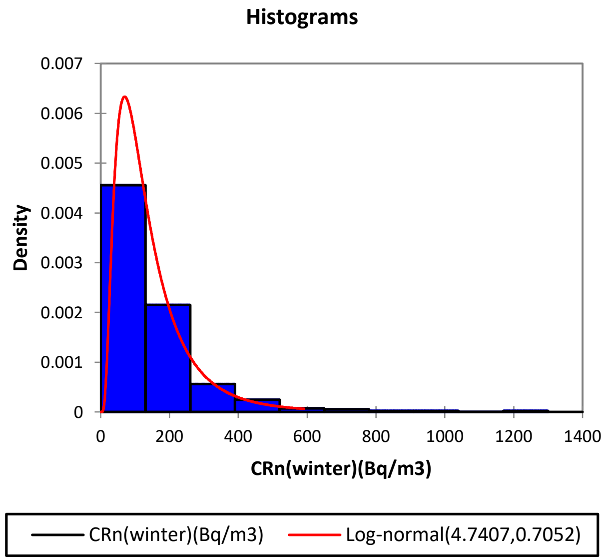
Rn seasonal variation

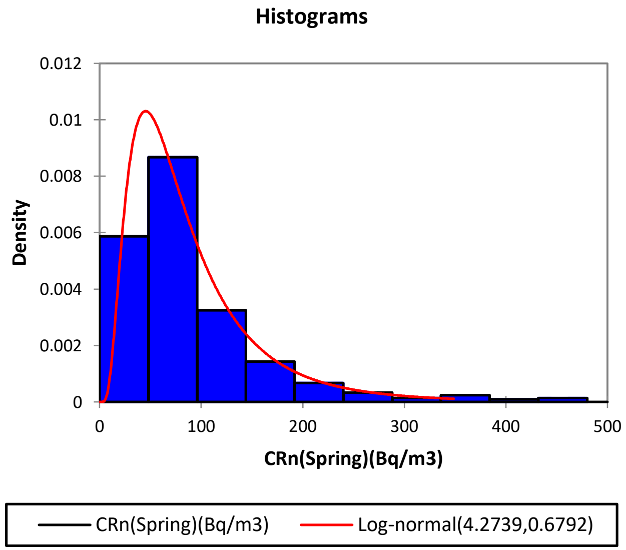
Descriptive statistic of indoor radon measurements in different season (2008/2009)

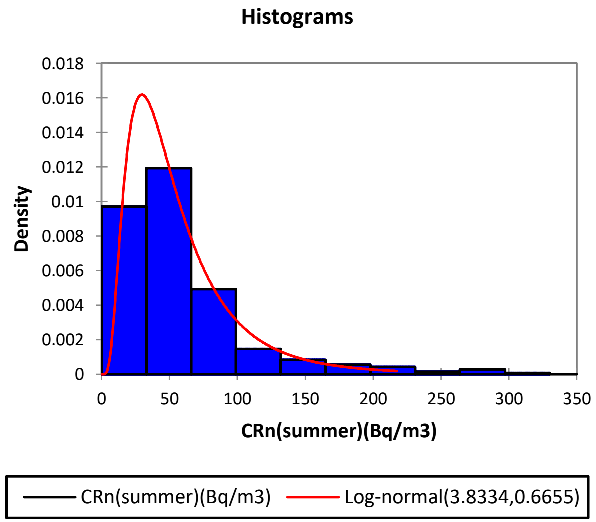
Sample	N	Minimum	Maximum	Median	AM	SD	CV(%)	GM	GSD
CRn(winter)(Bq/m3)	437	17	1276	106	150	136	91%	115	2.02
CRn(Spring)(Bq/m3)	437	9	478	70	91	73	80%	72	1.97
CRn(summer)(Bq/m3)	437	9	323	44	59	48	82%	46	1.95
CRn(autumn)(Bq/m3)	437	7	935	93	119	104	87%	92	2.02

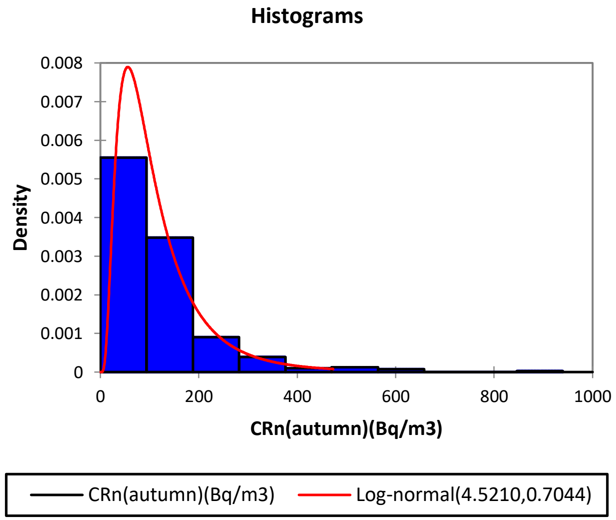


GM of indoor radon concentration in different season







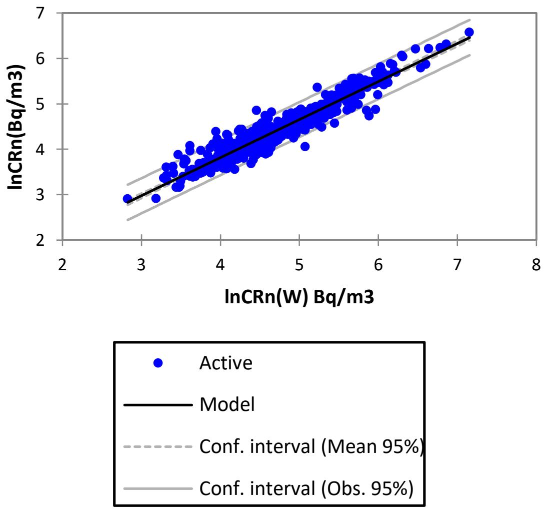


Seasonal Rn variation model

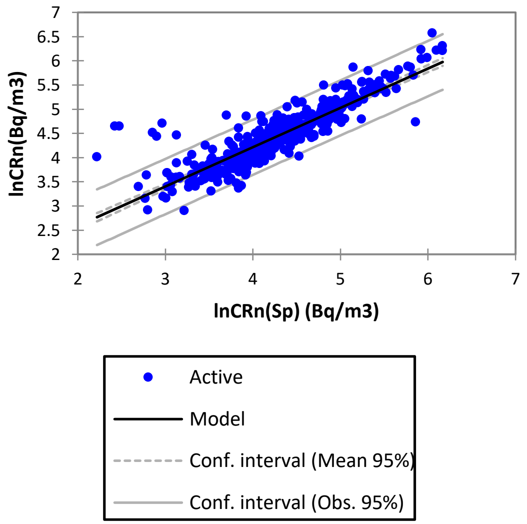
Linear regression model (y=Ax+B) parameters:

y	X	A	B
ln CRn (annual)	lnCRn, winter	0.84±0.01	0.46±0.06
ln CRn (annual)	lnCRn, spring	0.81±0.02	0.96±0.09
ln CRn (annual)	llnCRn, summer	0.71±0.03	1.71±0.11
ln CRn (annual)	lnCRn, autumn	0.80±0.02	0.83±0.08

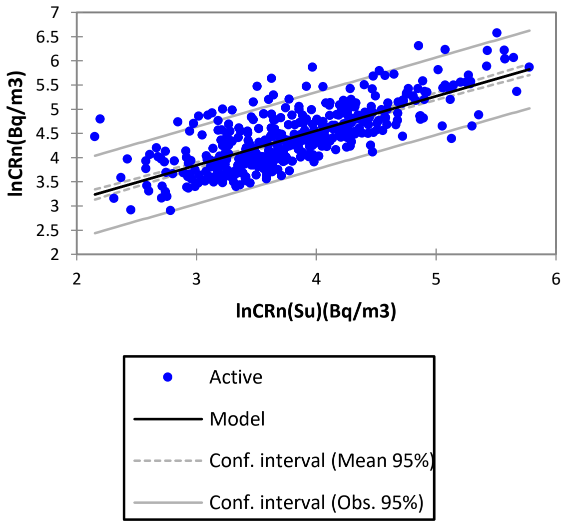
Regression of lnCRn(Bq/m3) by lnCRn(W) Bq/m3 (R2=0.9021)



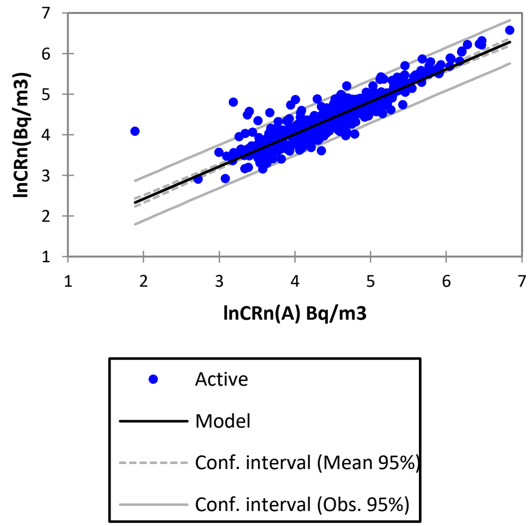
Regression of lnCRn(Bq/m3) by lnCRn(Sp) (Bq/m3) (R2=0.7850)



Regression of lnCRn(Bq/m3) by lnCRn(Su)(Bq/m3) (R2=0.5789)

