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ANOMALOUS RADON EMISSION AS PRE-SIGNAL OF MODERATE TO STRONG EARTHQUAKES IN VRANCEA GEOTECTONIC ACTIVE REGION IN ROMANIA

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Abstract. *As a potential precursor of earthquakes, this study aims to investigate temporal variations of radon (^{222}Rn) concentration levels in air near the ground by the use of solid-state nuclear track detectors (SSNTD) CR-39 (short term-10 days exposure time) in relation with some important seismic events recorded in Vrancea geotectonic active region, in Romania. The experimental observations reveal a strong correlation between the recorded radon emissions peaks associated with some moderate earthquakes of moment magnitude Mw ≥ 5.0 recorded during 2012-2022 period. The standard deviation of the radon measurements (s) was about 10% of the average radon concentration. The recorded pre-signals radon anomalies of earthquakes during eleven years monitoring period performed with solid state nuclear track detectors CR-39 suggest that earthquake precursors registered before moderate or strong seismic events are associated with some physical processes in or near the Vrancea earthquake fault zones or its neighbouring. This paper considered also the effects of meteorological parameters (air temperature, pressure, relative humidity, wind intensity and rainfall) on radon in air near the ground concentrations. The present results show existence of coupling between lithosphere-surfacesphere-atmosphere-ionosphere associated with preparation and seismic event occurring. Continuously monitoring of radon concentration anomalies in air near the ground in relation with Vrancea seismicity is an important issue and a surveillance tool in the field of earthquake hazard for Romania.*

Keywords: *radon anomalies, CR-39 Nuclear Track Detectors, earthquake, Vrancea, Romania*

1. INTRODUCTION

Earthquakes are the worst disastrous natural geological hazard that strikes the Earth surface. Cumulative stress energy in tectonic active regions worldwide manifests various earthquake precursors. Earthquakes are associated with stress/strain changes caused by seismic activity, ongoing rocks deformation and associated several geophysical fields' disturbances along the main active geologic faults. Is well known that fracto-emission signals (acoustic emissions, electromagnetic emissions, neutron emissions, radon and other gases like as helium, methane or carbon dioxide emissions from the crust to the surface) triggered by high-frequency pressure waves at the different scales, can be used as pre-signals tools for earthquake prediction and environmental protection [1].

Radon (^{222}Rn) is an odorless, colorless, inert, and radioactive noble gas (with half-life T1/2 = 3.8 days), occurring naturally in the environment, that emanates from rocks and soils as a result of the alpha decay of its parent, radium (^{226}Ra) in the decay series of uranium-238 (^{238}U). Among the noble gases it is the densest, having (atomic mass = 222), highest melting point (202 ºK), boiling point (211.5 ºK), critical temperature (377 ºK) and critical pressure (6.28 MPa). It is soluble in water, and its solubility decreases with increasing temperature [2]. Physical and chemical processes of ^{222}Rn gas emitted from the fault involve physical transport, diffusion in the atmosphere and disintegration in its α- and β-progeny. The great part of the natural radioactivity is not radioactive from radon itself, but from its short-lived alpha particle-emitting radon daughters, most notably ^{218}Po (radioactive T1/2=3 minutes), and ^{214}Po (radioactive T1/2=0.164 milliseconds), along with beta particles from ^{214}Bi (T1/2=19.7 minutes), where T1/2 is physical half-life [3].

Previous published studies regarding pre-signals seismic radon anomalies suggested that the recorded of amplitude anomalies were independent of moment magnitudes and epicentral distances of the related earthquakes, while time and duration of anomalies increase with magnitudes and epicentral distance [4].

The radon migrates through fractures and openings pore spaces escaping toward the surface by a combination of diffusive and advective transport processes [5]. Its concentration from the crust to the surface is significantly controlled by the spatial local uranium/radium abundance in rocks and soils distribution, and is mainly influenced by different geophysical and geochemical and meteorological factors systematically explained by Lithosphere–Atmosphere–Ionosphere Coupling (LAIC) model [6]. Earthquake preparation process is a transient dynamic The process and the integration of multi-sources geophysical, geochemical and seismic time series data will provide a complex pattern for earthquake forecasting.

