ISSN 1330–0008
CODEN FIZAE4

RADON CHAOTIC REGIME IN THE ATMOSPHERE AND SOIL

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Dedicated to the memory of Professor Vladimir Šips

Received 22 October 2004; Revised manuscript received 8 March 2005
Accepted 4 April 2005 Online 4 December 2005

Radon concentrations were continuously measured outdoors, in a basement and in soil. The readings were made every 10 minutes and the series of records were analyzed to extract phase-space dynamical information. The application of fractal methods allowed exploration of the chaotic nature of radon in the atmosphere and soil. The computed fractal dimensions, such as the Hurst exponent (H) from the rescaled-range analysis, Lyapunov exponent (λ) and attractor dimension, provided estimates of the degree of chaotic behaviour. The obtained low values of the Hurst exponent ($0 < H < 0.5$) indicate anti-persistent behaviour (non-random changes) of the series, but the positive values of λ have pointed out a great sensitivity to initial conditions and the deterministic chaos that appeared in the variations of the radon concentrations. The calculated fractal dimensions of attractors indicate strong influence of meteorological parameters on radon in the atmosphere and soil.

PACS numbers: 87.52.-g, 92.30.Px, 05.45.Tp UDC 539.164

Keywords: radon concentration in atmosphere, deterministic chaos, fractal dimensions, meteorological parameters

1. Introduction

Radon, ^{222}Rn, is an inert radioactive gas emitted ubiquitously by soil. The exhalation rate of radon depends on the concentration of its parent (^{226}Ra) in the Earth's crust, on meteorological conditions and on properties of the soil, like porosity and water content [1]. Radon concentration in the air is influenced by indoor and outdoor climatic variations, such as temperature, barometric pressure, ventilation and rain. Atmosphere can be considered as a complex and non-linear dynamical system whose variables can be subject to a chaotic regime.

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To examine the dynamics of radon concentrations, it is necessary to continuously measure radon in the air over a period of time. The radon time series can be analyzed to determine the degree of chaotic behaviour, to predict the variations in the future using fractal methods and to investigate the correlations with meteorological parameters by comparing some of the fractal dimensions [2].

Assuming a deterministic system, the causal relationship between the radon concentration and a given controlling parameter may yield a correlation between radon concentrations measured in two adjacent time intervals, while no correlation exists in a purely stochastic system.

2. Methods

We applied fractal methods to the results of radon measurements using several approaches. To examine the fractal properties of ^{222}Rn indoor and outdoor series of readings using the Hurst's rescaled-range analysis, we calculated the Hurst exponent, H, that is used to indicate whether a time series is random or successive steps are not independent, and the Lyapunov exponent, λ, whose positive value can indicate a regime of deterministic chaos. Furthermore, we calculated the attractor fractal dimensions, such as capacity dimension (or box-counting dimension, D_b), correlation dimension (D_c) and Lyapunov dimension (or Kaplan-Yorke dimension, D_L) which is a non-integer or fractal dimension of attractor belonging to a chaotic behaviour.

The rescaled-range analysis examines a time-dependent variable of a physical dynamic system by studying its time series, average values over sampling intervals, accumulation of the differences between a value and the average, as well as the differences between the maximum and minimum of the accumulation, or the range. Fitting the logarithm of the relative range (the range divided by the standard deviation) against the logarithm of time yields the slope that is the Hurst exponent H. The Hurst exponent lies in the region 0 to 1. If its value is close to 0.5, the time series indicates random walk, i.e., that successive steps are independent. When $0.5 < H < 1$, the recorded values show a persistent behaviour, i.e., the trend over the observed interval will continue. In that case, if the time series for some time in the past had a positive increment, then on the average it will also show an increase in the future. Conversely, a decreasing trend in the past implies on the average a continued decrease in the future. When $H < 0.5$, the time series has an anti-persistent behaviour, i.e., the observed trend will reverse. In that case, an increasing trend in the past implies a decreasing trend in the future, and a decreasing trend in the past indicates a probable increasing trend in the future [3].

