Nonlinear chaos-dynamical approach to analysis of atmospheric radon ^{222}Rn concentration time series

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Abstract. We present the theoretical foundations of an effective universal complex chaos-dynamical approach to the analysis and prediction of atmospheric radon ^{222}Rn concentration using the beta particle activity data of radon monitors (with a pair of Geiger–Muller counters). The approach presented consistently includes a number of new or improved available methods (correlation integral, fractal analysis, algorithms of average mutual information and false nearest neighbors, Lyapunov's exponents, surrogate data, nonlinear prediction schemes, spectral methods, etc.) of modeling and analysis of atmospheric radon ^{222}Rn concentration time series. We first present the data on the topological and dynamical invariants for the time series of the ^{222}Rn concentration. Using the data measurements of the radon concentration time series at SMEAR II station of the Finnish Meteorological Institute, we found the elements of deterministic chaos.

Keywords. Chaotic dynamics; time series of the ^{222}Rn concentration; universal complex chaos-dynamical approach; analysis and prediction.

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1. Introduction

At the present time, studying regular and chaotic dynamics of nonlinear processes in different classes of geophysical, chemical, physical and other systems is of great theoretical and practical interest because of its very important roles in both fundamental science and applied technologies [1–36]. The importance of studying the phenomenon of stochasticity or chaos in dynamical systems is due to a number of applications, including the necessity of understanding chaotic features in different geophysical (hydrometeorological, environmental, etc.) systems. New fields of investigations of these systems have been made possible by development of chaos and dynamical systems theory methods [1–15]. In our previous papers [11–34] we presented some results and review of new methods and algorithms for the analysis of different systems of environmental and earth sciences, quantum (atomic and molecular) physics, electronics and photonics. The nonlinear methods of chaos theory and recurrence spectra formalism have been applied to study the stochastic and chaotic elements

in the dynamics of hydrometeorological, environmental and physical (namely, atomic, molecular, nuclear systems in free state and in an external electromagnetic field) systems. These studies allow to discover unusual manifestations of chaos phenomenon. The studies concerning nonlinear behavior in the time series of atmospheric constituent concentrations are sparse, and their outcomes are ambiguous. In Ref. [35] the chaotic elements in the O_3 concentration time series for the Cincinnati (Ohio) and Istanbul regions have been found. Analysis of the NO_2, CO, O_3 concentration time series has been presented in Ref. [36]. The detailed analysis of the NO_2, CO, CO_2 concentration time series in the Gdansk region (Polland) has been presented in Ref. [22]. This analysis found the elements of a deterministic chaos in the corresponding time series. Moreover, it has been shown that even though a simple procedure is used to construct the nonlinear prediction model, the results are quite satisfactory. These studies show that the methodology of chaos theory can be applied and the short-range forecast by the nonlinear prediction method can be satisfactory. The time

series of concentrations are however not always chaotic, and chaotic behavior must be examined for each time series.

In this paper we present the theoretical foundations of an effective universal complex chaos-dynamical approach to the analysis and prediction of the atmospheric radon ^{222}Rn concentration changes. We also present the results of analysis of the atmospheric radon ^{222}Rn concentration time series using the beta particle activity data of radon monitors (with a pair of Geiger–Muller counters) at SMEAR II station of the Finnish Meteorological Institute [37]. The approach used consistently includes a number of new or improved methods of analysis: correlation integral, fractal analysis methods, algorithms of average mutual information and false nearest neighbors, the Lyapunov's exponents analysis, surrogate data, nonlinear prediction schemes, spectral methods, etc. (see details in Refs [11–34]). Data on the topological and dynamical invariants for the studied time series of the ^{222}Rn concentration are presented and show evidence of deterministic chaos.