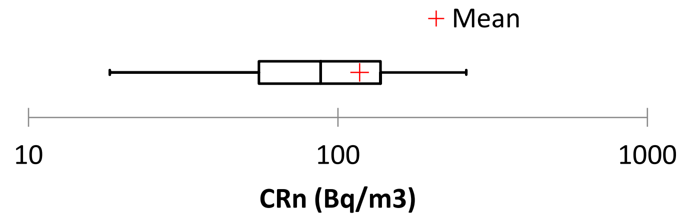


Regression of lnCRn(Bq/m3) by lnCRn(A) Bq/m3 (R2=0.8150)

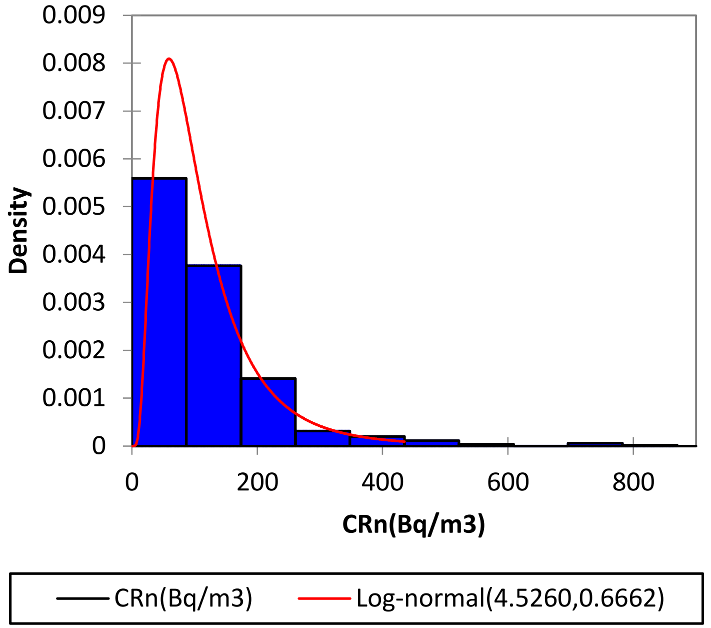


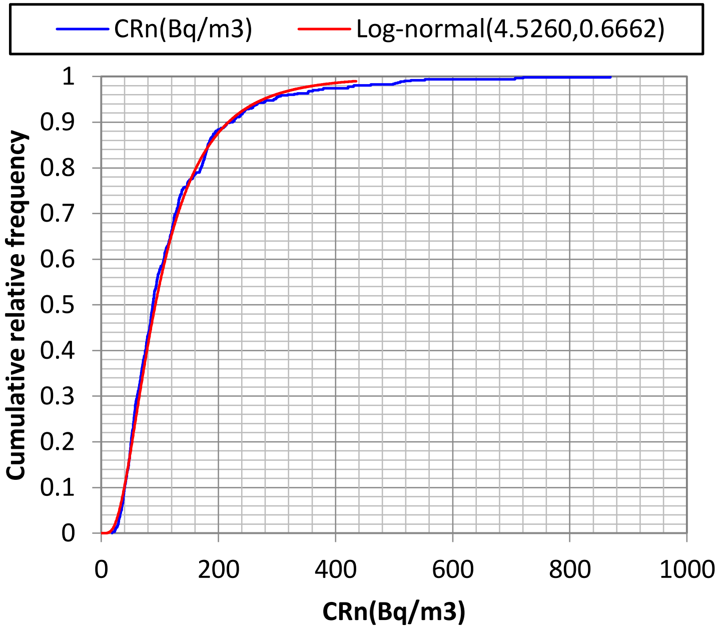
Indoor radon data characterization

Statistic	Rn(Bq/m3)
No. of observations	629
Minimum	18
Maximum	869
Median	88
Mean	118
Standard deviation	99
Variation coefficient	84%
Geometric mean	92
Geometric standard deviation	1.95

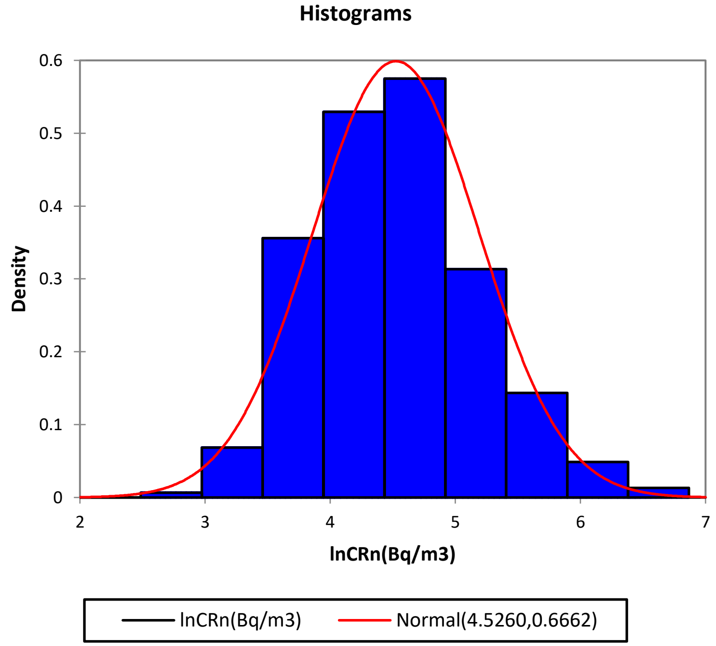


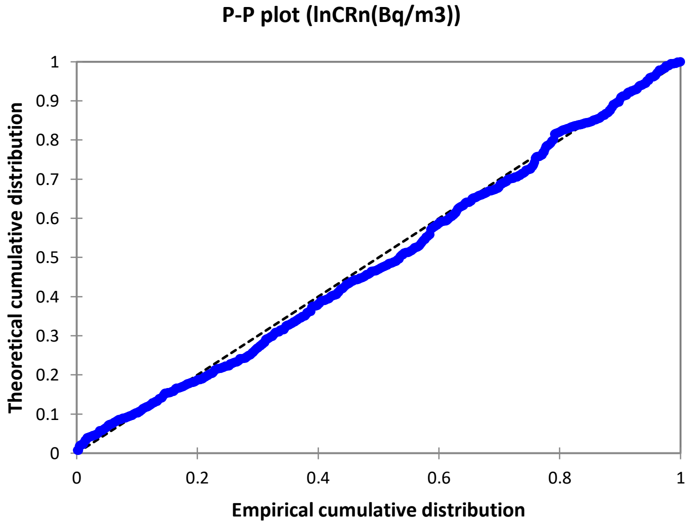
Rn data





ln(Rn) data

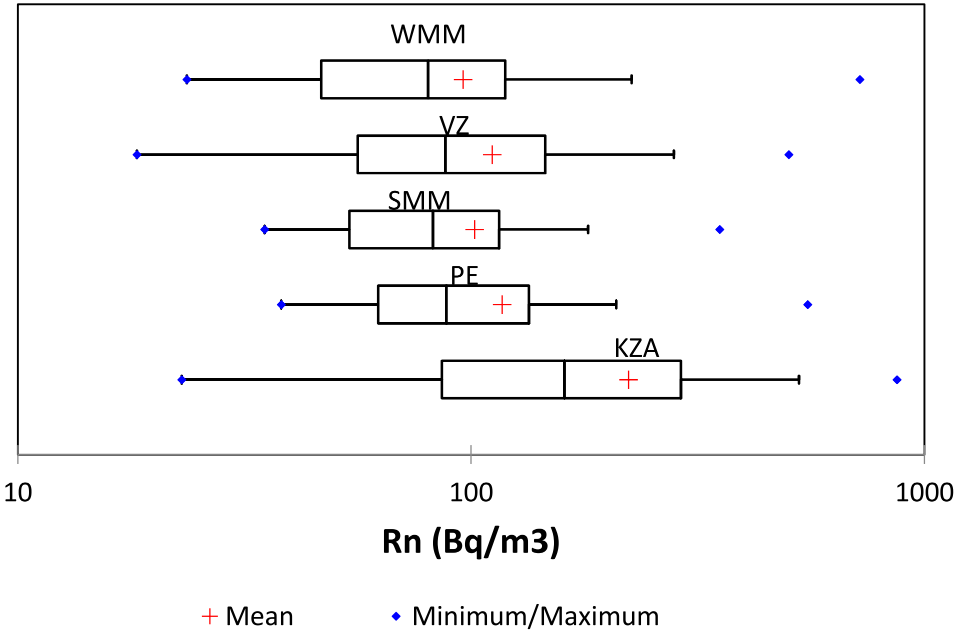




Factor affecting Rn variation

1. Geotectonical Zone

- Non- homogenies (Bartlett test, $p=0.012$)
- Significant variation between Zones (Kruskal-Wallis test, $p< 0.0001$)

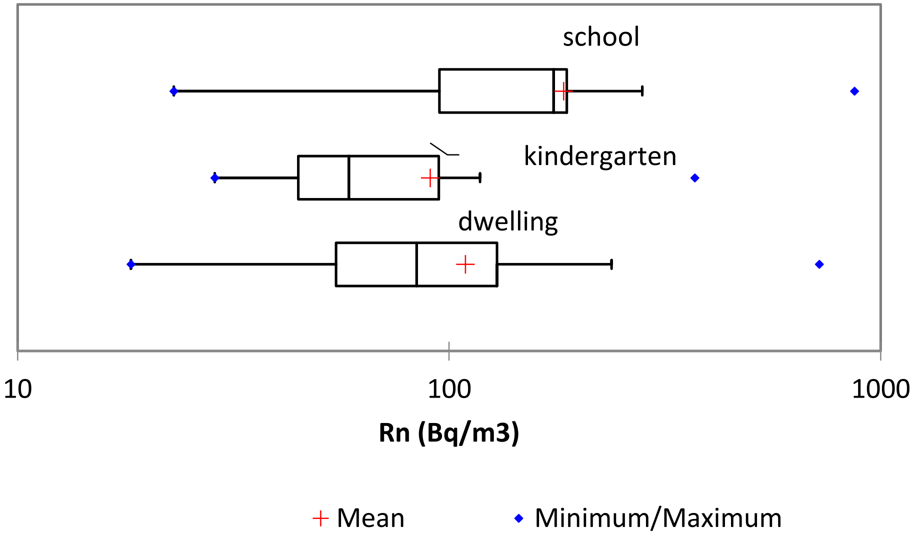


Zone	Frequency	GM	GSD	Groups Mann-Whitney test ($p<0.05$)		
WMM	114	77	1.89	A		
SMM	59	85	1.78	A	B	
VZ	353	91	1.87		B	
PE	49	95	1.81		B	
KZA	54	158	2.36			C