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The main aim of this study was to identify radon anomalies possibly associated with the seismic activity in the Vrancea geotectonic active area. To this end, the temporal patterns of atmospheric parameters were also obtained and analyzed to account for their effect on radon time series.

In order to establish earthquake precursory phenomena using sudden changes in radon emanation, the present investigation aims to assess the role of seismo-induced radon (^{222}Rn) anomalies as significant precursors of moderate and strong earthquakes in Vrancea region. The temporal variability of the radon concentration was continuously monitored with CR-39 nuclear track detectors from January 2012 to April 2022.

2. Seismotectonic Background

Seismogenic Vrancea region in Romania features a complex geologic structure characterized by a sharp relief. From seismo-tectonic viewpoint, the Vrancea geotectonic active zone in Romania (Figure 1) placed in the curvature sector of the Eastern Carpathians (Romania) presents a peculiar source of seismic hazard, which represents a major concern in Europe [7]. Seismicity in this region is related to the subcrustal earthquakes located at the sharp bend of the Southeast Carpathians, and at the intersection of the East-European plate (North and North-West) with the Moesian (South) and Intra-Alpine (West) subplates, Vrancea region is considered one of the most seismically active areas in Europe, with a high potential of seismic hazard for the neighboring countries (Bulgaria, Hungary, Serbia, Republic of Moldavia, Ukraine). It is characterized by the occurrence of low moment magnitude (M_w) crustal (0-40 km depth) earthquakes (M_w<5.5 and moderate seismic activity) and intermediate depth (70-200 km) strong earthquakes ($6 \leq M_w \leq 8$) in a narrow epicentral and hypocentral region [8].

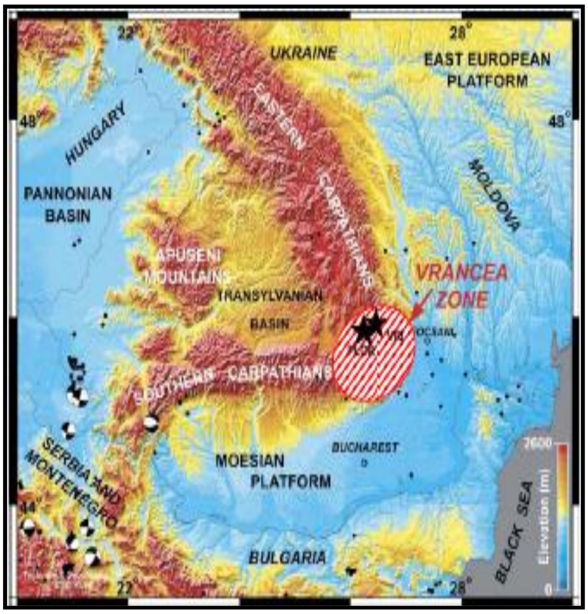

Figure 1. Location of Vrancea seismic zone on geomorphologic map of Romania.

Located at the border of the great East-European Platform, Romanian territory is an area of complex geological structure dominated by the presence of the Alpine Orogenic Belt of the Carpathian Mountains. The compressive stress field due to subduction of the Black Sea Sub-Plate under the Pannonian Plate generates faulting processes. The resulted fault plane is approximately parallel North Eastern-South Western oriented to the Carpathian Bend. A deep crustal fracture with dextral slip, the Peceneaga- Camena Fault is considered to be the North-Eastern boundary of the Moesian Platform. However, the Eastern unit of the Moesian Sub-Plate is described by a series of principal faults with a North-Western orientation and by a secondary system of North Eastern-South Western orientated faults. East of the Peceneaga-Camena Fault there are North-Western trending crustal fractures. The Black Sea Sub-Plate has a North-Western displacement along the “markers” formed by the Moesian and Eurasian Sub-Plates. Due to its peculiar tectonic regime and geodynamic processes, during the last 300 years, Vrancea seismogenic region featured 13 earthquakes with moment magnitudes (Mw) above 7, out of which seven events had Mw above 7.5 and three between 7.7 and 7.9 on Richter scale. Due to higher permeability and porosity relative to surrounding rocks, crustal faults and fractures are the main migration channels for carrier gases and radon gas from the deep layers of the crust to the surface [7].