2. Universal chaos-dynamical approach in analysis of chaotic dynamics of the radon concentration time series

As many ideas of the present approach have been developed earlier and need only to be reformulated in regard to the problem studied in this paper, we only need the key finding of Refs [11–18,22–25]. Let us formally consider scalar measurements $s(n) = s(t_0 + n\Delta t) = s(n)$, where t_0 is the start time, Δt is the time step, and n is the number of measurements. Further it is necessary to reconstruct phase space using, as well as possible, information contained in the $s(n)$. Such a reconstruction results in a certain set of d-dimensional vectors $y(n)$ replacing the scalar measurements. Packard *et al.* [1] introduced the method of using time-delay coordinates to reconstruct the phase space of an observed dynamical system. The direct use of the lagged variables $s(n + \tau)$, where τ is some integer to be determined, results in a coordinate system in which the structure of orbits in phase space can be captured. Then using a collection of time lags to create a vector in d dimensions,

$$y(n) = [s(n), s(n+\tau), s(n+2\tau), \ldots, s(n+(d-1)\tau)], \quad (1)$$

the required coordinates are obtained. In a nonlinear system, the $s(n + j\tau)$ are some unknown nonlinear combination of the actual physical variables that comprise the source of the measurements. The dimension d is called the embedding dimension, d_E. In order to

perform the subsequent reconstruction of phase space, it is very important to choose a proper time lag. If τ is chosen too small, then the coordinates $s(n + j\tau)$ and $s(n + (j + 1)\tau)$ are so close to each other in numerical value that they cannot be distinguished from each other. Similarly, if τ is too large, then $s(n + j\tau)$ and $s(n + (j + 1)\tau)$ are completely independent of each other in a statistical sense. Also, if τ is too small or too large, then the correlation dimension of the attractor can be under- or over-estimated respectively [3]. It is therefore necessary to choose some intermediate (and more appropriate) position between the above cases. The first approach is to compute the linear autocorrelation function

$$C_L(\delta) = \frac{\frac{1}{N}\sum\limits_{m=1}^{N}[s(m + \delta) - \overline{s}][s(m) - \overline{s}]}{\frac{1}{N}\sum\limits_{m=1}^{N}[s(m) - \overline{s}]^2}, \quad (2)$$

$$\overline{s} = \frac{1}{N}\sum\limits_{m=1}^{N}[s(m)]$$

and to look for that time lag where $C_L(\delta)$ first passes through zero. This gives a good hint for the choice of τ such that $s(n + j\tau)$ and $s(n + (j + 1)\tau)$ are linearly independent. However, a linear independence of two variables does not mean that these variables are nonlinearly independent since a nonlinear relationship can differ from a linear one. It is therefore preferable to utilize an approach with nonlinear independence, e.g. the average mutual information. Briefly, the concept of mutual information can be described as follows. Let there be two systems, A and B, with measurements a_i and b_k. The amount one learns about a measurement of a_i from measurement of b_k is determined within information theory [3,8,9]

$$I_{AB}(a_i, b_k) = log_2\left(\frac{P_{AB}(a_i, b_k)}{P_A(a_i)P_B(b_k)}\right), \quad (3)$$

where the probability of observing a out of the set of all A is $P_A(a_i)$, and the probability of finding b in a measurement B is $P_B(b_i)$, and the joint probability of the measurement of a and b is $P_{AB}(a_i, b_k)$. The mutual information I of two measurements a_i and b_k is symmetric and non-negative, and equals zero only if the systems are independent. The average mutual information between any value a_i from system A and b_k from B is the average over all possible measurements of $I_{AB}(a_i, b_k)$,

$$I_{AB}(\tau) = \sum_{a_i, b_k} P_{AB}(a_i, b_k) I_{AB}(a_i, b_k). \tag{4}$$

To place this definition to the context of observations from a certain physical system, let us think of the sets of measurements $s(n)$ as the A set and of the measurements a time lag τ later, $s(n + \tau)$, as the B set. The average mutual information between observations at n and $n + \tau$ is then