Factor affecting Rn variation

2. Indoor

- Homogenies (Bartlett test, p=0.71)
- Significant variation between different indoor (Kruskal-Wallis test, p< 0.0001)

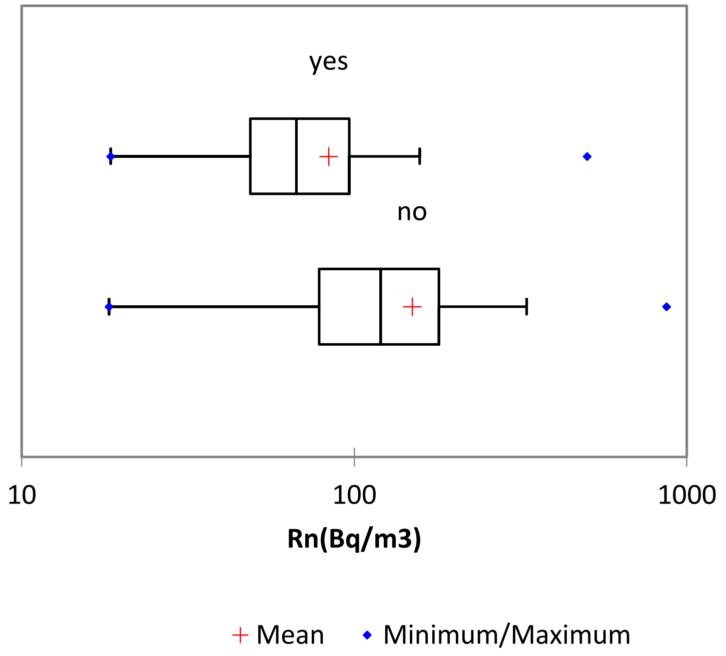


Indoor	Frequency	GM	GSD	Groups Mann-Whitney test (p<0.05)	
kindergarten	14	87	1.89	A	
dwelling	540	68	2.04	A	
school	75	146	1.96		B

Factor affecting Rn variation

4. Presence of basement

- Homogenies (Bartlett test, p=0.09)
- Significant variation (Kruskal-Wallis test, p< 0.0001)

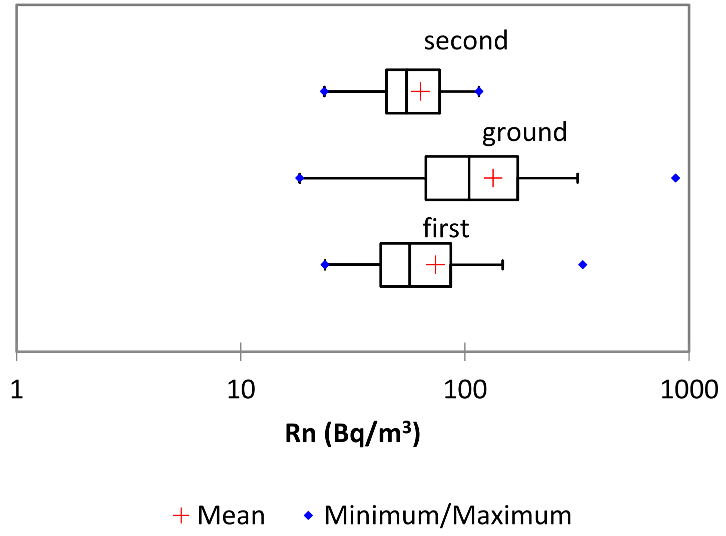


Presence in basement	Frequency	GM	GSD	Groups Mann-Whitney test (p<0.05)	
yes	306	70	1.75	A	
no	323	119	1.92		B

Factor affecting Rn variation

3. Floor

- Non-homogenies (Bartlett test, p=0.0097)
- Significant Rn variation between different floors (Kruskal-Wallis test, p< 0.0001)

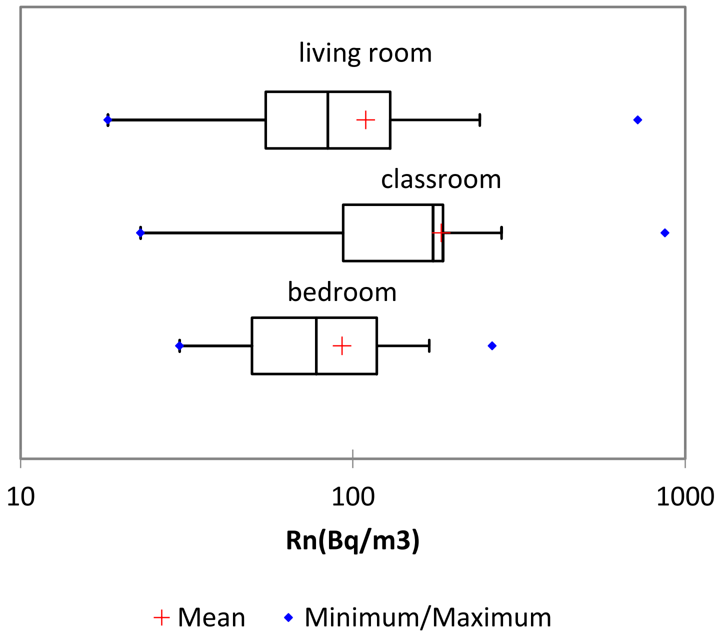


Floor	Frequency	GM	GSD	Groups Mann-Whitney test (p<0.05)	
second	15	57	1.60	A	
first	149	64	1.67	A	
ground	465	106	1.94		B

Factor affecting Rn variation

5. Type of room

- Homogenous data (Bartlett test, $p=0.39$)
- Significant Rn variation between group (Kruskal-Wallis test, $p< 0.0001$)

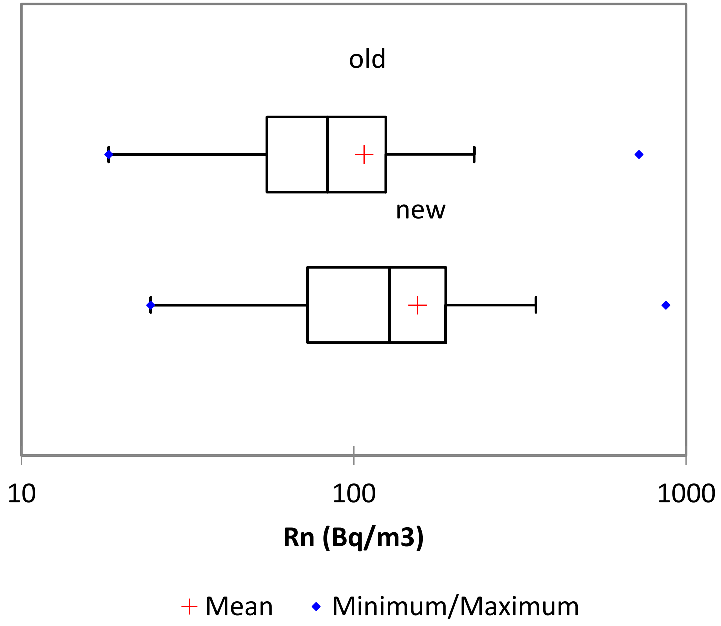


Floor	Frequency	GM	GSD	Groups Mann-Whitney test (p<0.05)	
bedroom	44	79	1.76	A	
living room	508	87	1.91	A	
classroom	77	146	1.98		B

Factor affecting Rn variation

6. Type of windows

- homogenous (Bartlett test, p=0.16)
- Significant variation (Kruskal-Wallis test, p< 0.0001)



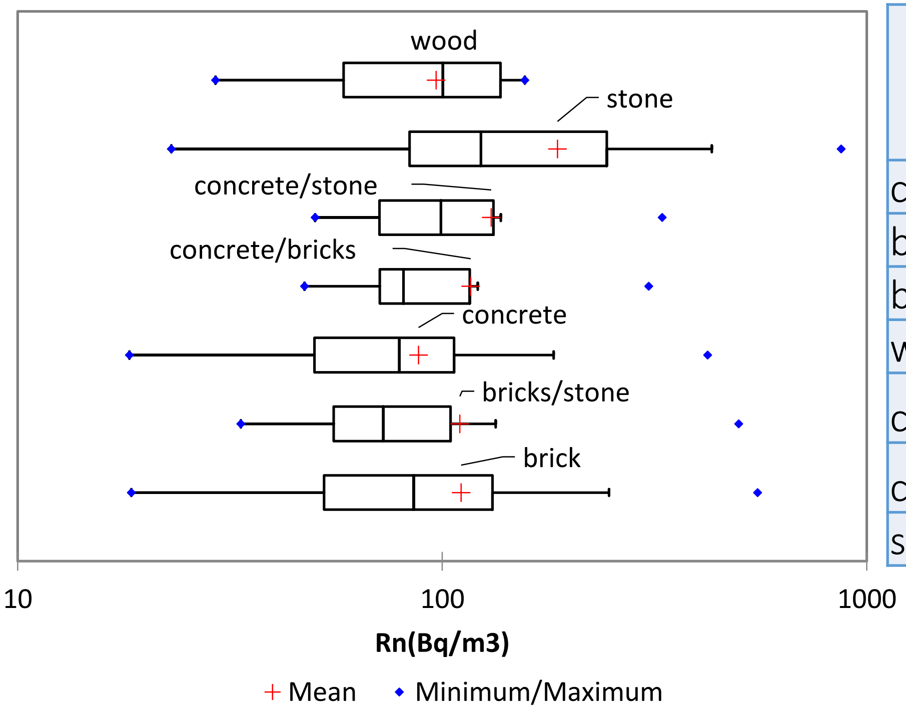
Type of windows	Frequency	GM	GSD	Groups Mann-Whitney test (p<0.05)	
Old	498	86	1.89	A	
New	131	121	2.05		B