To be precise, the above quantifications come from the correlation coefficient, ρ, of the successive increments in the following form: $\rho = 2^{2H-1} - 1$. If $H < 0.5$, the correlation function is negative and we have anti-persistence. If $H = 0.5$, there is no correlation and we have a random walk or an uncorrelated white noise. If $H > 0.5$, a positive correlation is present and the respective time series exhibits persistence. Since H is a statistical parameter, analysis of large sets of at least 2500 observations is required.

The Lyapunov exponent, λ, is a measure of the rate at which nearby trajectories in phase space diverge. If two nearby trajectories start off with a separation d_0 at time $t = 0$, then the trajectories diverge so that their separation at time t, $d(t)$, satisfies the expression: $d(t) = d_0 e^{\lambda t}$ (as a matter of fact, the value of λ is calculated as the limes of logarithms, or by a computer program). If λ is positive, then we say the behaviour is chaotic [4]. Generally, there are as many Lyapunov exponents as there are dynamical equations. For periodic trajectories, all Lyapunov exponents are negative. The larger is the positive exponent, the more chaotic is the system. i.e., the shorter is the time scale of the system's predictability. The sum of positive Lyapunov exponents is called entropy; its reciprocal is roughly the time over which a meaningful prediction is possible.

A second method of quantifying chaos is focusing on the geometrical aspects of the attractors; if the attractor has an non-integer or fractal dimension, the dynamical system has characteristics of a chaotic regime. The dimensionality is also important in determining the range of possible dynamical behaviour, namely, the dimensionality of the attractor, D, must be less than that of d of the full phase space; d is given by the minimum number of variables needed to describe the state of the system.

The embedding-space method allows extracting a multi-dimensional description of the phase-space dynamics from the time series data of a single dynamical variable. If the embedding space is generated properly, the behaviour of trajectories in this embedding space will have the same geometrical and dynamical properties that characterize the actual trajectories in the full multi-dimensional phase space of the system. The embedding procedure uses a recorded series of values X for some dynamical system and forms the series of values X_1, X_2, ...,X_d, so the embedding dimension, d, is the dimension of the embedding space [4].

A way of determining the embedding dimension is given by the false-nearest-neighbors (FNN) method which chooses the minimum embedding dimension of a one-dimensional time series. This method finds the nearest neighbor of every point in a given dimension, then checks whether these points are still close neighbors in the next higher dimension. The percentage of FNN should drop to a minimum, or zero, when the appropriate embedding dimension has been reached.

According the Takens theorem, d is a little larger than $2D$, $2D < d < 2D + 1$.

What we do in practice is to compute D (e.g. the so-called correlation dimension, D_c) for $d = 1, 2, 3, \ldots$ and plot the values of D as a function of d. We expect D to vary with d until d is equal to or becomes greater than about twice the dimension of the phase space attractor for the system. So we get the value of d; the known value of d tells us about a number of physical variables of the dynamical system (though we have only observed the time series of one variable).

Among many different definitions of dimensionality of an attractor, we use the tree ones and we try to compare them, and find out how close are their numerical values.

The capacity dimension of a geometrical object is determined as follows: $D_\mathrm{b} = -\lim_{R\to 0}(\log N/\log R)$, where N is the number of boxes needed to contain all points of the geometric object, and R is the side length of the box (e.g., for a point, the geometrical object in two-dimensional space, the box, is just a square of side R; $N = 1$, $D_\mathrm{b} = 0$).

The correlation dimension is determined as follows: $D_\mathrm{c} = -\lim_{R\to 0}(\log C/\log r)$, where C is the probability of finding two points in the same sphere of radius r (the probability is determined using the number of points in the sphere).

The Lyapunov dimension, D_L, can be determined from the calculated Lyapunov exponents, λ_i (the number of Lyapunov exponents corresponds to the dimension of the phase space), in the following way: $D_\mathrm{L} = p + (1/|\lambda_{p+1}|)\sum_{i=1}^{p}\lambda_i$, where p is the largest integer number according the expression, $\sum_{i=1}^{p}\lambda_i \geq 0$, $\lambda_1 > \lambda_2 > \lambda_3 > \ldots \lambda_n$.

It can be shown that $D_\mathrm{c} \leq D_\mathrm{L} \leq D_\mathrm{b} \leq d$ [5,4].