$$I_{AB}(\tau) = \sum_{a_i, b_k} P_{AB}(a_i, b_k) I_{AB}(a_i, b_k). \tag{5}$$

Now we have to decide what property of $I(\tau)$ we should select, in order to establish which among the various values of τ we should use in making the data vectors $y(n)$. One can recall that the autocorrelation function and average mutual information can be considered as analogues of the linear redundancy and general redundancy, respectively, which was applied in the test for nonlinearity. The general redundancies detect all dependences in the time series, while the linear redundancies are sensitive only to linear structures. Further, it can be concluded that the nonlinear nature of the process possibly results in chaotic growth in the concentration level. The goal of determining the embedding dimension is to reconstruct a Euclidean space R^d large enough so that the set of points d_A can be unfolded without ambiguity. In accordance with the embedding theorem, the embedding dimension d_E must be greater than, or at least equal to, a dimension of attractor, d_A, i.e. $d_E > d_A$. However, two problems arise with working in dimensions larger than really required by the data and time-delay embedding [1–20]. First, many of the computations for extracting interesting properties from the data require searches and other operations in R^d whose computational cost rises exponentially with d. Second, but more significant from the physical point of view, in the presence of noise or other high-dimensional contamination of the observations, the extra dimensions are not populated by dynamics, already captured by a smaller dimension, but entirely by the contaminating signal [3,11,22]. Further it is necessary to determine the dimension d_A.

There are several standard approaches to reconstruct the attractor dimension (see, e.g., [1–11]), but let us consider in this study two methods only. The correlation integral analysis is one of the widely used techniques to investigate the signatures of chaos in a time series. The analysis uses the correlation integral, $C(r)$, to distinguish between chaotic and stochastic systems. To compute the correlation integral, the algorithm of Grassberger and Procaccia [5] is the most commonly used approach. According to this algorithm, the correlation integral is

$$C(r) = \lim_{N \to \infty} \frac{2}{N(n-1)} \sum_{i,j_1 < i < j < N} H(r - ||y_i - y_j||), \tag{6}$$

where H is the Heaviside step function with $H(u) = 1$ for $u > 0$ and $H(u) = 0$ for $u < 0$, r is the radius of the sphere centered on y_i or y_j, and N is the number of data measurements. If the time series is characterized by an attractor, then the integral $C(r)$ is related to the radius r given by

$$d = \lim_{r \to 0, N \to \infty} \frac{\log C(r)}{\log r}, \tag{7}$$

where d is the correlation exponent that can be determined as the slope of the line in the coordinates $\log C(r)$ versus $\log r$ by a least-squares fit of a straight line over a certain range of r, called the scaling region. If the correlation exponent attains saturation with an increase in the embedding dimension, then the system is generally considered to exhibit chaotic dynamics. The saturation value of the correlation exponent is defined as the correlation dimension, d_2, of the attractor. The method of surrogate data is an approach that makes use of the substitute data generated in accordance to the probabilistic structure underlying the original data (see [2,11]). Often, a significant difference in the estimates of the correlation exponents, between the original and surrogate data sets, can be observed. In the case of the original data, a saturation of the correlation exponent is observed after a certain embedding dimension value (i.e., 6), whereas the correlation exponents computed for the surrogate data sets continue increasing with increasing embedding dimension.

It is worth considering another method for determining d_E which comes from asking the basic question addressed in the embedding theorem: one eliminated false-crossing of the orbit with itself which arose by virtue of having projected the attractor into a too low-dimensional space? By examining this question in dimension one, then dimension two, etc., until there are no incorrect or false neighbors remaining, one should be able to establish, from geometrical consideration alone, a value for the necessary embedding dimension. The advanced version is presented in Refs [16–18].

The Lyapunov's exponents are the dynamical invariants of the nonlinear system. In a general case, the orbits of chaotic attractors are unpredictable, but there is the limited predictability of chaotic physical system which is defined by the global and local Lyapunov's

exponents. A negative exponent indicates a local average rate of contraction while a positive value indicates a local average rate of expansion. In a chaos theory, the spectrum of Lyapunov's exponents is considered as a measure of the effect of perturbing the initial conditions of a dynamical system. In fact, if one manages to derive the whole spectrum of the Lyapunov's exponents, then other invariants of the system, i.e., Kolmogorov entropy and the attractor's dimension, can be found. The inverse of the Kolmogorov entropy is equal to an average predictability. An estimate of the dimension of the attractor is provided by the Kaplan and Yorke conjecture:

$$d_L = j + \frac{\sum_{\alpha=1}^{j} \lambda_\alpha}{|\lambda_{j+1}|}, \qquad (8)$$