Factor affecting Rn variation

7. Building materials

Homogenous data (Bartlett test, p=0.36)

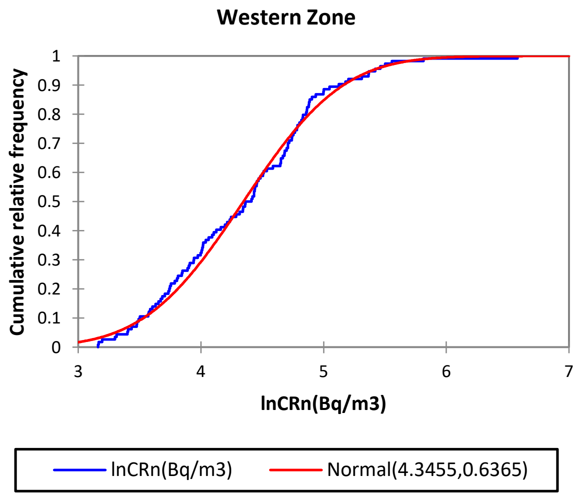
Significant variation between groups (Kruskal-Wallis test, p< 0.0001)



Building material	Frequency	GM	GSD	Groups Mann-Whitney test (p<0.05)	
concrete	104	74	1.78	A	
bricks/stone	45	86	1.92	A	
brick	326	88	1.93	A	
wood	6	82	1.94	A	B
concrete/bricks	10	98	1.80	A	B
concrete/stone	6	107	1.94	A	B
stone	68	137	2.15		B

Spatial variability of building factors influence

Western Zone



lnRn concentrations were normally distributed

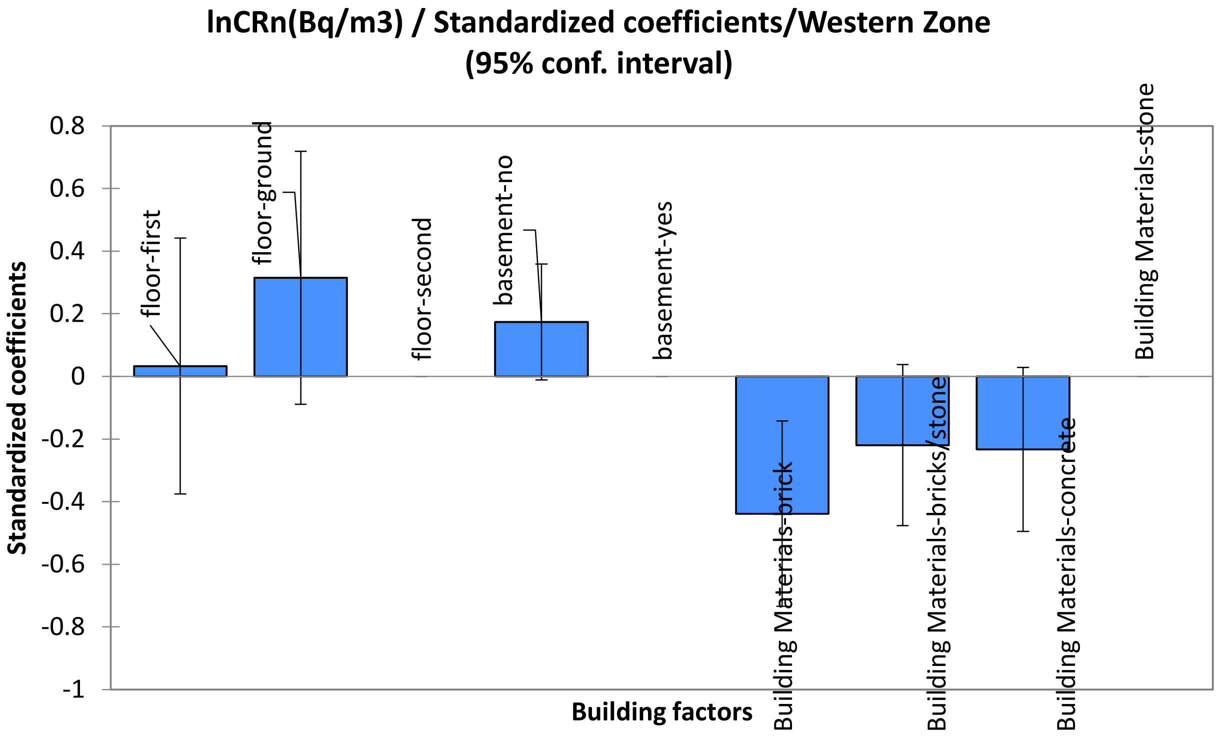
Univariable linear models results:

	Western Zone	p	R^2	RMSE
1	indoor			
2	floor	0.0046	0.1163	0.5652
3	basement	0.0016	0.0851	0.6115
4	room	0.9856		
5	building material	0.0490	0.0634	0.6243
6	windous	0.1133		

Multivariable linear model (R^2=0.21; RMSE=0.58)

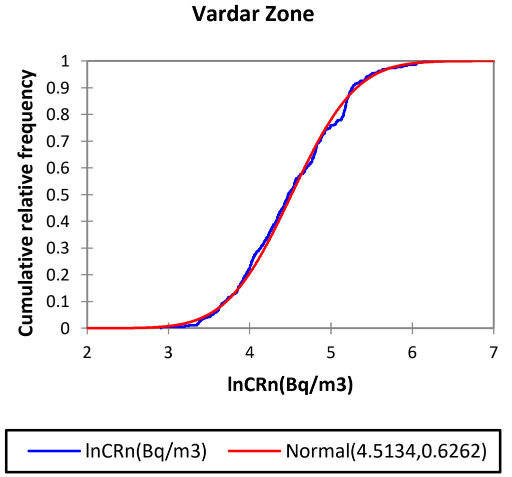
Equation of the multiple regression model:

lnCRn(Bq/m3) = 4.39197+0.04551*floor-first+0.41828*floor-ground+0.22329*basement-no-0.55592*Building Materials-brick-0.35870*Building Materials-bricks/stone-0.37402*Building Materials-concrete



Spatial variability of building factors influence

Vardar Zone



lnRn concentrations were normally distributed

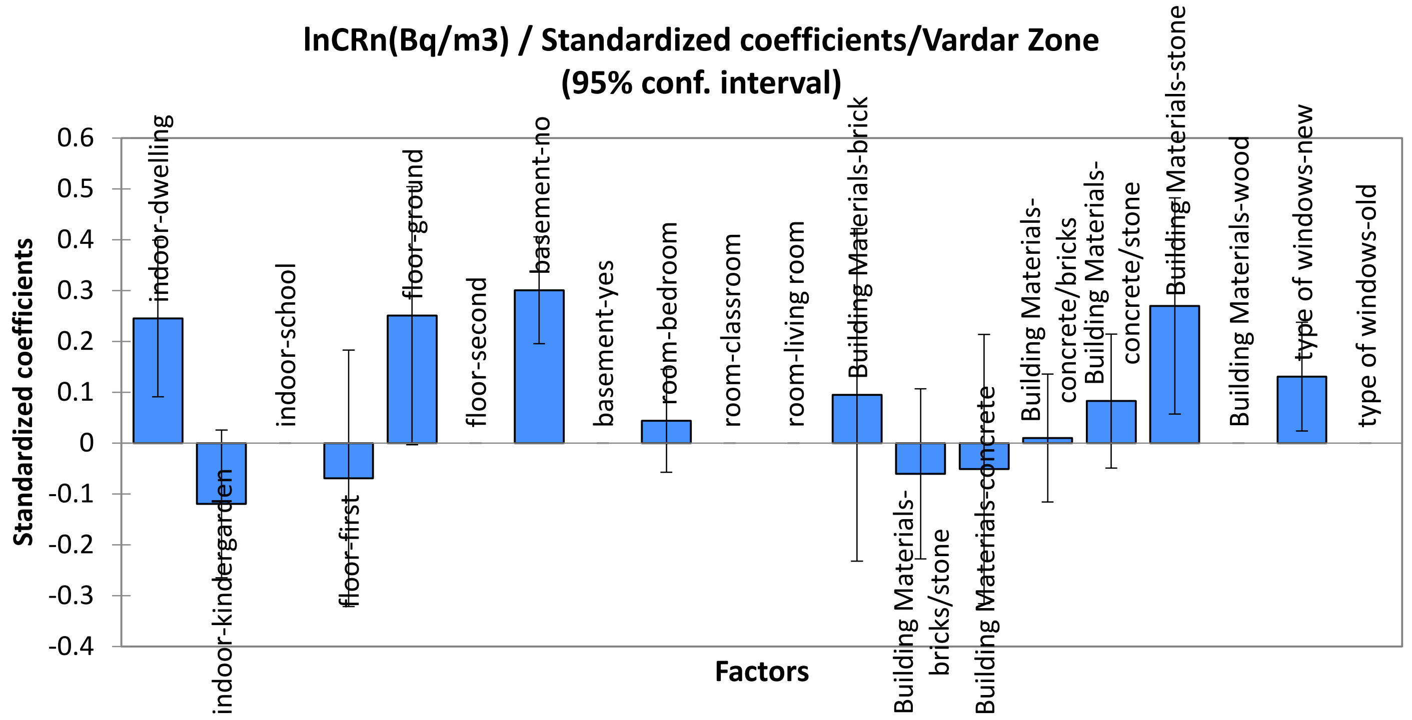
Univariable linear models results:

	Vardar zone	p	R^2	RMSE
1	indoor	< 0.0001	0.11	0.59
2	floor	< 0.0001	0.11	0.59
3	basement	< 0.0001	0.12	0.59
4	room	< 0.0001	0.08	0.60
5	building material	0.0009	0.08	0.62
6	windows	0.0065	0.02	0.62

Multivariable linear model (R^2=0.32; RMSE=0.54)

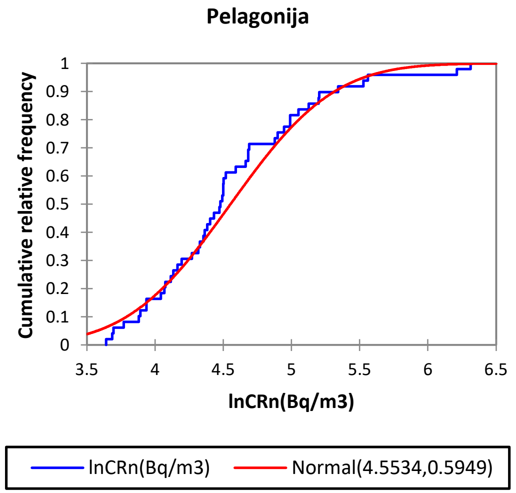
Equation of the multiple regression model:

lnCRn(Bq/m3) = 3.33447+0.58409*indoor-dwelling-0.39647*indoor-kindergarden-0.10662*floor-first+0.36619*floor-ground+0.38119*basement-no+0.10066*room-bedroom+0.12733*Building Materials-brick-0.19183*Building Materials-bricks/stone-0.08971*Building Materials-concrete+0.05245*Building Materials-concrete/bricks+0.40046*Building Materials-concrete/stone+0.62988*Building Materials-stone+0.20736*type of windows-new



Spatial variability of building factors influence

Pelagonija



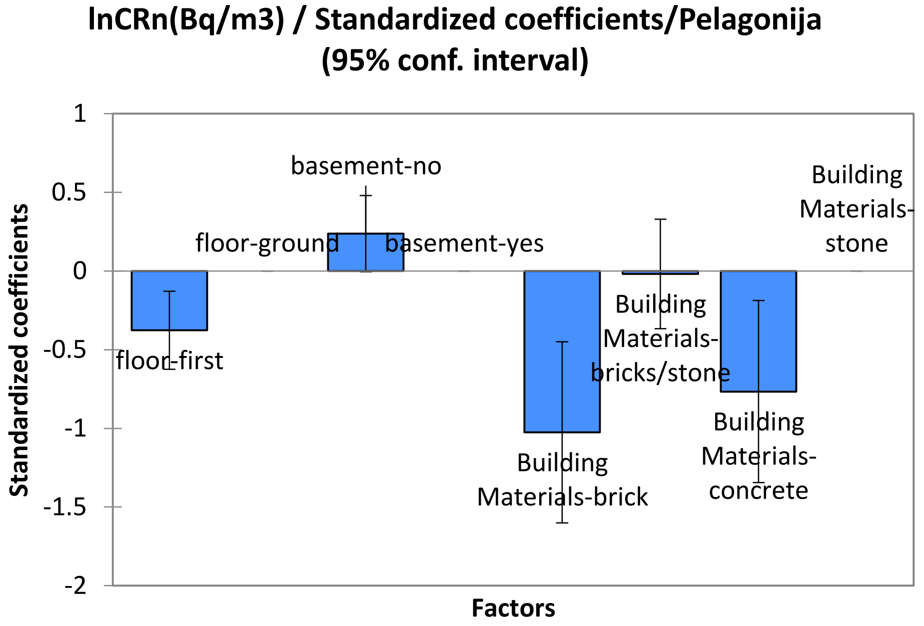
lnRn concentrations were normally distributed

Univariable linear models results:

	Pelagonija	p	R^2	RMSE
1	indoor			
2	floor	0.0165	0.1163	0.5652
3	basement	0.0300	0.0952	0.5719
4	room			
5	building material	0.0050	0.2508	0.5318
6	windous	0.4151		

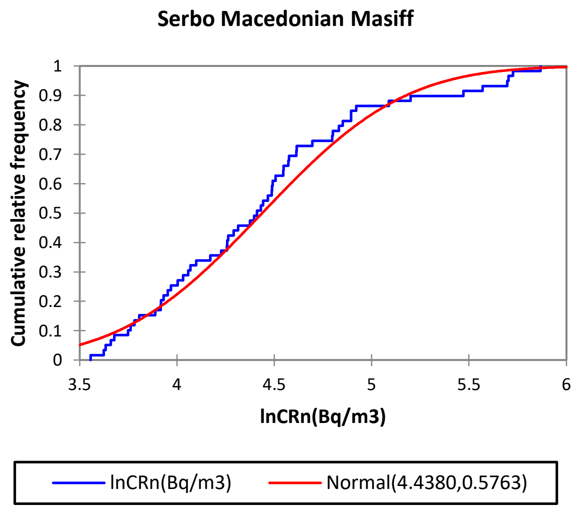
Multivariable linear model (R^2=0.49; RMSE=0.45)

Equation of the model:
lnCRn(Bq/m3) = 5.58556-0.44397*floor-first+0.27989*basement-no-1.22853*Building Materials-brick-0.04550*Building Materials-bricks/stone-0.90219*Building Materials-concrete



Spatial variability of building factors influence

Serbo-Macedonian Massif



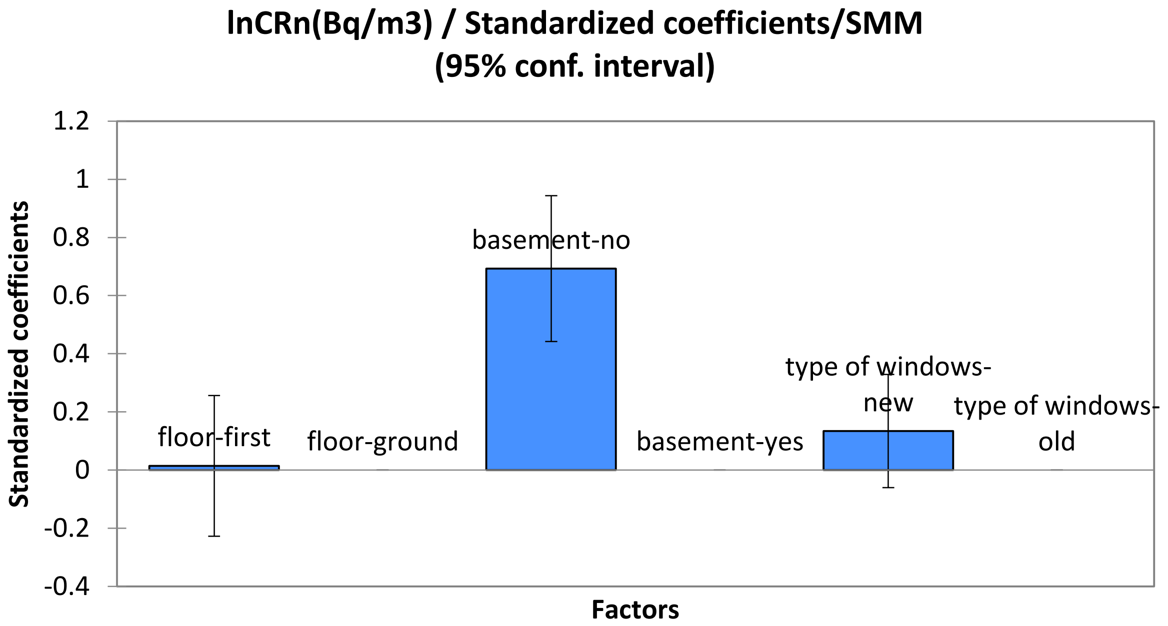
lnRn concentrations were normally distributed

Univariable linear models results:

	Serbo Macedonian Massif	p	R^2	RMSE
1	indoor			
2	floor	0.0001	0.2344	0.5087
3	basement	< 0.0001	0.5392	0.3947
4	room	0.6051		
5	building material	0.2300		
6	windows	0.0021	0.2344	0.5087

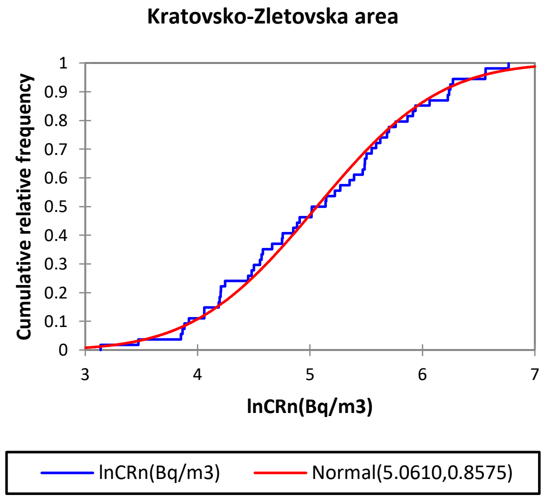
Multivariable linear model (R^2=0.56; RMSE=0.40)

Equation of the model:
lnCRn(Bq/m3) = 4.05379+0.01685*floor-first+0.79762*basement-no+0.25412*type of windows-new