In calculating the described fractal dimensions, we used the programs of the Chaos Data Analyzer (CDA) for calculating H, λ, D_b and D_c, Dataplore (DP) for calculating D_L and Visual Recurrence Analysis (VRA) for calculating d [6].

3. Experiments, results and discussion

Radon concentrations were measured with an Alpha Guard PQ 2000 detector (Genitron Instrument, Germany), outdoors (O), indoors (I) and in the soil (S) at a location in Valpovo (a town near Osijek). The sites O and S were positioned outside in the garden, 1 m above the ground and 0.8 m deep in the soil, respectively, while the site I was in a closed basement room of a single house. Simultaneously, the atmospheric temperature and pressure were measured.

Considerable difficulties were encountered in the radon measurements in the soil; the Alpha Guard measuring procedure, according to the instructions, envisages the burying of a narrow tube into the soil and pumping of the soil gas into the ionization chamber in order to measure the radon concentration. However, the soil material at the point of measurement was rather compact and because of the very low permeability, it was not possible to pump the soil gas into the measuring chamber. Therefore, we secured a cylinder-shaped hole of a diameter of 0.3 m and 0.2 m deep from which the soil gas was pumped through a tube.

At each site, the Alpha Guard detector was set to authomatically read and make a record of the number of counts every 10 minutes. Many readings of the radon concentration were carried out: in June (site O; 2835 readings), in July (site I; 3679) and in September (site S; 2967), in the year 2004. The mean radon concentrations of 16.4 Bq/m^3, 66.7 Bq/m^3 and 25.01 kBq/m^3, with the standard deviations of 11.9 Bq/m^3, 36.1 Bq/m^3 and 3.13 kBq/m^3, respectively, were obtained.

The time variations of the radon concentration in the soil (site S) and the barometric pressure, are presented in Fig. 1. In addition to the daily periodicity of the radon concentration, that can also be observed in variations of the barometric pressure, one can see irregularities in the radon concentration visible as large peaks and deep valleys. We examined whether the radon concentrations are chaotic, or their changes are random and they behave as a noise.

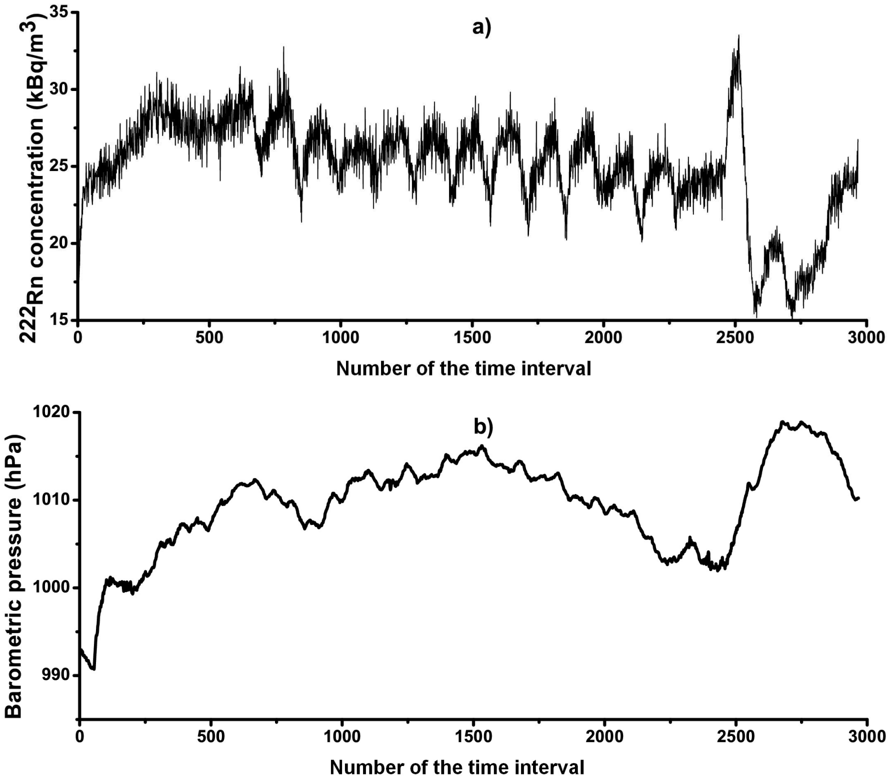

Fig. 1. Variation of the radon concentration in the soil versus the number of 10 min *intervals (a), as well as variations of the barometric pressure (b).*

A particular radon signal appeared around the 2500^{th} reading at the site S that we previously interpreted as a radon anomaly when searching for earthquake precursors [7]. The interpretation of radon anomalies as precursors of seismic activity has some uncertainties. One of them is that the time variation of radon concentration can give false signals due to the deterministic chaos [8]. So, we undertook the task to examine the origin of the registered radon signal.

