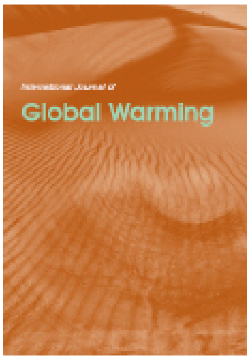

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Chaotic time series analysis of acid rain in Türkiye

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Abstract: Acid rain is an environmental problem that negatively affects nature and people globally. As a signal processing method, chaotic time series analysis is useful for understanding the behaviour of complex dynamic systems. This study aims to identify the chaotic behaviour of the dynamic system of acid rain characteristics, including the pH level, nitrate, and sulphate concentrations. A dataset from four rain collection stations in Türkiye between 2005 and 2022 is used to investigate the underlying dynamics and spatial distribution. The phase space is reconstructed, and the embedding parameters are obtained using Lyapunov exponentials, power spectral density, Hurst exponential, and mutual information functions to determine and predict the nonlinear properties of acid rain. Time series analyses revealed that the chemical components in acid rain demonstrated strongly nonlinear properties. Positive Lyapunov exponents prove the exponential divergence of the trajectories, which support the presence of chaos in the acid rain characteristics.

Keywords: nonlinear processes; dynamic system modelling; Lyapunov exponent; acid rain; Türkiye.

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1 Introduction

Acid rain has become an important environmental problem that has recently concerned the world (Ren et al., 2023). Acid rain is one of the consequences of excessive deposition of atmospheric pollutants. The two types of acid depositions can be classified as wet and dry. Wet acid deposition occurs when acidic materials penetrate water bodies in clouds (Dukic and Jerkovic, 2012). Acidic waters are carried from the atmosphere to the earth by rain, snowfall, sleet, and hail. Acid rain is formed by the dissolution of sulphur dioxide (SO_2) and nitrogen oxide (NOx) gasses in water droplets inside clouds (Burns et al., 2016). When these gases react with water and oxygen in the atmosphere, the result is the acidification of the water body with the inputs of sulphuric acid (H_2SO_4) and nitric acid (HNO_3) (Tontu and Tontu, 2022). The prevailing wind can transfer sulphur dioxide and nitrogen dioxide after being released from polluting sources. Dry deposition is the accumulation of acidic aerosols, particles, fly ashes, and atmospheric gases. This dry accumulation can then be transferred to the soil through the atmosphere. Even if these materials or gases are not acidic, they can turn into acids after contact with water in rain, snow, and fog. Dry deposition can increase the acidic behaviour of the rain waters and make surface waters acidic.

Acid rain adversely affects soil, water, plants, microorganisms living in soil and water, and man-made materials such as buildings and structures (Singh and Agrawal, 2008). Chemical compositions in acidic rainwater decrease the soil's pH level. Acid rain releases harmful materials (such as heavy metals) into the soil (Probst et al., 2000). This mobilisation of heavy metals in the soil can cause serious problems in the ecosystem (Cui et al., 2004; Abbasi et al., 2013). Acid rain affects human and animal health regarding the respiratory and food chain (Panwar and Shrirame, 2009). In recent years, it has been known that the toxic effects of acid rain damage rainforests.

Wet deposition mechanisms are more important in transporting pollutants from the atmosphere to the earth than dry deposition mechanisms. From this point of view, it is possible to obtain important information about the degree of air pollution in the local atmosphere by analysing the pollutant parameters in wet depositions (Beryland et al., 1982). Linear analysis methods are generally not useful for determining the structure of natural systems. Chaotic time series analysis is mostly used in nonlinear dynamic system theory (Kamislioglu and Kulalı, 2021). The chemical composition of acid rain is important to understand how local and regional pollutants are emitted and transported, and their potential ecological effects (Zhang et al., 2024). Understanding the dynamics of acid rain will provide a more reliable base for choosing a proper modelling and prediction method. Although several studies have been carried out to model acid rain with various methods, as far as we know, there is no study in the literature on the chaotic structure of acid rain. In this study, we conducted a chaotic analysis of the acid rain on the time series of pH value, nitrate, and sulphate concentrations in the wet rainwater samples from four rain collection stations in Türkiye. We reconstructed the relevant phase spaces based on 11,360 measurement data and then obtained the largest Lyapunov exponents, Hurst exponents, and mutual information functions in MATLAB. Furthermore, we interpreted the power spectrum graphs of the time series. The main contribution of this paper is to provide a detailed analysis of four chaotic indicators to examine the chemical characteristics in the acid rain time series. The largest Lyapunov exponent can be considered as a quantitative measurement of the chaotic extent of time series, as it is closely related to the degree of divergence of adjacent orbits in the attractors of the acid rain dynamic system.