3. Data & Methods

3.1. Data sets

Seismicity data have been provided by ROMPLUS catalogue (www.infp.ro/romplus) and (earthquakes.usgs.gov). Daily time series of the mean air temperature, rainfall and wind speed data were retrieved from http://www.soda-pro.com/web-services/meteo-data/merra and National Administration of Meteorology www.anm.ro, as well as by meteorological data recorded at the seismic stations.

3.2. Solid State Nuclear Track Detectors (SSNTD)

This study used passive sampling mode for radon monitoring in the surface air near the ground by Solid State Nuclear Track Detectors (SSNTD-CR-39) short time expositmeters, 10 days exposure, at 50 cm height above the ground, provided by Radon Analytics in Germany (http://www.radon-analytics.com), placed at four seismic stations: Plostina-PLOR and Vrancioaia-VRI, Cheia- Muntele Rosu-MLR, located on active fault in Vrancea zone, and Bucharest- Magurele- BUC (Figure 2).

The measuring range of approx. 15 Bq/m^3 up to over 5.000 Bq/m^3 enables the accurate determination of all radon concentrations usually occurring in air just above the ground in seismic fault zones. The detector itself - a plastic film, SSNTD CR-39 type consisting of a polycarbonate foil (approx. 1 to 10 cm^2 in size, approx. 0.1 to 1 mm thick) - is located inside the exposimeter housing. Air containing radon diffuses through the housing into the measuring chamber; radon decay products, dust and humidity are retained here. In addition, the sufficiently large diffusion resistance prevents the intrusion of thoron (Rn-220; half-life:

approx. 55 seconds). In the measuring chamber radon decays emitting alpha particles into its so-called radon daughters. These short-lived decay products are alpha-emitters too. The alpha particles create microscopic tracks in the film, which may be revealed and become visible under an ordinary optical microscope when treated with a properly chosen chemical reagent. A specific radon concentration and corresponding uncertainty are calculated in Bq/m^3, based on the number of the tracks, the size of the detector and the exposure time. This method proved to be fairly successful for seismic pre-signals exploration under varied meteorological conditions.

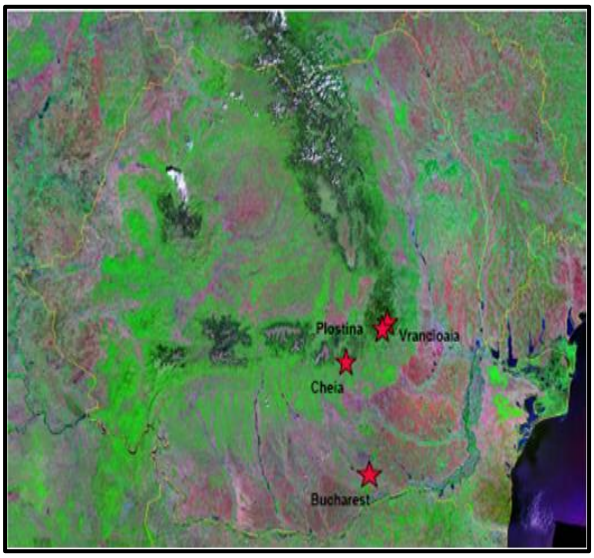

Figure 2. Location of pilot test areas for radon monitoring: Vrancioaia (VRI), Plostina (PLOR) in Vrancea seismogenic region, Cheia – Muntele Rosu (MLR), and Bucharest (BUC).

3.3. Statistical analysis used

Statistical methodologies have been applied to discriminate the effect of meteorological variables (air pressure, air temperature and rainfall rate) for precise identification of seismic induced anomalies.

The effect of atmospheric parameters variability on radon fluctuations should be reduced before attempting to associate radon anomalies with seismic events.