Spatial variability of building factors influence

Kratovsko-Zletovska Area



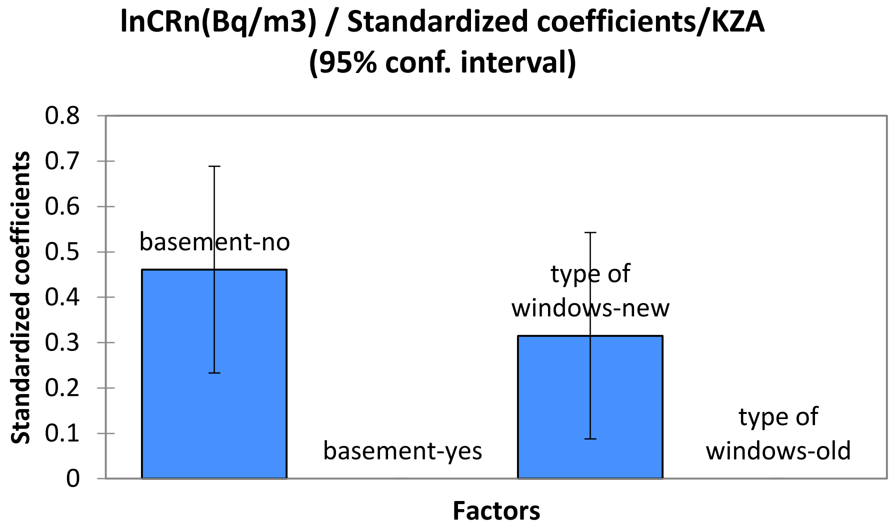
lnRn concentrations were normally distributed

Univariable linear models results:

	Kratovsko-Zletovska area	p	R^2	RMSE
1	indoor	0.9400		
2	floor	0.1800		
3	basement	< 0.0001	0.2821	0.7335
4	room	0.7295		
5	building material	0.6530		
6	windows	0.0017	0.1748	0.7864

Multivariable linear model (R^2=0.38; RMSE=0.69)

Equation of the model:
lnCRn(Bq/m3) = 4.35603+0.81964*basement-no+0.58634*type of windows-new



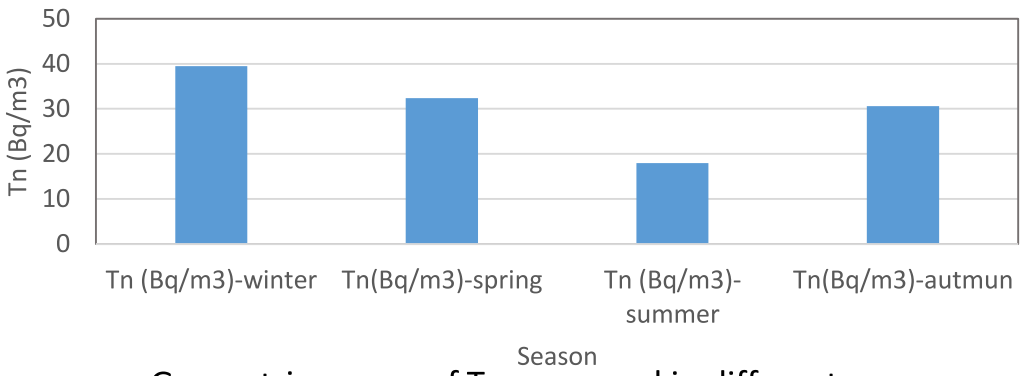
Variations of indoor thoron concentrations in Republic of Macedonia

- Indoor thoron seasonal variation
- Data characterization
- Factors affecting Tn variation

Tn seasonal variability

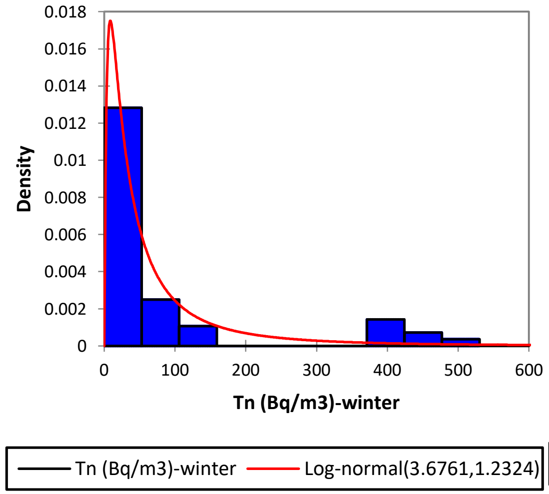
- Difference in Tn between the four seasons was observed in the results.

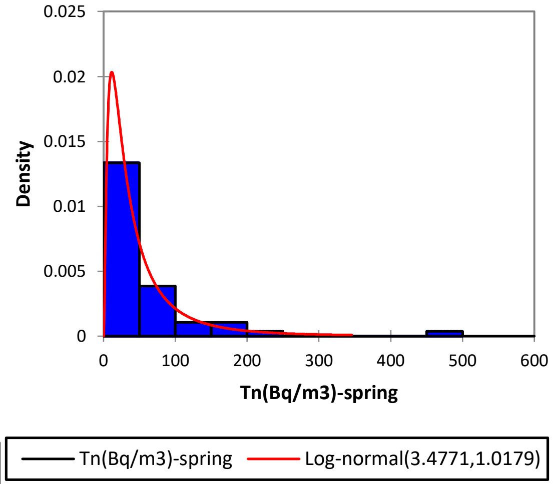
Sample	No. of observations	Maximum	Median	Mean	SD	CV	GM	GSD
Tn (Bq/m3)-winter	53	525	33	90	137	151%	39.4911	3.43
Tn(Bq/m3)-spring	57	495	28	56	77	138%	32.3659	2.77
Tn (Bq/m3)-summer	122	245	19	30	38	125%	17.9315	2.78
Tn(Bq/m3)-autmun	300	395	34	52	64	122%	30.6038	2.93

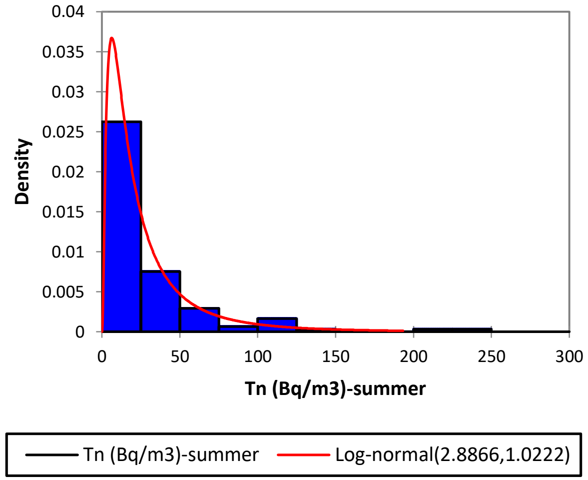


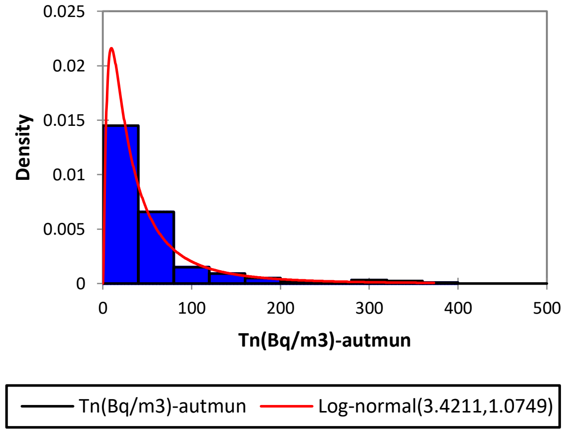
Geometric means of Tn measured in different season.

- The Tn concentrations in all season were fitted with log normal distribution:





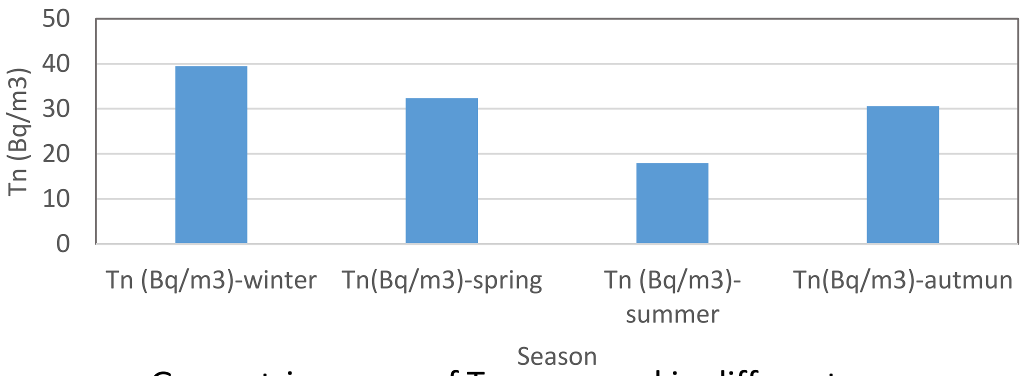




Tn seasonal variability

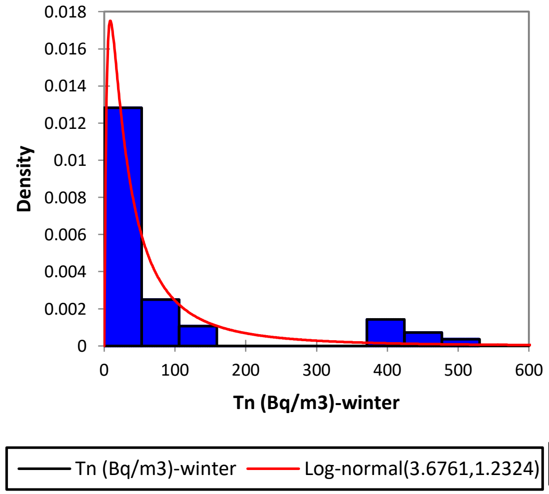
- Difference in Tn between the four seasons was observed in the results.

