



# The path from geology to indoor radon

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**Abstract** It is generally accepted that radon emission is strongly influenced by the geological characteristics of the bedrock. However, transport in-soil and entry paths indoors are defined by other factors such as permeability, building and architectural features, ventilation, occupation patterns, etc. The purpose of this paper is to analyze the contribution of each parameter, from natural to man-made, on the radon accumulation indoors and to assess potential patterns, based on 100 case studies in Romania. The study

pointed out that the geological foundation can provide a reasonable explanation for the majority of the values recorded in both soil and indoor air. Results also showed that older houses, built with earth-based materials, are highly permeable to soil radon. Energy-efficient houses, on the other hand, have a tendency to disregard the radon potential of the geological foundation, causing a higher predisposition to radon accumulation indoors and decreasing the general indoor air quality.

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## Introduction

Radon is considered a carcinogenic gas, attributed between 9 and 15% of the 14,000 annual cases of lung cancer in Europe (Darby et al. 2005; Krewski et al. 2005). Worldwide, it is considered to be the second leading cause of lung cancer after smoking. Prolonged exposure to levels exceeding 100 Bq/m<sup>3</sup> is treated as a serious environmental problem (WHO 2009; UNSCEAR 2000). The European Union has stipulated in the C.E. 2013/59/Euratom the obligation of Member States to monitor and report levels of radioactivity from natural and anthropogenic sources. Romania has since adopted Law 526/2018, establishing a National Action Plan to tackle the radon issue.

Radon ( $\text{Rn}^{222}$ ) is a decay product directly descending from radium ( $\text{Ra}^{226}$ ) in the natural uranium decay series. The local geology is the main controlling factor on the sources of radon generation in the atmosphere, and consequently, indoors too (Sachs et al. 1982; Kemski et al. 2001, 2005, 2009; Ciotoli et al. 2017), since uranium is widely distributed in rocks and soils throughout the earth's crust. It is well known that any rock may have a higher or lower content of radioactive elements, depending on its genesis (Sachs et al. 1982; Gundersen et al. 1992; Appleton 2007; Drolet et al. 2013). Magmatic and metamorphic rocks, for example, are known to have a higher content of radioactive elements (U/Ra) than most sedimentary rocks (Stoici and Tătaru 1988; Cosma and Jurcuț 1996). However, some sedimentary rocks, mainly clastic sedimentary rocks, consisting of fragments (clasts) of preexisting magmatic or metamorphic rocks, loosened from weathering process, then transported and accumulated to some sedimentary basins, are also known to have a significant radioactive potential (Drolet et al. 2013). The causal relationship between the amount of radioactive elements in a certain type of rock and radon emanations is not always easy to plot, even if the latter is dependent on uranium concentrations and the nature of the parental mineral. Certain factors such as rock granulometry, permeability, fractures, cracks, rock weathering and decay characteristics may play an important role in the generation and migration of radon to the subsurface (Ball et al. 1991; Nazaroff 1992; Drolet et al. 2013). Besides the geological bedding of a building site, the transfer of radon indoors is dependent on environmental factors such as ventilation, occupation patterns or building and architectural features (Sachs et al. 1982; Gundersen et al. 1992; Kemski et al. 2005; Bossew et al. 2008; Appleton and Miles 2010).

Recent studies have shown that although the variation of the radon levels indoors depends primarily on the physico-chemical characteristics of the substrate (type of rock and permeability), the parameters that influence the inlet and transport paths can play a decisive role in the accumulation of radon (Demoury et al. 2013; Cosma et al. 2013; Bossew 2015; Ciotoli et al. 2017). With this in mind, investigations concerning the radon concentration of in-soil gas, soil permeability, surface radon exhalation (house permeability) and indoor radon concentration were

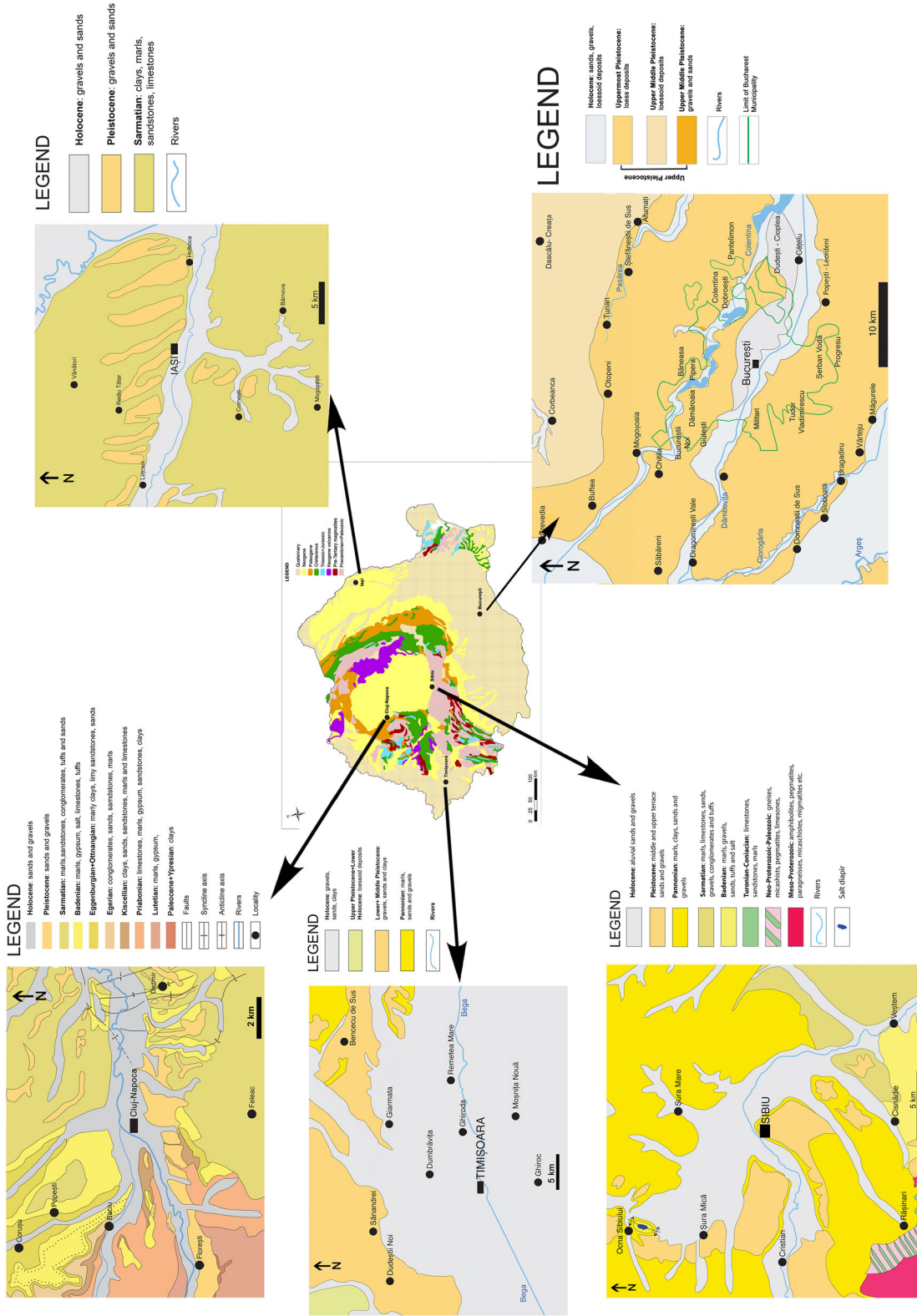
performed in five of the largest municipalities in Romania.