The paper is organised as follows: Section 2 explains the background information about the chaotic analysis methods. Section 3 is devoted to revealing the chaotic behaviour of acid rain by applying the chaotic analysis methods. Finally, the concluding remarks and potential extensions are presented in Section 4.

2 Theory and methodology

Since linear systems, logistic mapping, and similar theories are insufficient to explain many empirical findings, the need for a new theory comes out in science. Lorenz (1963), one of the leading researchers in chaos theory, was the first to explain chaotic behaviour by mathematical modelling in weather systems. The main purpose of chaotic analysis is to obtain information about the dynamics of the system. However, the qualitative properties of the chaotic attractor alone do not provide precise information about the overall behaviour of the system in chaotic time series analysis. Therefore, quantitative analysis is also required to identify systems. Quantitative identifiers allow us to distinguish noise behaviour from chaotic behaviour and define how many variables are required to model the system's dynamics. Various studies show that mathematical relationships between quantitative identifiers can be useful for understanding the universal properties of chaotic systems (Hilborn, 2000).

Chaotic time series have a nonlinear structure. Accordingly, nonlinear models should be preferred to represent a chaotic system. We can divide naturally occurring facts (for dynamic systems that vary in one or more directions) into discrete time or continuous in terms of the measurement technique. Measurements taken at discrete time intervals can be derived from a subsample of continuous time measurements. Therefore, discrete-time-varying dynamics can be solved by different equations or iteration methods. The iteration method is a mathematical method obtained by repetition of iterations. If time is measured continuously and differential equations model the dynamic system, the solution of derivative equations is used for continuous time variations. However, there is no differential equation in time series derived from a variable which evolves through time. In this case, the measured signal is dominated by the irregularity/noise caused by environmental conditions. The irregularity/noise caused by environmental conditions prevails in the measured signal. One way of cleaning the data from the additive noise is by filtering (Aguirre and Billings, 1995). After removing this noise, chaotic time series analysis methods can be applied.

Analysis of chaotic time series is used to determine how the variables of a dynamic system change according to which parameters. The outcome of the analysis is important, as it helps us to determine how the system will behave in the next step (Reiss, 2001). When nonlinear analysis methods are applied, the signal must be reconstructed in phase space. Information about the nonlinearity of the system can be obtained from the point derived by analysing the equations of the dynamic system in phase space and the size attractor such as a closed curve.

Although several studies have been carried out to model acid rain, there is no study conducted on the chaotic structure of acid rain. However, as with many other natural systems, demonstration of the chaotic structure of acid rain will provide evidence that nature is controlled by chaos. In this study, the nonlinear behaviour of the rainwater data was determined using Lyapunov exponentials, power spectral density, Hurst exponential, and mutual information functions, among the chaotic analysis methods. Chaotic determinants can be obtained using the formulation summarised in Table 1 (Hilborn, 2000; Sprott, 2003; Kodba et al., 2005; Kamislioglu and Kulalı, 2021).