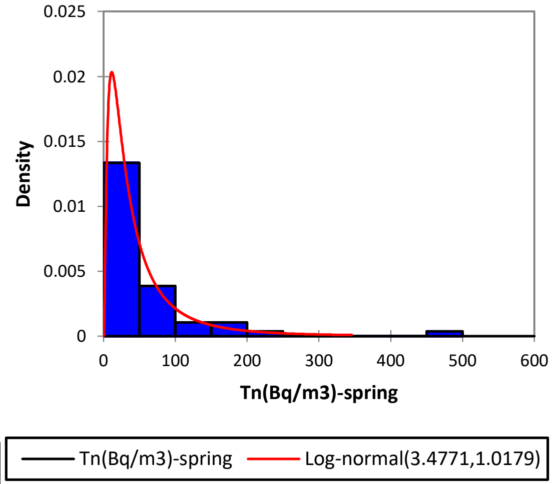
Sample	No. of observations	Maximum	Median	Mean	SD	CV	GM	GSD
Tn (Bq/m3)-winter	53	525	33	90	137	151%	39.4911	3.43
Tn(Bq/m3)-spring	57	495	28	56	77	138%	32.3659	2.77
Tn (Bq/m3)-summer	122	245	19	30	38	125%	17.9315	2.78
Tn(Bq/m3)-autmun	300	395	34	52	64	122%	30.6038	2.93

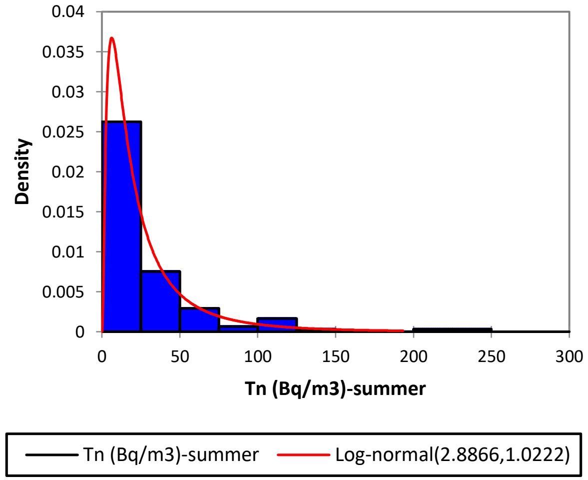


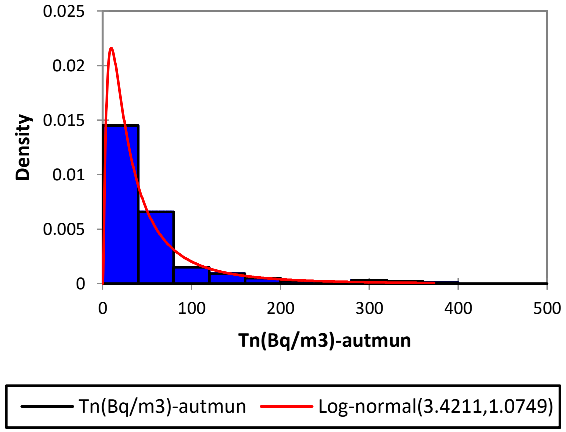
Geometric means of Tn measured in different season.

- The Tn concentrations in all season were fitted with log normal distribution:









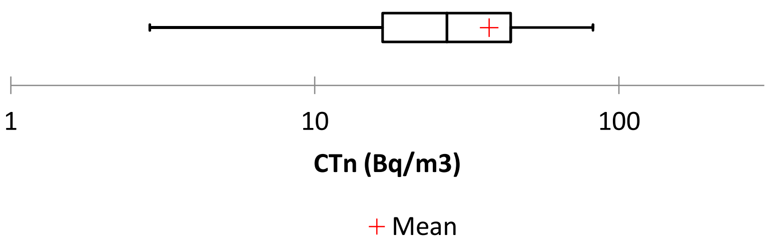
Tn seasonal correction

- The parametric linear regression analysis was applied to determined relationships between the log transformed Tn measured in autumn to Tn in winter, spring, as well as Tn measured in autumn to measured in summer for the houses with and without basement separately.
- Regression model: y=Ax+B.

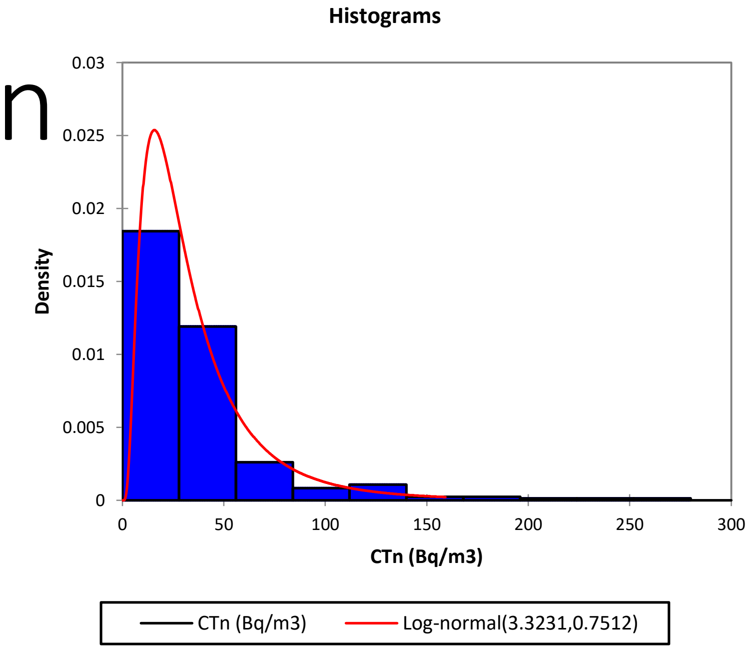
y	X	A	B	R^2
logTn, winter	logTn, autumn	0.648±0.109	0.542±0.186	0.410
logTn, spring	logTn, autumn	0.590±0.074	0.626±0.118	0.534
log Tn, summer; house with basement	logTn, autumn	0.472±0.093	0.640±0.137	0.269
log Tn summer; house without basement	logTn, autumn	0.468±0.121	0.504±0.185	0.488

Indoor annual Tn data characterization

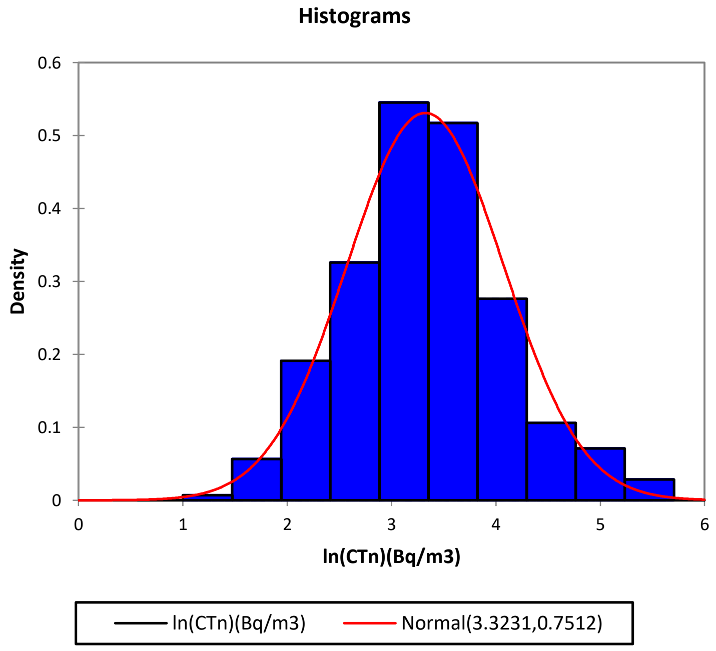
Statistic	CTn (Bq/m3)
No. of observations	300
Maximum	272
Median	27
Mean	37
Standard deviation	36
Variation coefficient	96%
Geometric mean	28
Geometric standard deviation	2.12