### Study areas

The five cities considered for the present study, Bucharest, Cluj-Napoca, Iași, Sibiu and Timișoara, are scattered throughout the country. Three cities are located on the east, south and west plains, while the other two are situated within the intra-Carpathian depression. The geological background of Romania and the five cities considered are presented in Fig. 1.

From a geological point of view, the city of Bucharest is located in the central part of the Wallachian sector of the Moesian Platform, involving two major structural units: a metamorphic sole and cross-cutting magmatic rocks, overlain by a sedimentary cover (Mutihac 1990; Ionesi 1994). Quaternary sedimentary deposits (gravel and sands of Colentina Formation and loess deposits of the Upper Middle Pleistocene; loess deposits and sands and gravels of the lower terraces of uppermost Pleistocene; detrital deposits of the Holocene lower terraces and riverbeds) are spread on the whole area of Bucharest. Borehole data (Liteanu 1952) indicate the presence in subsurface of two important Pleistocene lithostratigraphic units: The Frățești Formation (Lower Pleistocene) and Coconi Formation (Middle Pleistocene). The basal late Pleistocene (Mostișteea Fm.) is not cropping out in the studied area (Liteanu 1953, 1956; Andreescu et al. 2011, 2013).

Timișoara municipality is located on the southeastern border of the Pannonian Basin, the Miocene-Quaternary back-arch extensional basin, occurred after the Styrian phase (*sensu* Stille). The geological structure of this area consists of the units of the pre-Neogene basement (i.e., thrusting nappes, suture zones, magmatic bodies, sedimentary deposits, etc.), bordered by fault systems reactivated successively during Alpine movements and neo-structures (deep blocks, grabens, horsts, etc.) and controlled by normal faults (Polonic 1985). The Quaternary is largely exposed all over this area of the basin and mainly consists of clastic and clay deposits. The older Pleistocene fluvial terrace deposits are covered by Holocene alluvial deposits consisting of interbedded clays, sand and gravels. (Codarcea et al. 1968; Mircescu 1982; Simionescu et al. 1989).



**Fig. 1** The geological background of Romania and the setting of the five investigated perimeters, after Geological map of Romania, IGR, 1: 200000 (Sheet L-35-XXXIII for Bucharest, Sheet L-34-XXII for Cluj-Napoca, Sheet L-35-X: L-35-XI for Iași, Sheet L-34-XXII for Timișoara)

Iași municipality and its surroundings are located in the central-eastern sector of the Moldavian Platform. This platform consists of two major units: a metamorphic and magmatic sole, transgressed by a sedimentary cover (Mutihac 1990; Ionesi 1994; Răileanu et al. 2012). Carbonate, terrigenous and clastic Sarmatian rocks and Quaternary deposits (loess, as well as sand, gravel of the Pleistocene fluvial terraces, as well as Holocene alluvial deposits of the Bahlui River) are exposed in Iași and its neighborhoods area (Brânzilă 1999).

The city of Sibiu is situated on the southern border of the Transylvanian Depression, nearby the contact area with the Carpathians metamorphic rocks. The basement of the whole area concerns Precambrian metamorphic schists and upper Cretaceous sedimentary formations, transgressed by Neogene formations, on their turn overlapped by Quaternary deposits (Mutihac 1990). In Sibiu area, Badenian contains marls, gravels and sands, Sarmatian contains sandy marls, dolomite volcanic tuffs, sands, gravel and conglomerates, Pannonian contains marls, clays, sands and gravels, Pleistocene contains gravel and sands specific to medium and upper fluvial terraces and Holocene contains sands and gravels belonging to the lower alluvial terraces (Vancea and Ungureanu 1960; Ghiurcă 1966; Ciupangea et al. 1970; Lubenescu 1981; Gheorghian et al. 1970; Gheorghian and Gheorghian 1994).

The city of Cluj-Napoca is located in the north-western side of the Transylvanian Depression, to east of the Gilău Mountain's metamorphic rocks. On the area of this city, the post-“Iaramian” deposits evidence three partly superposed sedimentary basins: i. latest Cretaceous-early Miocene; ii. early Miocene; iii. Middle Miocene (Badenian)-Pannonian, occurred after the erection of the thrusting nappes of Pienides and Peri-Getides (Balintoni et al. 1998). The Cluj-Napoca area exposes mainly the rocks of the first and the last of these sedimentary basins. Quaternary deposits (various Pleistocene fluvial terraces and alluvial Holocene rocks of Someșul Mic and Nadăș rivers and their tributaries) are completing the sedimentary succession. (Răileanu and Saulea 1955, 1956; Mészáros and Clichici 1976, 1988; Mészáros and Ianoliu 1989; Filipescu 1999, 2011 and references therein).