Table 1 Main formulas of chaotic analysis methods

Lyapunov exponent	$\delta\dot{X} = M(X(t))\delta X$	$\lambda^{Lp}\left(X_0, \delta X\right) = \lim\limits_{t\to\infty}\frac{1}{t}\log\frac{d\left(X_0, t\right)}{d\left(X_0, 0\right)}$
Power spectrum analysis	$F(t) = \sum\limits_{k=-\infty}^{\infty} C_{k^{e^{ikt}}}$	$C_k = \frac{1}{2\pi}\int_0^{2\pi} F(t)e^{-ikt}dt$
Hurst exponent	$R(N)/\ S(N) = kN^H$	$R(N) = \max\{X(n, N)\} - \min\{(n, N)\}$
Mutual information	$I(\tau) = \sum\limits_{n=1}^{N}(x(n), x(n+\tau))\log_2\left[\frac{P(x(n), x(n+\tau))}{P(x(n))P(x(n+\tau))}\right] I(\tau) \geq 0$	

3 Chaotic analysis of acid rain in Türkiye

In this study, the pH values, nitrate, and sulphate concentrations of rainwater samples from four different rainwater collection stations in Türkiye were analysed mathematically, and the nonlinear time series analysis methods interpreted the chaotic behaviour of acid rain. Automatic rain collection stations are in Amasra, Antalya, Balıkesir, and Çatalca. These rain collection stations provide periodical rainwater samples as wet and dry samples. A total of 11,360 samples from 2004 to 2022 based on the Turkish State Meteorological Service database were used to measure pH, nitrate, and sulphate concentrations. Figure 1 shows the locations of the rain collection stations under consideration.

Figure 1 Rain collection stations (see online version for colours)

3.1 Nonlinear time series analysis

Natural events mostly have nonlinear behaviour. To determine this nonlinearity, analysing dynamic state variables help us to observe information about the time series of dynamic systems. We used MATLAB, TISEAN, and MINITAB software programs to analyse the data. Figure 2 illustrates the time series of pH value, nitrate, and sulphate concentrations. As the time series graphs indicate, the parameters do not contain any periodicity. This is one way of starting to investigate chaotic behaviour in these systems. We applied chaotic detection methods after a nonlinear filtering procedure to remove irregularity or noise in the rainwater data to recover noise-free orbits (Kamislioglu and Kulalı, 2021). This prediction-based filtering includes a resetting mechanism that enables the reconstruction of attractors to reduce the effect of noise. Finite impulse response (FIR) filters are used in this study. FIR is a simple and effective structure in digital filter design; it gives fixed results constantly in non-repetitive applications, and it is easy to get results in the linear phase.

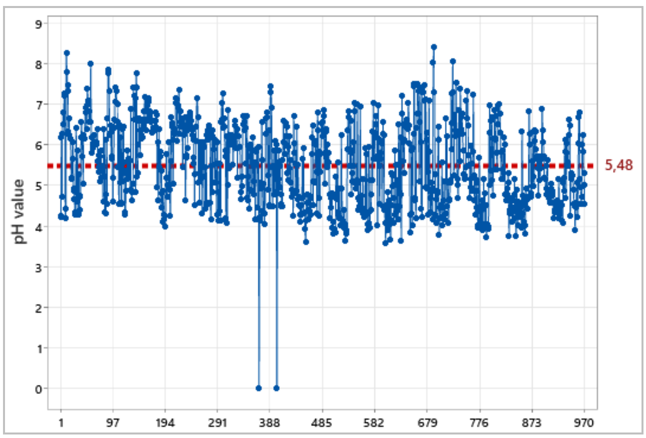
3.1.1 Lyapunov exponents

The presence of the largest positive Lyapunov exponent is the most important indicator of the chaotic behaviour of the time series (Yıldırım and Altınsoy, 2018). The largest Lyapunov exponents were calculated using MATLAB. The largest Lyapunov exponents of pH, nitrate, and sulphate concentrations for all stations are depicted in Figure 3. All parameters have at least one positive Lyapunov exponent. According to the graphics, it can be said that this dynamic system exhibits a chaotic behaviour, and it is not possible to make long-term predictions. It means that neighbouring orbits in phase space diverge from each other and are sensitive to initial conditions. Analysing Figure 3, we observe that the largest value is positive because it is calculated as ‘the average logarithmic divergence or convergence ratio of two close points of two-time series separated by an initial distance’. The largest Lyapunov exponent of pH, nitrate, and sulphate for the Amasra region is associated with the small difference in concentration variation between time series.