The radon signal, after the peak of 33.53 kBq/m^3, had a minimum that typically corresponded to the shape of a radon anomaly preceding an earthquake coming in about three weeks. The maximum reading of radon concentration shows a deviation from the mean value of more than two standard deviations [$(33.53-25.01)$ kBq/m^3 $= 8.52$ kBq/m^3, while two standard deviations amount to 2×3.13 kBq/m^3 $= 6.26$ kBq/m^3)] [7].

Studies of the radon concentration in soil have shown a barometric effect. Namely, a negative correlation between the radon concentration in the soil and the barometric pressure has been found. They may be seen as changes of the radon concentration and atmospheric pressure presented in Figs. 1a and 1b about the 2500^{th} reading. The decrease in barometric pressure corresponds to an increase in the radon concentration, and conversely [7].

We considered the time variation of radon concentration in the soil (Fig. 1a) and calculated the embedding dimension by means of the VRA program and the false nearest neighbors (FNN) method that clearly showed the dimension of 3 (Fig. 2, the minimum position of the curve at 49 % of false neighbors); otherwise the FNN calculations for the uniform white noise gave nearly 50 % of false neighbors for all embedding dimensions (i.e. the respective FNN calculation does not show any minimum).

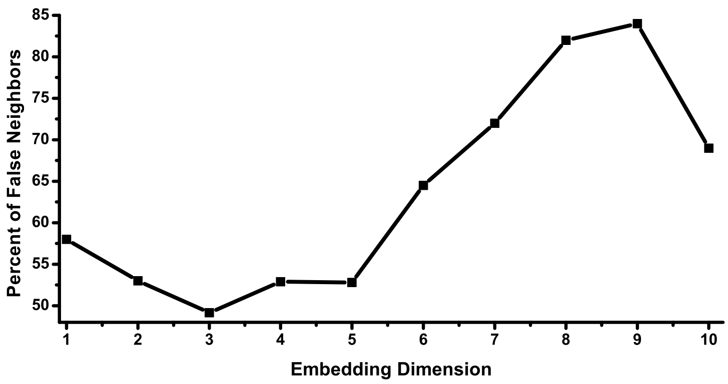

Fig. 2. The FNN calculation for the radon variation in the soil: percentage of false neighbors versus embedding dimension.

There is also another approach to the analysis of time series of a number of equidistant points, separated by an interval of time, the so-called power spectrum (mean square amplitude as a function of frequency), that can refer to the chaotic behaviour of given data. With the CDA program, we obtained the cumulative periodogram, which was the integral of the power spectrum over frequency, presented in Fig. 3; the integral followed the 45° line for the white noise, but in the case of the radon anomaly observed at the site S (radon in soil, Fig. 1a), large deviations were found.

The application of the fractal methods using the CDA program to the three series of measurements of radon concentration (at the sites O, I and S) and to the readings of barometric pressure and temperature gave the results presented in Table 1 for the Hurst (H) and Lyapunov exponent (λ in 10 minute time steps), and capacity (D_b), correlation (D_c), Lyapunov dimension (D_L), while the values of embedding dimension (d) were calculated using by the VRA program. The values of D_L were calculated for 1 h intervals, because the DP program did not give reliable results for data of 10 min intervals, because the differences between the neighboring data were to small.