## Materials and methods

Solid-state nuclear track detectors were placed indoors in 1000 residential buildings for two consecutive campaigns. Each campaign lasted 6 months. In each building were placed 2 passive detectors; one for each of the two most occupied rooms of the house. The working protocol for passive measurements has been previously described elsewhere (Cucoș et al. 2012). Based on passive measurement results, 100 houses with radon concentrations above 200 Bq/m<sup>3</sup> in at least one room were chosen for detailed diagnostics. Only these 100 houses were analyzed in the present study.

Soil gas radon concentration was determined in situ, using RM-2 (Radon v.o.s., Czech Republic) portable soil radon monitor and applying the Neznal method (Neznal et al. 2004). The method requires determining the concentration of radon from soil gas samples extracted from a depth of 80 cm using a metal probe, 15 sampling points for each 800 m<sup>2</sup> of bare soil. The number of the samples collected varied according to the available soil surface of each property. Graphical representations of the radon measurement protocol in soil, implemented in the present study, have been published by Cosma et al. (2013). The quality assurance has been achieved by participating in frequent intercomparison exercises with regard to radon measurements in soil (Burghele et al. 2019). Soil permeability measurements were carried out using Radon-Jok (Radon v.o.s., Czech Republic) in order to assess the radon potential (RP) and radon index (RI) of the building site, essential for the identification of “hot” areas in terms of radon activity concentration. The permeability of the soil is calculated based on Darcy's equation, regarding the flow of fluids through porous media (Neznal and Neznal 2005; Lupulescu et al. 2018). The principle of the Radon-Jok measurements is based on the ability to extract gas from the soil with the help of negative pressure. The RP is calculated according to the equation (Neznal et al. 2004):

$$RP = \frac{\text{3rd quartile of Radon Concentration in Soil} - 1}{-\log(\text{3rd quartile of Soil Permeability}) - 10}$$

A building site's radon index indicates the level of risk of radon release from bedrock, and it is the qualitative expression of the RP of a building site. If  $RP < 10$ , then RI is *low*; if  $10 \leq RP < 35$ , then RI is

medium; if  $35 \leq RP$ , the RI is high. The radon index of a building reflects the degree of radiation protection a building needs, which depends on the soil characteristics and the building's foundation type.

Gamma spectrometry measurements were performed on soil and building materials collected from the investigated houses. The activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  was measured. Present study is discussed only the  $^{226}\text{Ra}$  content due to its direct correlation with indoor radon. The activity concentration of  $^{226}\text{Ra}$  was measured after a month of storage using the gamma lines of the short-lived radionuclide daughters of  $^{222}\text{Rn}$  ( $^{214}\text{Pb}$  at 295 keV and 351 keV and  $^{214}\text{Bi}$  at 609 keV). Samples were analyzed with a high-resolution gamma spectrometer equipped with a high-purity germanium (HPGe) well-type ORTEC GEM detector, having a FWHM of 1.92 keV at 1.33 MeV permitting the detection of low gamma energies. The activity concentration was calculated using the relative method with IAEA 385,327,447 standards. The method has been described in a previous work by Cosma et al. (2013).

Measurements to determine radon exhalation ( $\Phi$ ) and leakages from cracks were performed for each house using several devices. Rad7 (DurrIDGE Company Inc., USA), RTM1688-2 (Sarad GmbH, Germany), Alpha Guard (Saphymo GmbH, Germany) and Radim 3A-Eman (Jr Plch, Czech Republic), each coupled to an accumulation chamber, were used to determine the exhalation rate of radon from the flooring in the investigated houses. In order to assess the distribution of radon entry in a room, samples of air were collected from all visible cracks, in the slab (Nezmal et al. 2004). The samples were collected using a 150-ml Janet syringe, and the Luk3P (J. Plch, Czech Republic) detector equipped with Lucas cells was used to determine the radon concentrations. The determination of radon exhalation and presence in leakages have a qualitative significance, which helps to establish the radon tightness of the building.

Data on  $\text{CO}_2$ , CO, volatile organic compounds (VOC), relative humidity, pressure and temperature were collected using ICA system, developed by LiRaCC (Tunyagi et al. 2019). Parameters were reported as quantitative data, except for VOC that was reported as qualitative data. The energy efficiency of the building was calculated based on the national methodology (Romanian Energy Performance Methodology 2006) for energy certification by taking

into account the climate data, building envelope properties, glazed area, heating, domestic hot water and lighting system. Several steps were necessary to determine the energy consumption: determination of the global insulation level; calculation of heat losses and heat gains; other data: HVAC systems efficiency, occupancy scenario, building shading, type of heat emissions system. The sum of these factors was taken into consideration representing the influence of the occupational factor on the indoor radon concentration.

Statistical analysis of the data was performed by using SPSS software, version 24 (SPSS Inc., USA). The statistical distribution of the data was evaluated using the D'Agostino-Pearson test. For the sample comparison, the nonparametric Mann-Whitney test was performed. The Chi-square ( $\chi^2$ ) test was used to evaluate the degree of association between qualitative variables. In order to evaluate the intensity of the relationship between the examined quantitative variables, the Pearson correlation coefficient was calculated. The multivariate analysis was conducted using log-transformed indoor radon data as dependent variable. The stepwise regression procedure was used, and only the factors that had a statistically significant influence on dependent variable ( $p < 0.05$ ) were taken into account. The significance level  $\alpha$  was chosen at 0.05.

All radon investigations were carried out in situ by the *Constantin Cosma* Radon Laboratory (LiRaCC) from Faculty of Environmental Sciences and Engineering, while gamma measurements were performed in the laboratory of Environmental Radioactivity and Nuclear Dating Centre of Interdisciplinary Research Institute on Bio-Nano-Science. Both institutions, belonging to the Babeş-Bolyai University, Cluj-Napoca, Romania, have implemented standardized international procedures for quality control and assurance of measurements.