Enhanced radon and other gases emissions from the earth's surface preceding an earthquake, which are often perceivable by instrumental sensors, can be called an anomaly. Several studies have tried to determine the physical basis behind the radon anomaly that causes fluctuations in the air ionization. These studies discuss the theoretical and experimental results.

The identification of an anomaly in radon time series and the definition of the anomaly parameters (the duration, amplitude and precursor time) have been used by different researchers. However, radon variation crossing the 2σ confidence interval from the mean value at the monitoring site is typically identified as a significant anomaly. Radon concentration anomaly was calculated based on the following formula:

$$C_{Rn} \geq \overline{C_{Rn}} + 2\delta$$

where δ is standard -deviation, and radon variation crossing the 2δ confidence interval from the mean value at the monitoring site is typically identified as a significant anomaly. In good accordance to scientific literature on earthquake precursor anomalies, shape of the peak anomaly can be used as a diagnostic variable of forthcoming seismic events [14]. ORIGIN 10.0 software version 2021 for Microsoft Windows was used for data processing.

4. Results and Discussion

4.1. Seismicity of Vrancea region

Regarding seismicity in Vrancea region during 1 January 2012- 1 May 2023 investigated period have been recorded hundreds of events of moment magnitude in the range of 2.0 ≤ M_w ≤ 5.7. As Figure 3 shows, during more than 11 years period have been recorded 111 earthquakes of moment magnitude M_w in the range of 4.0 ≤ M_w ≤ 5.7 on Richter scale, of which only five earthquakes recorded M_w ≥ 5.0 in the range depths of 21 km-148 km. The crustal seismicity evidenced an increase of microseismic events at the depths of 67 km- 173 km in Vrancea area (registered at seismic stations Plostina and Vrancioaia), but also an increase of crustal earthquakes number in the range of 0 – 30 km depth.

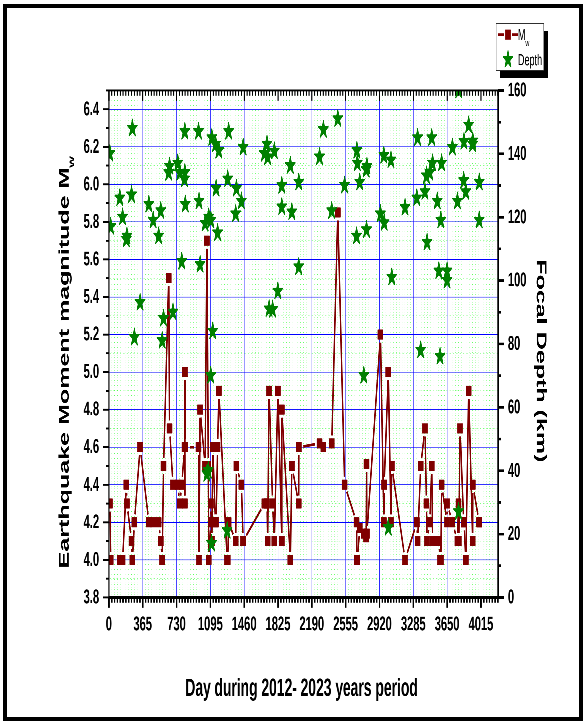

Figure 3. Seismic events recorded in Vrancea region during 2012-2023 period.

4.2. Temporal variability of radon concentration

The preparatory process of earthquakes is rich in complex features, from stochastic to chaotic or pseudo-periodic dynamics, function of the different geo-tectonic regime which is reflected in the focal mechanisms.

This study aims to establish a relationship between the intensity of radon anomaly and size of impending earthquake that may be valid for a specified source distance range from the recording site.