From Table 1, we can see that for radon λ is positive at all measurement sites, thus pointing at a chaotic regime of the radon concentration in the atmosphere and

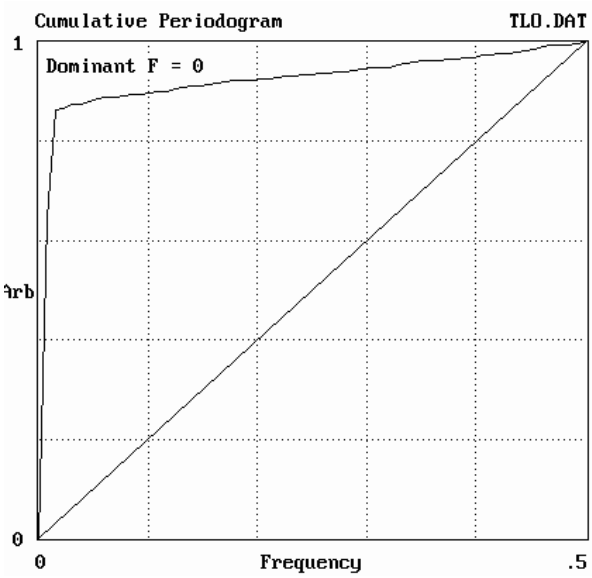

Fig. 3. The curve of cumulative periodogram (integral of the power spectrum over frequency) for time series of the radon in the soil; the 45° line belongs to the white noise.

TABLE 1. The values of the Hurst exponent (H), Lyapunov exponent (λ, for 10 min *time steps), capacity dimension (D_b), correlation dimension (D_c), Lyapunov dimension (D_L) and embedding dimension (d), calculated for radon, barometric pressure and air temperature, at the sites O (air outdoors), I (indoors) and S (in the soil outdoors), for N data, measurements in 10* min *intervals; the D_L values were obtained for $N/6$ data, i.e., for measurements in 1* h *intervals.*

Radon							
Site	N	H	λ	D_b	D_c	D_L	d
O	2835	0.13	0.63 ± 0.03	1.92 ± 0.25	4.59 ± 0.08	1.73	4
I	3679	0.12	0.75 ± 0.03	2.64 ± 0.30	3.97 ± 0.15	1.43	8
S/O	2967	0.20	0.57 ± 0.03	1.45 ± 0.19	4.43 ± 0.01	1.15	3
Barometric pressure							
Site	N	H	λ	D_b	D_c	D_L	d
O	2835	0.67	0.15 ± 0.03	1.38 ± 0.18	3.13 ± 0.55	1.04	13
I	3679	0.70	0.13 ± 0.02	1.27 ± 0.15	2.56 ± 0.64	1.32	8
S/O	2967	0.71	0.08 ± 0.02	0.37 ± 0.05	2.45 ± 0.63	1.08	9
Temperature							
Site	N	H	λ	D_b	D_c	D_L	d
O	2835	0.88	0.14 ± 0.04	1.32 ± 0.17	2.53 ± 0.95	1.27	15
I	3679	0.50	0.18 ± 0.09	1.38 ± 0.16	4.27 ± 0.27	2.64	10
S/O	2967	0.72	0.12 ± 0.03	0.92 ± 0.12	2.20 ± 0.25	1.38	9

soil gas, though with a slightly lower grade of the deterministic chaos in the soil; that was expected considering the influence of meteorological parameters on the radon in the atmosphere. The Lyapunov exponent of radon was a little higher indoors than outdoors, what suggests that the atmosphere was less stable in July than in June (during the radon measurement at the site O); that indicate also the standard deviations of the barometric pressure: 2.7 mbar (about the mean of 1005.2 mbar) and 3.4 mbar (about the mean of 1004.5 mbar) for the sites O (June) and I (July), respectively.

The Hurst exponent for radon was small and less than 0.5, indicating anti-persistent behaviour, i.e. an increasing trend in the past implied a probable decreasing trend in the future, and conversely (Fig. 4). Different values of the Hurst exponent for radon ($H < 0.5$) were obtained by Paush et al. [2], Bossew and Lettner ($H > 0.5$) [9] and by Bejar et al. ($H > 0.5$) [8].