3.1.2 Hurst exponent

The presence of Hurst exponent equals 0.5, which displays random walking as a non-chaotic behaviour. If it gets values in the range $0.5 < H \leq 1$, then a possibility of chaos exists in the system. On the other hand, if it is in the range $0 \leq H < 0.5$, then the system has a chaotic fractal structure; hence, the same system depends on the initial conditions. In this paper, the Hurst exponents are found as 0.55 and 0.60, and the consequences are presented in Figure 4. The window interval is the time lag, and averaging is performed over the time window. The slope of the line gives us the Hurst value numerically.

Figure 2 Time series graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, (j), (k), (l) Çatalca (see online version for colours)



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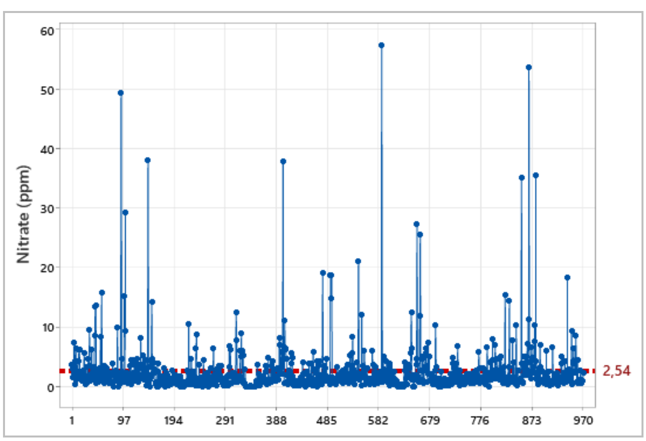
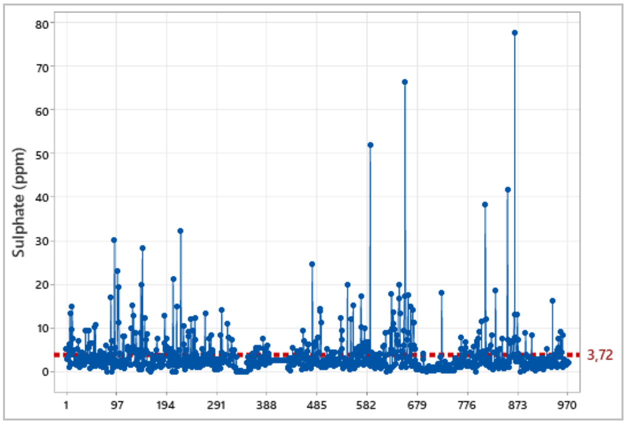


(b)

Figure 2 Time series graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, (j), (k), (l) Çatalca (continued) (see online version for colours)



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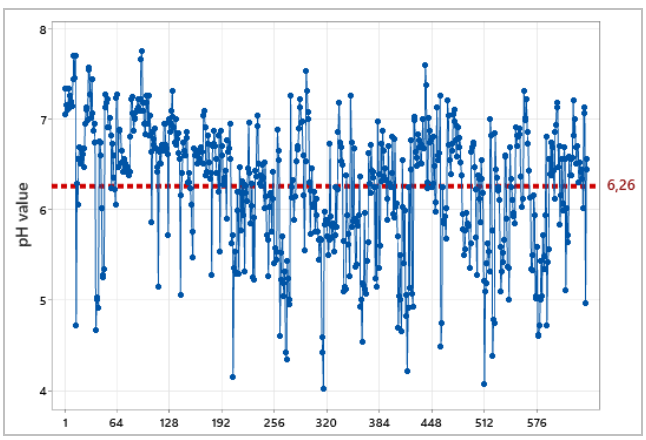