## Results and discussions

### Statistical analysis

The statistical analysis of the experimental results obtained for the 100 houses follows both the descriptive presentation of the obtained results and the evaluation of the degree of association between the investigated parameters.

**Table 1** Descriptive statistics of the parameters monitored in the 100 houses

Parameter	Min.	Max.	Median	A.M.	S.D.	G.M.	
Indoor Rn (Bq/m <sup>3</sup> )	150	1221	309	356	176	325	
$\Phi$ max $\times 10^{-3}$ (Bq/m <sup>2</sup> /s)	3	99	8	13	16	9	
Q3 Rn <sub>soil</sub> (kBq/m <sup>3</sup> )	6	97	34	39	20	34	
RP	5	133	29	33	21	28	
CO <sub>2</sub> (ppm)	432	3375	1083	1213	638	1078	
RH (%)	16	70	42	42	10	41	
T (°C)	13	31	21	22	3	22	
$\Phi$ max maximum exhalation rate per house, Q <sup>3</sup> third quartile, RP radon potential	Energy consumption (kWh/m <sup>2</sup> )	102	500	210	216	63	208
	<sup>226</sup> Ra (Bq/kg)	8	283	32	48	57	33

### Univariate analysis

The normal distribution of log-transformed data on the radon concentration was confirmed by the D'Agostino-Pearson test ( $p > 0.05$ ). Table 1 presents a geometric mean of measurements for residential radon concentration of 325 Bq/m<sup>3</sup>, being about 4 times the geometric mean reported for Romania (Cosma et al. 2013; Muntean et al. 2014). This is explicable, given that the selection of the 100 houses in this project had as a main criterion a radon concentration above 200 Bq/m<sup>3</sup>.

The CO<sub>2</sub> concentration, reported here for 1 week, had a geometric mean of 1078 ppm with limits ranging from 432 to 3375 ppm. Similar to the distribution of radon concentration indoors, the CO<sub>2</sub> concentration had a log-normal distribution, which is confirmed by applying the D'Agostino-Pearson test to the log-transformed data.

The diagnosis of houses involved both the determination of the exhalation rate in the floor and the identification of cracks through which radon can infiltrate from the soil into the indoor air. The rate of exhalation was below the detection limit ( $3 \times 10^{-3}$  Bq/m<sup>2</sup>/s) in 33 of the 100 investigated houses. In the rest of the houses, the exhalation rate exhibited an extremely high variation with limits ranging from  $3 \times 10^{-3}$  Bq/m<sup>2</sup>/s to  $99 \times 10^{-3}$  Bq/m<sup>2</sup>/s and an arithmetic mean of  $13 \times 10^{-3}$  Bq/m<sup>2</sup>/s. In 4 out of 6 houses where the exhalation rate is higher than  $28 \times 10^{-3}$  Bq/m<sup>2</sup>/s, the flooring is placed directly on soil or slag, without any form of concrete screed in-between. In terms of leakages, only those situations where the measured value was above the threshold of 1 kBq/m<sup>3</sup> were taken into account. In only 12 houses, the floor type (tiles, laminate flooring

**Table 2** Concentration of indoor radon activity versus radon index in the soil

Radon index	No. of houses with indoor radon conc. (Bq/m <sup>3</sup> )	
	< 300	≥ 300
Low	1	3
Medium	21	41
High	11	20

or the presence of concrete screed) did not allow radon leakages above this threshold. For the remaining 88 houses, between 1 and 18 cracks with an arithmetic mean of 6 cracks per house were identified as radon entry points. As a maximum value of radon measured from cracks, the variation is extremely high, ranging from 1 to 27 kBq/m<sup>3</sup>, the coefficient of variation being 85%.

In order to calculate the radon potential (RP), the 75% (Q3) percentile of soil radon concentration was considered. Thus, the value calculated for Q3 was between 6 and 97 kBq/m<sup>3</sup> with an arithmetic mean of 39 kBq/m<sup>3</sup>. The radon potential was calculated based on the Q3 of the radon concentration in soil, and the permeability of the soil ranged between 5 and 133 with an arithmetic mean of 33. Radon potential values < 10 indicate a low index, between 11 and 34 medium and > 35 a high index. The majority (62) of the investigated lots presented a medium radon index, a third (33) showed a high radon index, while only in 4 locations the radon index was low.

**Table 3** Statistical correlations determined between relevant parameters

Spearman correlation for <sup>226</sup> Ra	Soil	0.35	− 0.07	0.5	0.65**	n/a	n/a	n/a	n/a
	Building material	0.08	0.55**	− 0.17	− 0.3	n/a	n/a	n/a	n/a
Variable		Indoor Rn	Φ max	Q3 Rn <sub>soil</sub>	RP	CO <sub>2</sub>	Temp	RH	Energy
Pearson correlation	Indoor Rn		0.05	− 0.12	0.1	0.26*	− 0.11	0.30**	0.13
	Φ max			− 0.1	0.14	− 0.04	0.21	− 0.14	− 0.07
	Q3 Rn <sub>soil</sub>				0.33**	0.13	− 0.15	0.13	0.02
	RP					0.06	0.19	0.1	0.04
	CO <sub>2</sub>						0.01	0.51**	0.29**
	Temp							− 0.51**	− 0.02
	RH								0.27**
	Energy								

\*Correlation is significant at the 0.05 level (2-tailed)

\*\*Correlation is significant at the 0.01 level (2-tailed)

**Bivariate analysis**

By calculating the Pearson correlation coefficient, a weak correlation between the residential radon concentration and the CO<sub>2</sub> concentration ( $r = 0.26$ ,  $n = 97$ ,  $p = 0.01$ ) was obtained. This may indicate a poor ventilation that will lead to an accumulation of both CO<sub>2</sub> and radon concentration, even if sources of origin are different. No statistically significant correlation between indoor radon concentration and parameters such as floor radon exhalation, radon concentration in cracks, soil radon concentration, soil potential or radon index, was observed. Table 2 shows the distribution of houses based on the concentration of indoor radon and radon index of the surrounding soil. Although the radon index takes into account both soil radon concentration and permeability, this index cannot be a surrogate of indoor radon measurements or an indicator for the prioritization of houses that will be investigated from residential radon. This is also confirmed by the  $\chi^2$  test indicating that there is no statistically significant dependence between the radon index and the residential radon concentration ( $p > 0.05$ ). Table 3 presents a tabulated representation of correlated parameters.