The average ten days mean radon concentration C_{Rn} in air above the ground measured with CR-39 detectors recorded for eleven years period in Vrancea (Plostina seismic station) was 576.81 ±78.27 Bq/m^3, and ten days C_{Rn} fluctuations was in the range of 78±22 Bq m/^{3} and 2975± 296 Bq/m^3 .The average ten days mean radon concentration C_{Rn} in air above the ground measured with CR-39 detectors recorded for the same period in Bucharest-Magurele seismic station was of 629.03 ±89.45 Bq/m^3 and ten days C_{Rn} fluctuations in the range of 78±19 Bq/m^3 and 2619± 329 Bq/m^3. The values of ^{222}Rn concentrations in air near the ground at Plostina and Vrancioaia stations in Vrancea region are in the close ranges, and have strong seasonal variations. Changes in the air near the ground radon concentrations may be associated with the changes in radon in soil regime and geological settings in the epicentral and surrounding regions. At both Vrancea test sites (situated at a distance of almost 150 km of Bucharest), and Bucharest the radon concentrations were low in the summer and high in the winter periods. For some of the recorded earthquakes of moment magnitude 4.0 ≤ M_w ≤5.7 that occurred during eleven years observation period, at seismic stations Plostina and Vrancioaia in Vrancea region, Cheia-Muntele Rosu and Bucharest was recorded the increase of radon concentration with one month - two weeks before the quakes, followed by post-quake reduction of ^{222}Rn. The time series of air near the ground radon concentration recorded from 2012 to 2016 are displayed in Figure 4 for Vrancea-Plostina and Bucharest stations. The earthquake events are also marked. A coarse look at the data suggests that the periods of radon spikes coincide with periods of frequent seismic events.

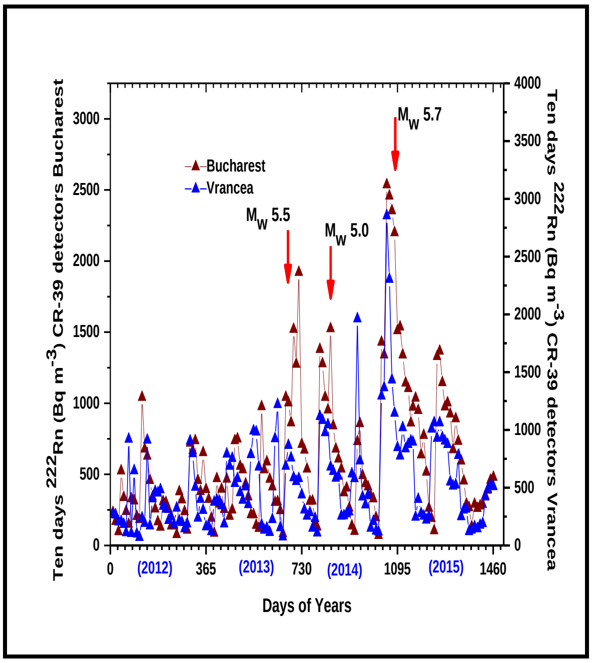

Figure 4. Sequence chart of measured air radon data in Bq/m^3 and radon spikes that coincide with periods of seismic events of M_w ≥ 5.

The duration of recorded radon anomalies is linearly related to the earthquake preparation radius and inversely to the distance from earthquake focus to the radon measuring site. This observation supports the theory that radon in air above the ground could be used as a presignal of seismic events. As the radon near the ground concentration values taken shortly after seismic events showed a sudden decrease returning to normal concentrations as before earthquakes, we concluded that these anomalies could be attributed to the seismic activity. This finding is in a good accordance with scientific literature in the world [9-12].

The relative changes of radon concentration from its average for a particular period of the year depend on the magnitude and depth of the impending earthquake, the distance of recording station from the earthquake source, and seasonal changes of meteorological parameters (temperature, pressure, humidity). Prior to identifying any radon anomalies possibly related to geodynamic processes, the effect of meteorological parameters on radon time series has been well examined. The fluctuations of atmospheric meteorological parameters were also obtained and analyzed to account for their effect on radon time series.