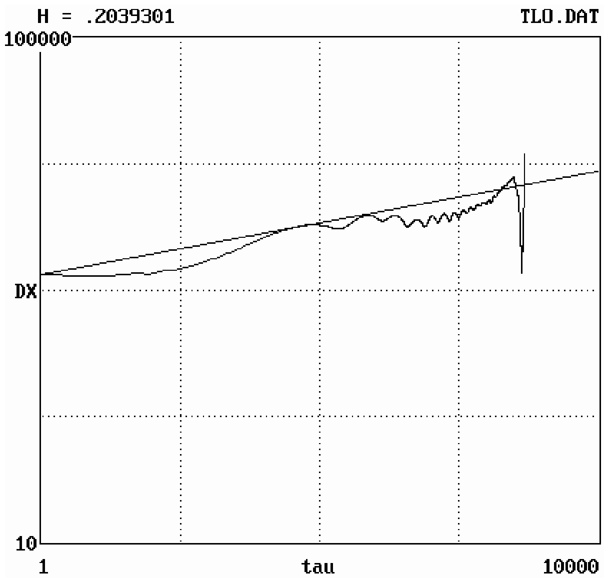

Fig. 4. The calculated values of the Hurst exponent for the time series of radon in the soil.

When H is determined, one can calculate an attractor's fractal dimension from $D = 2H$. If we assume the values of H were correctly obtained, then the calculated values of D_b and D_L dimensions for the outdoor radon, site O, and soil radon, site S, nearly satisfy the quoted equation. The D_b value for radon at the site I (2.64) is also fractal and low-dimensional, what is in accordance with the known investigation of the atmosphere, which was chaotic within a shallow layer of a few meters above the ground [1]. Similarly, one can comment the low-dimensional values of D_L for radon, while the D_c values are rather high and we did not consider them (they were not in accordance with the above mentioned low-dimensional values).

The obtained values of the embedding dimension for radon (d, Table 1) are mainly in accordance with the above relationship, $2D < d < 2D+1$, for the D_b and D_L values. Considering the relation $D_c \leq D_L \leq D_b \leq d$, our experimental data and computing methods of attractor's fractal dimension have mainly shown accordance with this relation (except for the already mentioned D_c, but the inequality $D_c < d$ is still fulfilled).

According to the attractor's non-integer or fractal dimension, as well as the embedding dimension (d) for radon in the outside atmosphere (O), the lower bound on the number of independent variables, required to describe the time evolution of the outdoor radon concentration, is 4; for radon in the soil ($d = 3$), the number of required variables is 3.

Of course, in the experiments with radon, the variables were meteorological parameters, like barometric pressure, temperature, rainfall, wind or ventilation.

From Table 1, the parameters for the fractal analysis of barometric pressure and temperature also show a chaotic behaviour, as for the radon series, although the positive λ values were lower than the ones for radon; it tells us of a lower grade of deterministic chaos of temperature and pressure; also the Hurst exponent ($H = 0.5$) indicates a random walk of data of the temperature series in the basement (site I).

The Lyapunov and capacity dimensions for pressure and temperature have similar values like the radon concentration (Table 1), indicating a correlation between the time variation of radon concentration, barometric pressure and temperature [2]; e.g., the statistical correlation, with a negative correlation coefficient, was found between radon concentration in soil and barometric pressure [7]; namely, similar D_L values, as well as the D_b values, indicate the same chaotic way of the mentioned variables. Of course, there were some discrepancies of other fractal parameters, like the D_c and d, but that may also show that some applied algorithms were not quite good. The high values of the Hurst exponent ($0.5 < H < 1$) for pressure and temperature indicate a persistent behaviour, i.e., an increasing trend in the past implied on the average a continued increase in the future, and conversely for a decreasing trend.

A dynamic dissipative and non-linear system becomes chaotic because of its sensitivity to initial conditions and limited precision of the measurement. Therefore, the trajectories of chaotic systems can only be predicted for relatively short time scales.

The sum of the positive Lyapunov exponents (base e) is called entropy; its reciprocal value is roughly the time over which a meaningful prediction is possible (the so-called Lyapunov time). Since the program gave positive values of λ for all series of measurements (presented in Table 1), the Lyapunov time, $1/\lambda$ was, e.g., in the basement (site I) $1/(0.75/6)$ h = 8 h for the radon concentration and $1/(0.18/6)$ h = 33.3 h for the temperature.