On the other hand, a moderate correlation between residential radon concentration and humidity was obtained ( $r = 0.3$ ,  $n = 97$ ,  $p < 0.01$ ). As expected, an inverse correlation was obtained between temperature

and humidity ( $r = - 0.5$ ,  $n = 97$ ,  $p < 0.01$ ). A moderate correlation was also obtained between CO<sub>2</sub> concentration and energy consumption ( $r = 0.3$ ,  $n = 92$ ,  $p < 0.01$ ).

A total number of 30 samples of soil (10) and building material (20) were submitted to gamma analysis. A good Pearson correlation ( $r = 0.72$ ,  $n = 10$ ,  $p < 0.05$ ) was obtained between the radon potential and the <sup>226</sup>Ra content for the assessed soil samples. Although such a correlation could be expected, the result should be treated with caution in view of the low variation range of <sup>226</sup>Ra in the evaluated samples (23–39 Bq/kg) and the low number of samples evaluated. According to EC RP (1999), the concentrations obtained for radionuclides analyzed in the samples taken were within normal limits. Due to the low number of samples, a Spearman’s correlation was also applied for the gamma data.

By applying the Mann–Whitney nonparametric test, a statistically significant difference was obtained between the medians of the radon exhalation rate based on the presence of concrete screed under the flooring, the latter’s absence yielding a significantly greater median ( $17.5 \times 10^{-3}$  vs.  $6.9 \times 10^{-3}$  Bq/m<sup>2</sup>/s). A similar situation was also obtained by the type of ceiling, a wooden ceiling leading to a significantly higher floor radon exhalation compared to the concrete ceiling ( $p < 0.01$ ). This result can be attributed to the stack effect caused by the inner–outer pressure

difference which is amplified by the lack of a sealed ceiling thus leading to an additional infiltration of sub-slab air. By determining the partial correlation coefficient between the radon exhalation rate and the ceiling type, the control variable, being the presence of the concrete screed, showed a reduction in the correlation coefficient from  $r = 0.26$  ( $p = 0.03$ ) to  $r = 0.14$  ( $p > 0.05$ ). Therefore, the presence of concrete screed has a mediating effect on the impact of the ceiling type on the rate of exhalation. On the other hand, the impact of the ceiling type on the indoor radon concentration is statistically insignificant, the median of the concentration of radon in houses with a wooden ceiling being similar to that of the houses with concrete ceiling ( $p > 0.05$ ).

Multivariate linear regression was used to investigate the impact of measured parameters on the mean value of residential radon concentration. The percentage of variability of residential radon concentration is explained by the model for only 15% of cases, the main contributors being the existence of concrete screed below the floor, respectively, the indoor humidity.

### Influence of geological factors

The studied locations in the area of Bucharest overlap the Middle Upper Pleistocene deposits (loessoid deposits, Colentina Formation) and the uppermost Pleistocene formations (loess deposits). The sedimentary evolution of the Colentina Fm. can be related to the actual water courses (e.g., Argeş and Dâmboviţa rivers). The Pleistocene discharge of these rivers used to be stronger than it is today. This hypothesis is supported by the amount of carried sediments and flow paths average. The springs of these rivers are located in metamorphic areas corresponding to the Făgăraş metamorphic lithostratigraphic units (Sebeş-Lotru terranes) (Balintoni 2014). The mineralogical composition of these detrital deposits may reflect the petrographic and mineralogical imprint of the above-mentioned crystalline units. They bear amphibolites, micaschists, ocular gneisses, paragneisses, which are known for their relatively increased radiation potential (Stoici and Tătaru 1988). The petrographic characteristics of these deposits may explain the relatively high values of in-soil and indoor radon concentrations. On the other hand, loess-like deposits are known to have a

low radioactive potential (Kozak et al. 2005). In addition, such low permeable formations should block the migration of radon in soil. However, indoor and in-soil measurements performed on emplacements overlapping loessoid and loess deposits revealed high radon concentration values. This phenomenon can be explained by the following hypothesis. A dry loess column can be formed under the constructions and by contracting can lead to the development of high amplitude cracks. As a consequence, radon gas can migrate through such flow paths from the underlying rock formations with a higher radioactive potential (Tondeur et al. 1996; Kozak et al. 2005).

In the Timișoara municipality, the emplacements are located on Holocene alluvial deposits. The existing geotechnical data indicate that these deposits are covered by up to 1-m-thick soils. This aspect suggests a possible link between in-soil radon values and these alluvial deposits. Soil and indoor radon values can be related to the lithological heterogeneity of these deposits and the possible occurrence of rock fragments with increased radioactivity. Such hypothesis may be explained by the source area of the alluvial deposits. The Bega River carries reworked material from the Poiana Ruscă Mountains, where metamorphic rocks are the main petrographic local component. They consist of various petrotypes (quartz schists, micaschists, quartzites, amphibolites, paragneisses) belonging to the Padeş and Făgăraş terranes (Balintoni et al. 2014). Such rocks are known for their increased radioactive potential (Stoici and Tataru 1988). Geotechnical studies indicate the presence of silty clay interbedding which can explain the reduced permeability measured in some points around the emplacements.

The majority of the studied locations from Iași overlap Sarmatian clay-rich deposits. In two situations, they lie directly on Pleistocene upper terrace detrital deposits. It is known that some Sarmatian clay varieties recorded a high radioactivity (Otton 1991; Cosma and Jurcut 1996; Appleton 2007; Drolet et al. 2013). The possible presence of such clays may explain both indoor and in-soil radon increased values recorded in this study. The lithological heterogeneity of these deposits and the presence of clayish soils (as indicated by geotechnical data) may explain the differences in permeability values recorded in various measurement points around the emplacements. Middle and high in-soil radon values recorded for the



Pleistocene deposits can be explained by their mineralogical and lithological heterogeneity together with the possible presence of rock fragments with increased radiation potential. These features may also explain both the indoor measurement values and the measured permeability differences from some points surrounding the emplacements.