The rank correlation Spearman coefficient between air near the ground ^{222}Rn concentration and air temperature for the entire investigation period in Vrancea region at Plostina seismic station was found to be (r= 0.63), significant for a positive correlation, while the correlation coefficient between air near the ground ^{222}Rn concentration and air pressure was found to be a weaker positive correlation (r= 0145, p=0.0027). No statistically significant correlation was observed with rainfall (r = - 0.049, p = 0.201) and wind speed (r = - 0.129, p = 0.011). Particular attention was paid to examining the effect of rainfall on air radon time series, as striking similarities have been reported by other studies between rainfall- and earthquake-induced fluctuations in radon profile [13-19].

Prior to the seismic events the earthquake precursors appear at different distances and heights over the active seismogenic areas. The earthquake preparation area on the ground can be estimated according to the formula:

$$R = 10^{0.43 M_w}$$

where R is the possible radius of the preparation zone in km and Mw is the earthquake moment magnitude [15-16]. For Vrancea earthquake of 22 November 2014 with Mw =5.7, the corresponding calculated radius would be R ≈ 282.5 km in the latitudinal and longitudinal directions from the epicenter, for earthquake 6 October 2013, moment magnitude M_w=5.5 the corresponding calculated radius would be R ≈ 231.7 km, while for the earthquake 29 March 2014, of moment magnitude M_w=5.0 the corresponding calculated radius would be R ≈141.3 km, in the latitudinal and longitudinal directions from the epicenter (Table 1).

Table 1. Radius values for earthquake preparation zones of seismic events of M_w ≥ 5.0 recorded during analyzed period

Data	Mw	Depth (km)	R (km)
06.10.2013	5.5	134	231.7
29.03.2014	5	134	141.3
22.11.2014	5.7	39	282.5
28.10.2018	5.8	151	311.9
31.01.2020	5.2	121	172.19

As Bucharest station is located at almost 150 km of Vrancea seismic area, it seems that preparation area of a moderate/strong earthquake will start from about the same distance of Vrancea.

To get an anomaly earthquake association, Dobrovolsky's earthquake preparation zone was used, and 5 of 6 observed radon anomalies were interpreted as presignals to at least one earthquake event. Analyzing the peak profile of radon in air anomalies, precursor times ranging from 2.9 to 15.8 days and anomaly durations ranging from 0.8 to 5.9 days were obtained.

The good spatial coupling between the Rn gas anomalies and the fault zones indicated that the anomalies are intrinsically related to earthquakes and tectonic activities. This conclusion is supported by the fact that Rn gas anomalies distributed along the fault zones were found by field observations using CR-39 detectors at the two seismic stations in Vrancea zone (Plostina and Vrancioaia). The above findings were compared with worldwide data, and we found a good concordance.

In earthquake preparation, pressure gradients, associated with movement of crustal plates/subplates, drive underground ^{222}Rn gas to flow in different directions, being able to reach the surface without chemically reacting with other entities. For this reason, its concentration anomaly in air provides significant information for seismic studies, being considered as a potential long-term/short-term earthquake precursor tool [20-29]. The spatio-temporal variations in its anomalies, correlation coefficients, and their correlation analysis between the epicenter and background area may provide information on the origins of the radon gas, the physical processes of gas migration and its vertical distribution. Such analysis can be used to identify the genetic mechanisms of gas anomalies and provide the basis for studying the lithosphere-surfacesphere-atmosphere-ionosphere coupling (LCAIC) mechanism.

4. Conclusion

To systematically study the temporal variation of radon in the air near the ground concentration related to seismicity, a series of field measurements using short-term nuclear track detectors CR-39 were carried out in Vrancea active area and Bucharest. Radon in air concentration and meteorological parameters were continuously monitored for nearly eleven seasons. The long-term time series analysis of radon in active fault zones shows strong seasonal fluctuations in its levels.

The ^{222}Rn concentrations recorded at the seismic stations in Vrancea region and Bucharest vary with air temperature and pressure and hence these environmental parameters must be discriminated for identification of seismic induced anomalies. Statistical methodologies were applied for identification of the seismic induced anomalies in ^{222}Rn such that the variability of air near the ground ^{222}Rn with pressure and temperature is discriminated.

Despite technological advances in geospatial and observational seismic events monitoring and early warning systems, the ability of precisely forecasting remains a major challenge for scientific community.

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