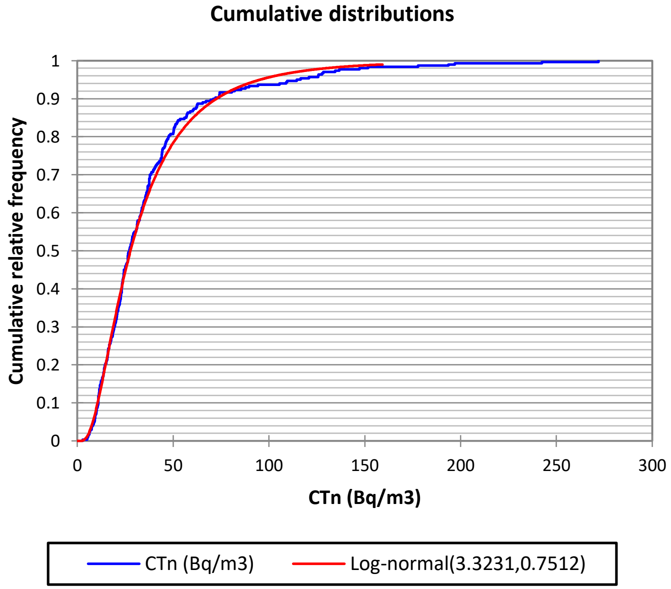


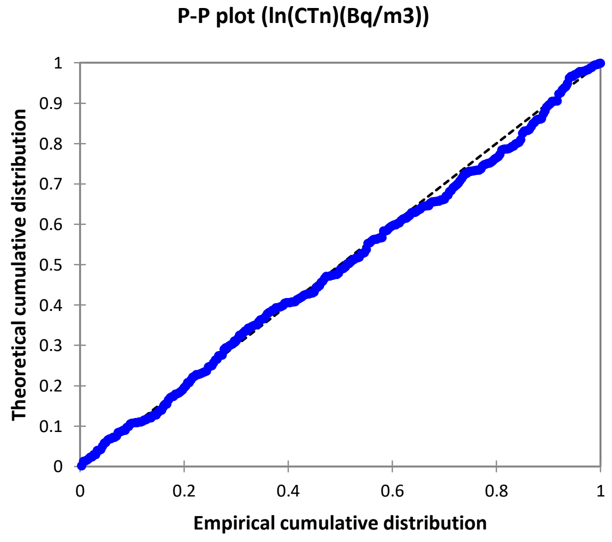
Tn data



ln(Tn) data



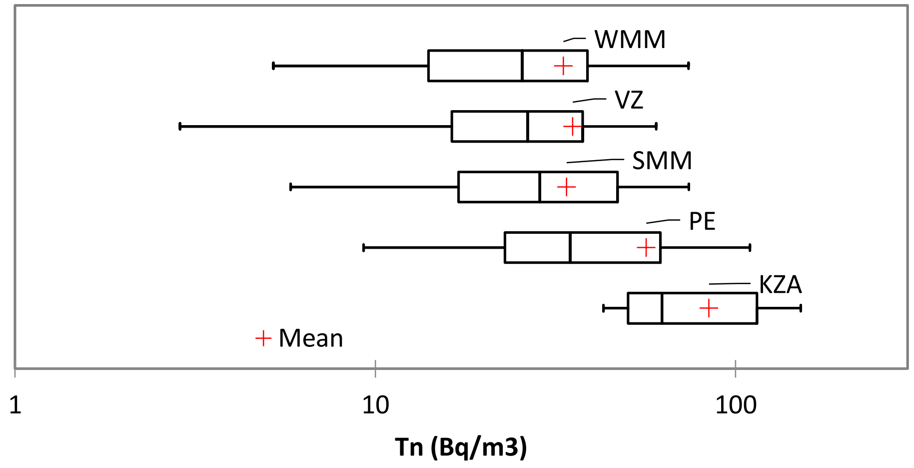




Factor affecting Tn variation

1. Geotectonical Zone

- Homogenies (p=0.75)
- Significant variation between Zones (ANOVA, p< 0.0013); R^2=0,06

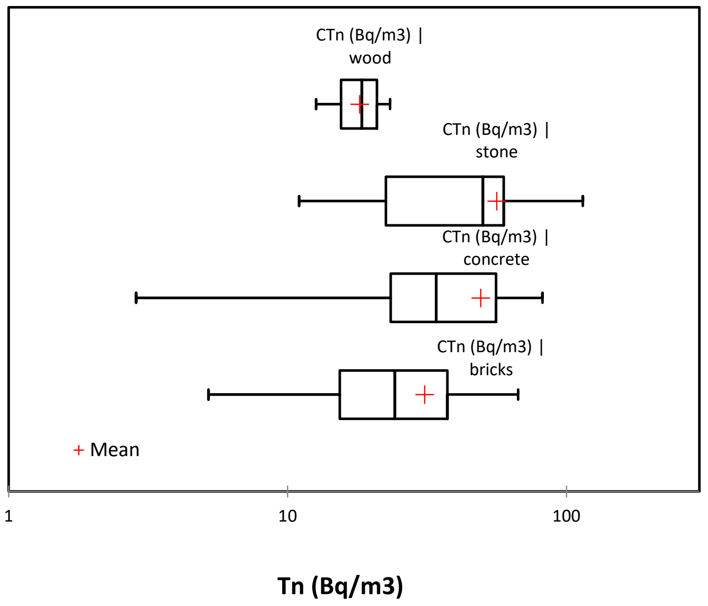


Geotectonical Zone	No.	GM	GSD	Groups (Fisher test)	
KZA	5	75	1.72	A	
PE	29	40	2.30	A	
SMM	43	27	2.06		B
VZ	142	27	2.03		B
WMM	81	25	2.12		B

Factor affecting Tn variation

2. Building materials

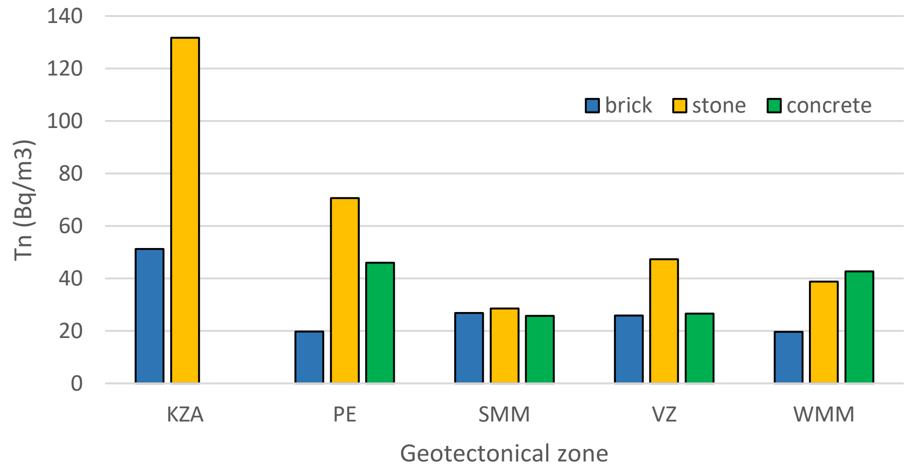
- Homogenies (p=0.10)
- Significant variation between building materials (ANOVA, $p < 0.0001$), $R^2=0.09$

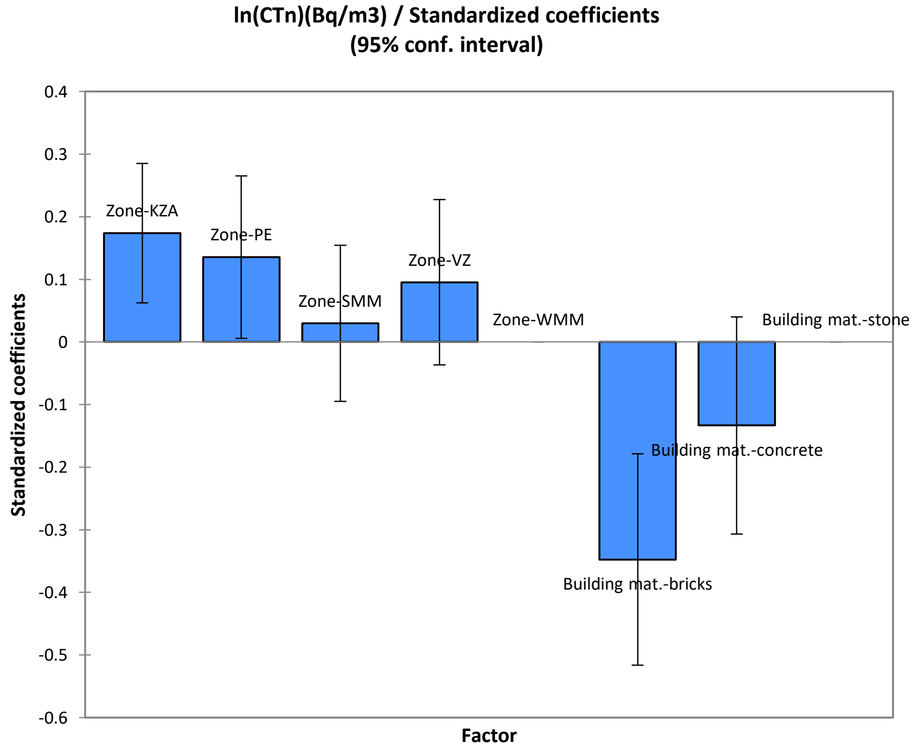


Building materials	No	GM	GSD	Groups (Fisher test)	
bricks	202	24	1.97	B	
concrete	63	35	2.33		A
stone	32	43	2.11		A
wood	3	18	1.36		

Factor affecting Tn variation

- Significant variation between Zones (ANOVA, $p < 0.0001$), $R^2=0.12$





Equation of the multiple regression model:

ln(CTn)(Bq/m3) = 3.63279+1.01791*Zone-KZA+0.34330*Zone-PE+0.06404*Zone-SMM+0.14380*Zone-VZ-0.56001*Building mat.-bricks-0.24482*Building mat.-concrete

Summary

- Seasonal variability of Rn (highest concentrations in the winter and lowest in summer) is confirmed;
- The models for assessing of annual Rn from 3 months measurement are developed. The model with the best performance refers to the linear relationship between the Rn measurements in winter and annual Rn (R^2=90%).
- Differences in the Rn in various geotectonic zones of the country is significant;

Summary

- In general, building factors: floor, basement, type of windows, the indoor type, type of room, building material significantly affect Rn variation. On the other hand, their impact is associated with geotectonic zones .
- Practically no all factors as well each factor separately has equal contribution to Rn variations in individual geotectonic units. Our results showed that Rn variations which originating by the building characteristics are in the range: from 21% in Western zone to 56% in the Serbo-Macedonian Massif.

Summary

- Just like a radon, the Tn seasonal and spatial variations is significant.
- Tn seasonal variation models are with lower coefficients of determination compared to Rn models. For Tn the best model is, with: $R^2 = 0.53$;
- Grouping Tn according to geotectectonic zones and building materials, we assumed that the used local materials for the buildings construction are the main source of Th variations. But these two factors explain only 12% of the Tn variability.