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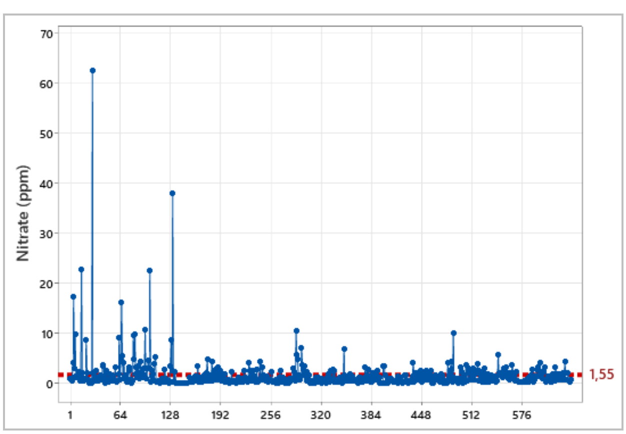
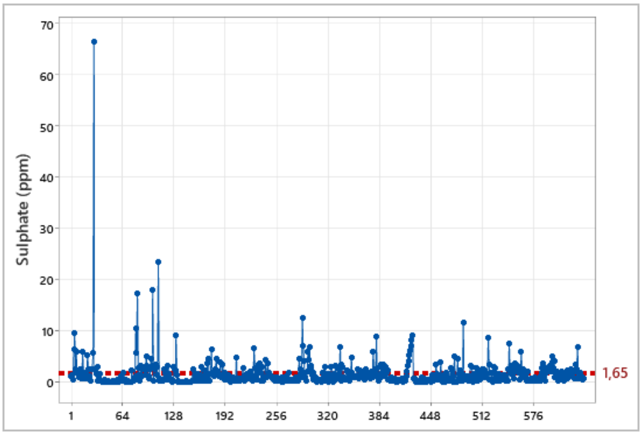


(e)

Figure 2 Time series graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, (j), (k), (l) Çatalca (continued) (see online version for colours)



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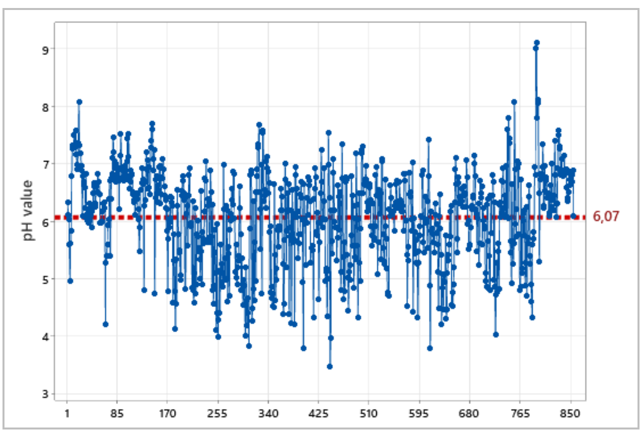

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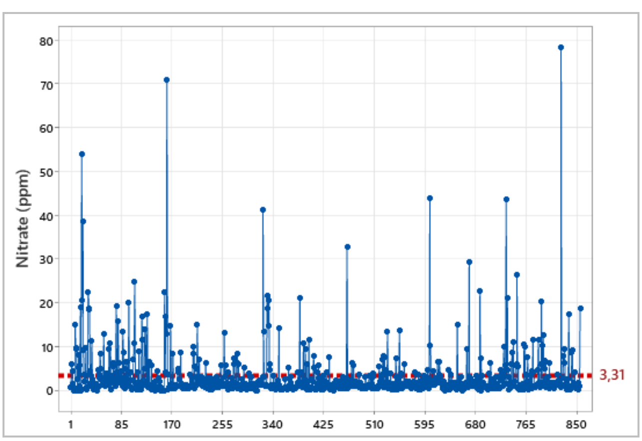
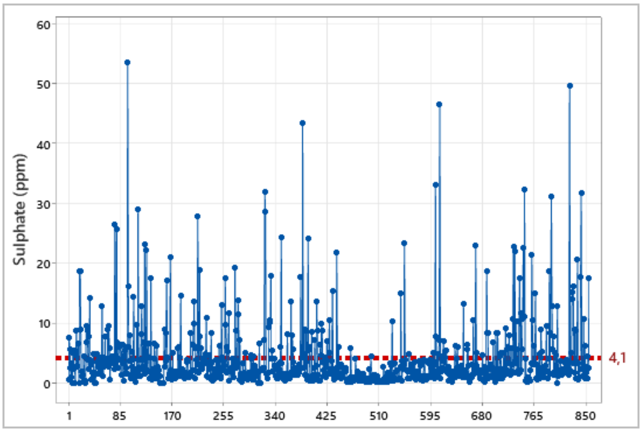