where j is such that $\sum_{a=1}^{j} \lambda_a > 0$ and $\sum_{a=1}^{j+1} \lambda_a < 0$ and the Lyapunov's exponents λ_α are taken in descending order. There are a few approaches to computing the Lyapunov's exponents. One of them computes the whole spectrum and is based on the Jacobi matrix of the system. In the case where only observations are given and the system function is unknown, the matrix has to be estimated from the data. In this case, all the suggested methods approximate the matrix by fitting a local map to a sufficient number of nearby points. To calculate the spectrum of the Lyapunov's exponents from the amplitude level data, one could determine the time delay τ and embed the data in the four-dimensional space. In this point it is very important to determine the Kaplan–Yorke dimension and compare it with the correlation dimension, defined by the Grassberger–Procaccia algorithm [5]. The estimations of the Kolmogorov entropy and average predictability can further show a limit, up to which the amplitude level data can be predicted on average. Other details can be found in Refs [5–22].

3. Data on chaotic elements in time series of the radon concentration and conclusion

The concentration of atmospheric radon ^{222}Rn was determined by measuring the activity of beta particles in atmospheric aerosol using radon monitors. Measurements of the radon concentrations at SMEAR II station (61°51′N, 24°17′E, 181 m above sea level; near the Hyytil, Southern Finland) was done by a group of experts of the Finnish Meteorological Institute (FMI) and was actually integrated into the system long-term measurements (see details in Refs [37] and [38–41] too). The continuous measurement was performed

during 2000–2006 on the seven heights (from 4.2 m to 127 m). Technologically, for the detection of beta particles, a device with a pair of the Geiger–Muller counters, arranged in the lead corymbs, was used. Registration of the beta particles was cumulatively carried out in 10-minute intervals. The effectiveness of a detection was 0.96 percent and 4.3 percent for beta radiation from ^{214}Pb and ^{214}Bi respectively. Estimate of the 1-σ counting statistics is ± 20 percent for a presumed stable a ^{222}Rn concentration of 1 Bq/m^3 [37]. The mean daily values of atmospheric ^{222}Rn concentrations were in the range from 0.1 to 11 Bq/m^3. In fact, the lower limit of this range was provided by a hardware detection limit of the radon monitors. The corresponding data meet the log-normal distribution with a geometric mean of 2.5 Bq/m^3 (a standard geometric deviation of 1.7 Bq/m^3). The average geometric value for the daily average radon concentrations amounted to 2.3 to 2.6 Bq/m^3 per year. In general, during 2000–2006 both hourly and daily values of a parameter, which corresponds to the radon concentration, ranged from about 1 to 5 Bq/m^3.

In figure 1 is presented the typical time-dependent curve of the radon concentration, received at SMEAR II station [37].

The resulting Kaplan–Yorke dimension is very close to the correlation dimension, which is determined by the algorithm of Grassberger and Procaccia. Moreover, the Kaplan–Yorke dimension is smaller than the dimension of embedding, which confirms the correctness of the choice of the latter. In table 1 we list the results of computing different dynamical and topological invariants and parameters (time delay τ, correlation dimension (d_2), embedding space dimension (d_E), Lyapunov's exponents (λ_i), Kolmogorov entropy (K_{ent}), Kaplan–Yorke dimension (d_L), the predictability limit (Pr_{max}) and chaos indicator (K_{ch})) for radon concentration time series (2001).

Therefore, using the uniform chaos-dynamical approach we have carried out modeling and analysis of
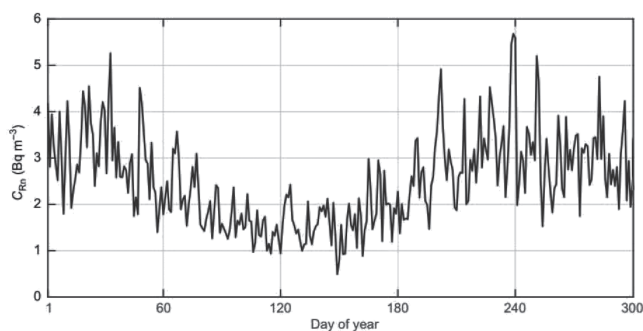

Figure 1. The time-dependent curve of the atmospheric radon concentration (see text).

Table 1. Time delay τ, correlation dimension (d_2), embedding space dimension (d_E), Lyapunov's exponents (λ_i), Kolmogorov entropy (K_{ent}), Kaplan-York dimension (d_L), the predictability limit (Pr_{max}) and chaos indicator (K_{ch}) for radon concentration time series (2001).