The studied locations of Sibiu overlap both the Holocene lower terrace deposits of Cibin and Sebeş rivers and the Pleistocene detrital upper terrace deposits of the Cibin Valley. The composition of the alluvial and upper terrace deposits may reflect the petrographic and mineralogical features of the geological formations crossed by the Cibin and Sebeş rivers in Quaternary. Such formations comprise metamorphic rocks (gneiss, micaschists, few amphibolites and pegmatites from the Sebeş-Lotru terranes) which are known for their higher radioactive potential (Stoici and Tătaru 1988). These aspects could explain both the relatively high indoor and in-soil radon values resulting from measurements performed around the emplacements.

The investigated houses from Cluj-Napoca area overlap Quaternary (Pleistocene and Holocene upper terrace and alluvial formations), Neogene (Sarmatian, Badenian) and Paleogene (Rupelian) deposits. The composition of Quaternary deposits can reflect the mineralogical and petrographical features of the geological formations crossed through time by the Someşul Mic River and its tributaries. Such formations include magmatic (e.g., Muntele Mare granite) and metamorphic rocks (micaschists and paragneisses belonging to the Someş terrane), which are known for their higher radioactive potential (Balintoni et al. 2009; Stoici and Tătaru 1988). This aspect can explain the medium and high radon values in soil and the relatively high values from indoor measurements. The lithological heterogeneity characterizing these detrital deposits could also explain the variation of in-soil measurements. The emplacements overlapping the Sarmatian and Badenian deposits presented a peculiar feature in terms of in-soil measurement values. Radon potential values tend to migrate toward a lower limit of the interval defining average values. In the meantime, these values are lower than the average estimated for emplacements overlapping Quaternary deposits. In a single case, the emplacement overlaps Lower Rupelian deposits with marls, sandy clays, limestones and coarse limestone interbeddings. Geotechnical data

indicates the presence of up to 0.5-m-thick black clays with plastic consistency, followed by 1-m-thick dusty clay with plastic consistency. This black clay horizon explains the medium radon potential together with relatively high values from indoor measurements (Drolet et al. 2013).

## Conclusions

The variation of indoor radon could not be directly correlated with radon variation in the soil. Considering that all houses in the study had indoor radon concentration values exceeding 200 Bq/m<sup>3</sup> and that radon variation in soil was quite significant (covering from low to high index), but not directly correlated with the variation of indoor radon, it can be certainly stated that, along with geology, the factors related to the construction characteristics play an extremely important role in the accumulation of radon indoor. It has been noted that all these energy-efficient houses have a rather high average of CO<sub>2</sub> which in turn has been positively correlated with indoor radon levels. This observation leads to the conclusion that energy-efficient houses tend to have a low air exchange rate that can produce negative effects on the indoor air quality through the accumulation of CO<sub>2</sub>, radon or humidity. On the other hand, the absence of concrete screed combined with thermal retrofit on old houses may present additional challenges for radon mitigators. The final conclusion drawn from this study is that preventive radon measures should be applied for all new buildings, regardless of the radon index, and that indoor long-term passive measurements are the most reliable method for assessing the radon risk for human health.

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## References

Andrescu, I., Codrea, V., Enache, C., Lubenescu, V., Munteanu, T., Petculescu, AI, et al. (2011). Reassessment of the pliocene/pleistocene (neogene/quaternary) boundary in the