Because a significant earthquake (with a magnitude $M > 2.5$) was not detected in the region of 300 km around the site of experiment (Osijek) (the seismic data of the Seismological Stations of the Geophysics Department of the University of Zagreb) five weeks after the appearance the radon signal shown in Fig. 1a, we concluded that the radon signal observed in the soil was not related to a tectonic activity. Hence, the chaotic regime of the radon time variation and the changes of barometric pressure (the barometric effect) could be possible causes of the radon signal.

Another our measurement with the Barasol device (radon silicon detector, made by Algade, France) was running at the same soil site (S) and the same time with the Alpha Guard detector. The radon time variation, registered by the Barasol, also showed a radon signal at the 2500^{th} reading, and we concluded that the origin of the radon signal was the large change of barometric pressure. Namely, it is improbable that the radon signal, caused by the deterministic chaos, appeared in two independent devices at the same time.

4. *Conclusion*

The application of the fractal methods to three radon series of measurements (outdoors, indoors and in the soil), and to the barometric pressure and temperature yielded the values of the Hurst (H) and Lyapunov exponent (λ), and of the capacity (D_b), correlation (D_c), Lyapunov (D_L) and embedding dimension (d).

The positive values of λ for all series of measurements of radon concentration, barometric pressure and temperature indicated their chaotic behaviour. The radon measurements showed higher values of λ (and higher grade of deterministic chaos) than the measurements of pressure and temperature. The Hurst exponent ($H = 0.5$) indicates random walk of data of the temperature time series in the basement (site I), what indicates the absence of deterministic chaos.

The Hurst exponent for radon is small and less than 0.5, indicating anti-persistent behaviour, i.e. an increasing trend in the past implied a probable decreasing trend in the future, and conversely; this behaviour was most expressed for the indoor radon, that had the smallest value of H (0.12). On the other hand, the high values of the Hurst exponent ($0.5 < H < 1$) for pressure and temperature indicated persistent behaviour, i.e., an increasing trend in the past implied on the average a continued increase in the future, and conversely for decreasing trend.

According to the attractor's non-integer or fractal dimension and the embedding dimension (d) for radon in the outside atmosphere (O), the lower bound on the number of independent variables required to describe the time evolution of the outdoor radon concentration is 4; for radon in the soil ($d = 3$), the number of required variables is 3.

Simultaneously with the measurements of radon concentration, the meteorological parameters barometric pressure, temperature, rainfall, wind and ventilation were also recorded.

The Lyapunov and capacity dimensions of pressure and temperature had similar values as for the radon concentration, indicating a multiple correlation between the time variation of radon, barometric pressure and temperature; namely, similar values of D_L and D_b indicate the same chaotic behaviour.

The Lyapunov time, for which a meaningful prediction was possible only in the basement, was 8 h and 33.3 h for the series of radon measurements and temperature, respectively.

The particular radon signal in the soil, that appeared about the 2500$^{\text{th}}$ reading, is explained as an effect of changes of the barometric pressure.

Acknowledgements

This work was supported by Croatian Ministry of Science.

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KAOTIČNI REŽIM ZA RADON U ATMOSFERI I TLU

Mjerili smo koncentracije radona u otvorenoj atmosferi, u podrumu i u tlu, neprekidno tijekom mjesec dana u 10-minutnim intervalima. Radonske vremenske nizove analizirali smo usporedbom algoritama kako bi se dobile dinamičke informacije o faznom prostoru. Primjena fraktalnih metoda omogućila je ispitivanje kaotične prirode radona u atmosferi i tlu. Izračunate fraktalne dimenzije, kao što su Hurstov eksponent (H), Ljapunovljev eksponent (λ) i dimenzija atraktora, omogućile su procjenu stupnja kaotičnosti. Dobivene niske vrijednosti Hurstovog eksponenta ($0 < H < 0,5$) ukazale su na antiperzistentno ponašanje vremenskih nizova (nenasumičnost promjena), dok su pozitivne vrijednosti pokazale veliku osjetljivost na početne uvjete i ukazale na deterministički kaos koji se pojavljuje u vremenskim varijacijama radona. Izračunate fraktalne dimenzije atraktora ukazale su na više (meteoroloških) parametara koji su utjecali na razine radona u atmosferi i u zemnom plinu.