Year	τ	d_2	d_E
2001	12	5,48	6
Year	λ_1	λ_2	K_{ent}
2001	0,0182	0,0058	0,024
Year	d_L	Pr_{max}	K_{ch}
2001	5,36	42	0,80

the atmospheric radon ^{222}Rn concentration time series, and received new data on the topological and dynamical invariants for the ^{222}Rn concentration time series. These results show evidence of deterministic chaos. Generally speaking, the results are of great theoretical and practical interest, as well as for applications in many fields, such as environmental (environmental radioactivity) and earth sciences, geophysics and physics, etc. [7–16, 40–46].

Acknowledgements

References

[1] N Packard, J Crutchfield, J Farmer and R Shaw, *Phys. Rev. Lett.* **45**, 712 (1988)

[2] M Kennel, R Brown and H Abarbanel, *Phys. Rev. A.* **45**, 3403 (1992)

[3] A Fraser and H Swinney, *Phys. Rev. A.* **33**, 1134 (1986)

[4] H Abarbanel, R Brown, J J Sidorowich and L Sh Tsimring, *Rev. Modern Phys.* **65**, 1331 (1993)

[5] P Grassberger and I Procaccia, *Physica D.* **9**, 189 (1983)

[6] F Takens, *In: Dynamical systems and turbulence*, Ed D Rand and L Young (Springer, Berlin 1981) p. 366

[7] R Mane, *In: Dynamical systems and turbulence*, Ed D Rand and L Young (Springer, Berlin 1981) p. 230

[8] M Sano and Y Sawada, *Phys. Rev. Lett.* **55**, 1082 (1995)

[9] J Hu, J B Gao and JK D White, *Chaos, Solitons & Fractals* **22**, 807 (2004)

[10] T Schreiber, *Phys. Rep.* **308**, 1 (1999)

[11] A V Glushkov, *Modern Theory of a Chaos* (OSENU, Odessa, 2012)

[12] A V Glushkov, O Khetselius, S Brusentseva, P Zaichko and V Ternovsky, Advances in Neural Networks, Fuzzy Systems and Artificial Intelligence. Recent Advances in Computer Engineering. Ed J Balicki. Lodz Univ., Lodz. **21**, 69 (2014)

[13] O Yu Khetselius, *Dynamical Systems Applications*. Ed J Awrejcewicz, M Kazmierczak, P Olejnik and J Mrozowski Wyd. Politechniki Ådzkiej, Lodz. T2, 145 (2013)

[14] O Yu Khetselius, S V Brusentseva and T B Tkach, Dynamical Systems Applications. Ed J Awrejcewicz, M Kazmierczak, P Olejnik and J Mrozowski Wyd. Politechniki Ådzkiej, Lodz. T2, 251 (2013)

[15] A V Glushkov, V V Buyadzhi and E L Ponomarenko, *Proc. Int. Geometry Center* **7(1)**, 24 (2014)

[16] A V Glushkov, O Yu Khetselius, Yu Ya Bunyakova, O N Grushevsky and E P Solyanikova, *Dynamical Systems Theory*. Ed J Awrejcewicz, M Kazmierczak, P Olejnik and J Mrozowski, Wyd. Politechniki Ådzkiej, Lodz. T1, 249 (2013)

[17] A V Glushkov, V Kuzakon, V B Ternovsky and V V Buyadzhi, *Dynamical Systems Theory*. Ed J Awrejcewicz, M Kazmierczak, P Olejnik and J Mrozowski, Wyd. Politechniki Ådzkiej, Lodz. T1, 461 (2013)

[18] A V Glushkov, O Yu Khetselius, Yu Ya Bunyakova, G P Prepelitsa, E P Solyanikova and E N Serga, *Dynamical Systems - Theory and Applications*. Lodz: Lodz Univ. Press (Poland) LIF111 (2011)

[19] A V Glushkov, G P Prepelitsa, S V Dankov, V N Polischuk and A V Efimov, *J. Tech. Phys.* **38**, 219 (1997)