- Dacian Basin (Eastern Paratethys) Romania. *Oltenia, Studii și comunicări, Științele Naturii*, 27(1), 197–220.
- Andrescu, I., Codrea, V., Lubenescu, V., Munteanu, T., Petculescu, A., Știucă, E., et al. (2013). New developments in the Upper Pliocene–Pleistocene stratigraphic units of the Dacian Basin (Eastern Paratethys), Romania. *Quaternary International*, 284, 15–29. <https://doi.org/10.1016/j.quaint.2012.02.009>.
- Appleton, J. D. (2007). Radon: Sources, health risks, and hazard mapping. *Ambio*, 36, 85–90. <https://doi.org/10.1579/0044-7447>.
- Appleton, J. D., & Miles, J. C. H. (2010). A statistical evaluation of the geogenic controls on indoor radon concentrations and radon risk. *Journal of Environmental Radioactivity*. <https://doi.org/10.1016/j.jenvrad.2009.06.002>.
- Balintoni, I., Balica, C., Cliveti, M., Li, L.-Q., Hann, H. P., Chen, F., et al. (2009). The emplacement age of the Muntele Mare Variscan granite (Apușeni Mountains, Romania). *Geologica Carpathica*, 60(2009), 495–504. <https://doi.org/10.2478/v10096-009-0036-x>.
- Balintoni, I., Balica, C., Ducea, M. N., & Hann, H. (2014). Peri-Gondwanan terranes in the Romanian Carpathians: A review of their spatial distribution, origin, provenance, and evolution. *Geoscience Frontiers*. <https://doi.org/10.1016/j.gsf.2013.09.002>.
- Balintoni, I., Mészáros, N., & Györfi, I. (1998). La Transylvanie, dépression et bassins. *Studia Universitatis Babeș-Bolyai, Geologia*, 43(1), 43–59.
- Ball, T., Cameron, D., Coleman, T., & Roberts, P. (1991). Behaviour of radon in the geological environment: A review. *Quarterly Journal of Engineering Geology*. <https://doi.org/10.1144/GSL.QJEG.1991.024.02.01>.
- Bossew, P. (2015). Mapping the geogenic radon potential and estimation of radon prone areas in Germany. *Radiation Emergency Medicine*, 4(2), 13–20.
- Bossew, P., Dubois, G., & Tollefsen, T. (2008). Investigations on indoor radon in Austria, part 2: Geological classes as categorical external drift for spatial modelling of the radon potential. *Journal of Environmental Radioactivity*. <https://doi.org/10.1016/j.jenvrad.2007.06.013>.
- Brânzilă, M. (1999). Geologia părții sudice a Câmpiei Moldovei. In Corson (Ed.), Iași.
- Burghel, B., Tenter, A., Cucuș, A., Dicu, T., Moldovan, M., Papp, B., et al. (2019). The FIRST large-scale mapping of radon concentration in soil gas and water in Romania. *Science of the Total Environment*. <https://doi.org/10.1016/j.scitotenv.2019.02.342>.
- Ciotoli, G., Voltaggio, M., Tuccimei, P., Soligo, M., Pasculli, A., Beaubien, S. E., et al. (2017). Geographically weighted regression and geostatistical techniques to construct the geogenic radon potential map of the Lazio region: A methodological proposal for the European Atlas of Natural Radiation. *Journal of Environmental Radioactivity*. <https://doi.org/10.1016/j.jenvrad.2016.05.010>.
- Ciupănean, D., Paucă, M., & Ichim, T. (1970). *Geologia Depresiunii Transilvaniei*. București: Academiei R.S.R.
- Codarcea, A., Drăgulescu, A., Hinculov, L., Mihăilă, N., Nica, E. (1968). *Geological map of Romania*. Geological Institute of Romania, 1:200000, sheet L-34-XXII, Timișoara.
- Cosma, C., Cucuș, A., Papp, B., Begy, R., Dicu, T., Moldovan, M., et al. (2013). Radon measurements and radon remediation in Băița-Ștei prone area. *Carpathian Journal of Earth and Environmental Sciences*, 8(2), 191–199.
- Cosma, C., & Jurcuș, T. (1996). *Radonul și mediul înconjurător*. Cluj-Napoca: Dacia.
- Cucuș Dinu, A., Cosma, C., Dicu, T., Begy, R., Moldovan, M., Papp, B., et al. (2012). Thorough investigations on indoor radon in Baita radon-prone area (Romania). *Science of the Total Environment*, 431, 78–83. <https://doi.org/10.1016/j.scitotenv.2012.05.013>.
- Darby, S., Hill, D., Auvinen, A., Barros-Dios, J. M., Baysson, H., & Bochicchio, F. (2005). Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies. *British Medical Journal*. <https://doi.org/10.1136/bmj.38308.477650.63>.
- Demoury, C., Ielsch, G., Hemon, D., Laurent, O., Laurier, D., & Clavel, J. (2013). A statistical evaluation of the influence of housing characteristics and geogenic radon potential on indoor radon concentration in France. *Journal of Environmental Radioactivity*. <https://doi.org/10.1016/j.jenvrad.2013.08.006>.
- Drolet, J.-P., Martel, R., Poulin, P., Dessau, J.-C., Lavoie, D., Parent, M., et al. (2013). An approach to define potential radon emission level maps using indoor radon concentration measurements and radiogeochemical data positive proportion relationships. *Journal of Environmental Radioactivity*. <https://doi.org/10.1016/j.jenvrad.2013.04.006>.
- EC RP 112 (1999). European Commission, Radiation protection 112, Radiological Protection Principles concerning the Natural Radioactivity of Building Materials, Directorate-General Environment, Nuclear Safety and Civil Protection, EC Luxembourg.
- Filipescu, S. (1999). The significance of foraminifera fauna from the Feleac Formation (Transylvanian Basin, Romania). *Studia Universitatis Babeș-Bolyai, Geologia*, 44(2), 125–131.
- Filipescu, S. (2011). Cenozoic lithostratigraphic units in Transylvania. In I. Bucur & E. Sășăran (Eds.), *Calcareous algae from Romanian Carpathians. Field trip Guidebook* (pp. 37–48). Presa: Universitară Clujeană.
- Gheorghian, M., & Gheorghian, D. (1994). Datation biostratigraphiques des formations Miocenes du sud de la Transylvanie a partir des foraminiferes. *Miocene From Transylvanian Basin, Romania, Cluj-Napoca*, 4, 125–134.
- Gheorghian, D., Lubenescu, V., & Olteanu, R. (1970). Contribuții la stratigrafia Miocenului din sudul Transilvaniei. *D.S.S. Com Geologic București*, 57(4), 55–56.
- Ghiurcă, V. (1966). Briozoarele tortoniene de la Tălmăcel și Cislăndia-Sibiu. *Studia Universitatis Babeș-Bolyai, Geologia-Geographia*, 1, 99–104.
- Gundersen, L.C.S., Schumann, R.R., Otton, J.K., Dubiel, R.F., Owen, D.E., Dickinson, K.A. (1992). Geology of radon in the United States. In A.E. Gates and L.C.S. Gundersen (Ed.), *Geologic controls on Radon*, Boulder, Colorado, Geological Society of America. Special Paper 271.
- Ionesi, L. (1994). *Geologia unităților de platformă și a orogenului nord-dobrogean*. București: Tehnică.
- Kemski, J., Klingel, R., Siehl, A., & Valdivia-Manchego, M. (2009). From radon hazard to risk prediction-based on geological maps, soil gas and indoor measurements in

- Germany. *Environmental Geology*. <https://doi.org/10.1007/s00254-008-1226-z>.
- Kemski, J., Klingel, R., Stegemann, A., & Siehl, R. (2005). Radon transfers from ground to houses and prediction of indoor radon in Germany based on geological information. *Radioactivity in the Environment*. [https://doi.org/10.1016/S1569-4860\(04\)07103-7](https://doi.org/10.1016/S1569-4860(04)07103-7).
- Kemski, J., Siehl, A., Stegemann, R., & Valdivia-Manchego, M. (2001). Mapping the geogenic radon potential in Germany. *Science of the Total Environment*. [https://doi.org/10.1016/S0048-9697\(01\)00696-9](https://doi.org/10.1016/S0048-9697(01)00696-9).
- Krewski, D., Lubin, J. H., Zielinski, J. M., Alavanja, M., Catalan, V. S., Field, R. W., et al. (2005). Residential radon and risk of lung cancer: a combined analysis of 7 North American case-control studies. *Epidemiology*, 16(2), 137–145.
- Liteanu, E. (1952). Geologia zonei oraşului Bucureşti. *Comitetul Geologic Inst Bucureşti*, 1, 3–80.
- Liteanu, E. (1953). *Geologia Ținutului de câmpie din bazinul inferior al Argeşului si a teraselor Dunării*. Inst. Geol., St. tehn. Econ., E/2 Bucuresti, 5–78.
- Liteanu, E. (1956). *Geologia si hidrogeologia ținutului dunărean dintre Argeş si Ialomița*. Bucureşti.
- Lubnescu, V. (1981). Studiul biostratigrafic al Neogenului superior din sud-vestul Transilvaniei. *Anuarul Institutului de Geologie și Geofizică*, 58, 123–202.
- Lupulescu, A., Dicu, T., Papp, B., & Cucuș, A. (2018). Determination of the monthly variation of radon activity concentration in soil. *Studia Universitatis Babeş-Bolyai, Ambientum*. <https://doi.org/10.24193/subbambientum.2018.1.05>.
- Mészáros, N., & Clichici, O. (1976). *Pe poteci cu banuței de piatră. Ghid geologic al zonei Cluj*. Bucuresti: Sport-Turism.
- Mészáros, N., & Clichici, O. (1988). La geologie du Municipie Cluj-Napoca. *Studia Universitatis Babeş-Bolyai, Geologia-Geographia*, 33(1), 51–56.
- Mészáros, N., Ianoliu, C. (1989). Nannoplankton zones in the Oligocene deposits of the northwestern Transylvanian Basin. The Oligocene from the Transylvanian Basin, Romania. *Special volume, "Babeş - Bolyai" University*, 157–162. Cluj-Napoca.
- Mircescu, V. (1982). Zăcămintele de ape geotermale din Câmpia de Vest, in Schimbul de experiență: Utilizarea apelor geotermale, Eds. Consiliul Popular al Judetului Timiș, Institutul de proiectari Timișoara, Com. Sin. Com. Ing. Techn., (2), 36–50.
- Muntean, L. E., Cosma, C., Cucos (Dinu), A., Dicu, T., & Moldovan, D. V. (2014). Assessment of annual and seasonal variation of indoor radon levels in dwelling houses from Alba county, Romania. *Romanian Journal of Physics*, 59(1–2), 163–171.
- Mutihac, V. (1990). *Structura geologică a teritoriului României*. Bucureşti: Tehnică.
- Nazaroff, W. W. (1992). Radon transport from soil to air. *Reviews of Geophysics*, 30(2), 137–160.
- Neznal, M., & Neznal, M. (2005). Permeability as an important parameter for radon risk classification of foundation soils. *Annals of Geophysics*. <https://doi.org/10.4401/ag-3192>.
- Neznal, M., Neznal, N., Matolín, M., Barnet, I., & Miksova, J. (2004). *The new method for assessing the radon risk of building sites*. Prague: Czech Geological Survey.
- Otton, J. K. (1991). Potential for indoor radon hazards-A first geologic estimate, In Doe, B.R., (Ed.), Proceedings of a U.S. Geological Survey Workshop on Environmental Geochemistry, U.S. *Geological Survey Circular*, 1033, pp. 171–173.
- Polonic, G. (1985). Neotectonic activity at the eastern border of the Pannonian depression and its seismic implications. *Tectonophysics*, 47, 109–115.
- Răileanu, G., & Saulea, E. (1955). Contribuții la orizontarea și cunoașterea variațiilor de facies ale paleogenului din regiunea Cluj și Jibou (NV bazinului Transilvaniei). *Revista Universității CI Parhon și a Politehnicii București*, 8, 233–245.
- Răileanu, G., & Saulea, E. (1956). Paleogenul din regiunea Cluj și Jibou (NV bazinului Transilvaniei). *Anuarul Institutului Geologic al României*, 29, 272–308.
- Răileanu, V., Tătaru, D., Grecu, B., & Bala, A. (2012). Crustal models in Romania-II. Moldavian Platform and adjacent Areas. *Romanian Journal of Physics*, 57, 1438–1454.
- Romanian Energy Performance Methodology, 2006. Metodologie de calcul al performanței energetice a clădirilor. [https://www.mdrap.ro/userfiles/reglementari/Domeniul\\_XXVII/27\\_11\\_MC\\_001\\_1\\_2\\_3\\_2006.pdf](https://www.mdrap.ro/userfiles/reglementari/Domeniul_XXVII/27_11_MC_001_1_2_3_2006.pdf).
- Sachs, H. M., Hernandez, T. L., & Ring, J. W. (1982). Regional geology and radon variability in buildings. *Environment International*. [https://doi.org/10.1016/0160-4120\(82\)90016-2](https://doi.org/10.1016/0160-4120(82)90016-2).
- Simionescu, T., Codrea, V., & Trelea, N. (1989). La faune quaternaire de Lovrin (dép. Timis)-Banat. *Analele Științifice ale Universității "Al. I. Cuza" Iasi (serie nouă)*. *Geologie-Geografie*, 35, 41–53.
- Stoici, S., Tătaru, S. (1988). Uranium si Thorium. *Seria substante minerale utile*. In Tehnică (Ed.), București.
- Tunyagi, A., Dicu, T., Cucuș, A., Burghel, B., Dobrei, G., Lupulescu, A., Moldovan, M., Niță, D., Papp, B., Zsacsvai, K., Țenter, A., Beldean-Galea, M.S., Anton, M., Grecu, Ș., Cioloca, L., Miloș, R., Botoș, M.L., Chiorean, C.G., Catalina, T., Istrate, M.A., & Sainz, C. An innovative system for monitoring radon and indoor air quality. *Romanian Journal of Physics*, 2019 (Accepted).
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). (2000). *Sources and Effects of Ionizing Radiation, Report to the General Assembly* (pp. 1–10). New York: United Nations.
- Vancea, A., & Ungureanu, L. (1960). Asupra corelării depozitelor miopliocene din bazinul Transilvaniei pe baza de microfauna. *Stud. si cerc. de geolog. București*, 4, 619–625.
- WHO (2009). World Health Organization. *Handbook on Indoor Radon, a Public Health Perspective*. In Zeeb, Hajo, Shannoun, Ferid (Eds.), p. 110.

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