[20] A V Glushkov, O Yu Khetselius and L Lovett L, *Advances in the theory of atomic and molecular systems: Dynamics, spectroscopy, clusters, and nanostructures, progress in theoretical chemistry and physics*, ed. P Piecuch, J Maruani, G Delgado-Barrio, and S Wilson (Springer, 2009), Vol. 20, p. 125

[21] A V Glushkov, G P Prepelitsa, Ya I Lepikh, V V Buyadzhi, V B Ternovsky and P A Zaichko, *Sensor Electr. Microsyst. Techn.* **11**, 43 (2014)

[22] A V Glushkov, V V Buyadzhi, A S Kvasikova, A V Ignatenko, A A Kuznetsova, G P Prepelitsa, V B Ternovsky, *Quantum Systems in Physics, Chemistry, and Biology*, pp. 169–180

[23] A V Glushkov, O Yu Khetselius, S V Malinovskaya, *Molec. Phys.* **106**, 1257 (2008)

[24] O Yu Khetselius, *J. Phys.: Conf. Ser.* **397**, 012012 (2012)

[25] A V Glushkov, S V Malinovskaya, O Yu Khetselius, A V Loboda, D E Sukharev and L Lovett, *Int. J. Quant. Chem.* **109**, 1717 (2009)

[26] A A Svinarenko, L V Nikola, G P Prepelitsa, T Tkach and E Mischenko, *AIP Conf. Proc.* **1290**, 94 (2010)

[27] A V Glushkov, Relativistic quantum theory. *Quantum Mechanics of Atomic Systems*, Astroprint, Odessa. 2008

[28] A V Glushkov, O Yu Khetselius, Yu M Lopatkin, T A Florko, O A Kovalenko and V F Mansarliysky, *J. Phys.: Conf. Ser.* **548**, 012026 (2014)

[29] A V Glushkov, *Atom in Electromagnetic Field* (KNT. Kiev, 2005)

[30] A V Glushkov, S V Malinovskaya, E P Gurnitskaya, O Yu Khetselius and Yu V Dubrovskaya, *J. Phys.: Conf. Ser.* **35**, 425 (2006)

[31] O Yu Khetselius, *Hyperfine Structure of Atomic Spectra*. Astroprint. Odessa, 2008

[32] A V Glushkov, S V Malinovskaya, I M Shpinareva, V P Kozlovskaya and V I Gura, *Int. J. Quant. Chem.* **104**, 512 (2005)

[33] A A Svinarenko, *J. Phys.: Conf. Ser.* **548**, 012039 (2014)

[34] T A Florko, S V Ambrosov, A A Svinarenko and T B Tkach, *J. Phys.: Conf. Ser.* **397**, 012037 (2012)

[35] M Lanfredi and M Macchiato, *Europhys. Lett.* **1997**, 589 (1997)

[36] K Koak, L Åaylan and O Åen, *Atm. Env.* **34**, 1267 (2000)

[37] X Chen, J Paatero, V-M Kerminen, L Riuttanen, *et al.*, *Boreal Environm. Res.* 299, p. 21

[38] D J Jacob and M J Prather, *Tellus. 42b*, 118 (1990)

[39] J-L Pinault and J-C Baubron, *J. Geophys. Res.* **102**, 101 (1997)

[40] A V Glushkov, V D Rusov, S V Ambrosov and A V Loboda, *New Projects and New Lines of Research in Nuclear Physics*. Ed G Fazio and F Hanappe (World Scientific, Singapore 2003) p. 126

[41] O Yu Khetselius, A V Turin, D E Sukharev, T A Florko, *Sensor Electr. Microsyst. Techn*. N1, 30 (2009)

[42] A V Glushkov, V V Buyadzhi and V B Ternovsky, *Proc. Int. Geometry Center* **6**, 6 (2013)

[43] A V Glushkov, V B Ternovsky, V V Buyadzhi and G P Prepelitsa, *Proc. Int. Geometry Center* **7**, 60 (2014)

[44] A V Glushkov, V M Kuzakon, V V Buyadzhi and E P Solyanikova, *Proc. Int. Geometry Center* **8**, 93 (2015)

[45] G P Prepelitsa, V V Buyadzhi and V B Ternovsky, *Photoelectronics* **22**, 103 (2013)

[46] V V Buyadzhi, *Photoelectronics* **24**, 128 (2015)