(h)

Figure 2 Time series graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, (j), (k), (l) Çatalca (continued) (see online version for colours)



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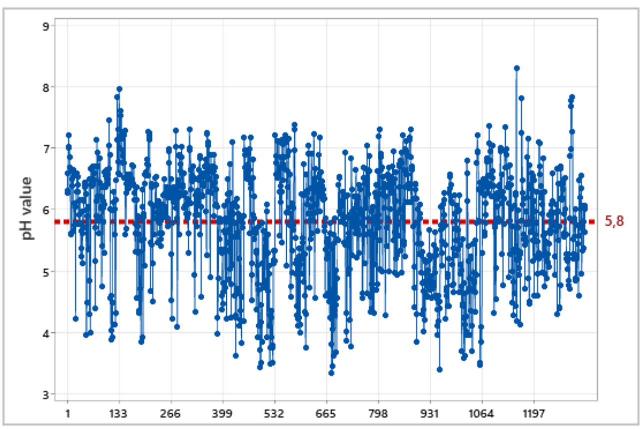

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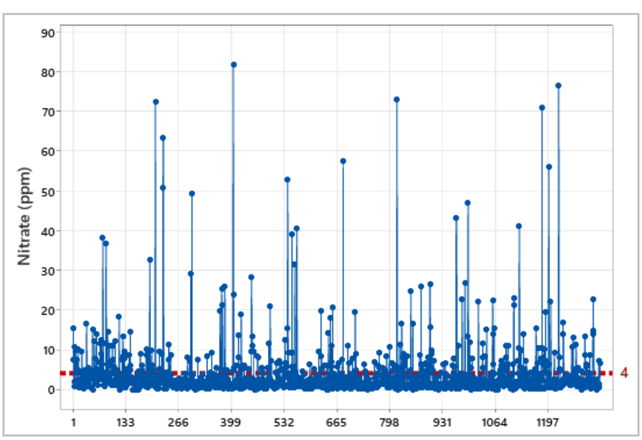

(k)

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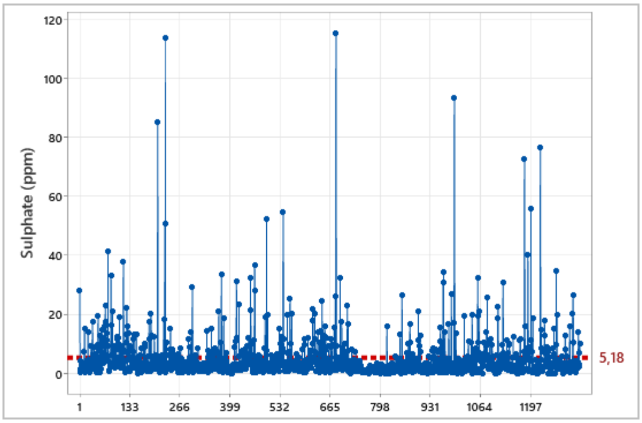
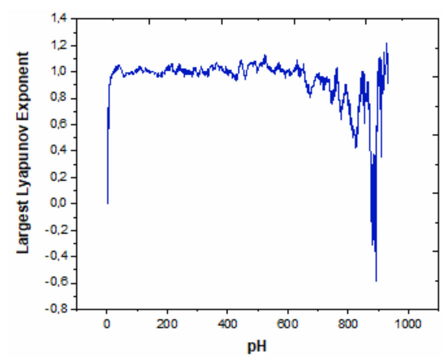


(l)

Figure 3 The largest Lyapunov exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (see online version for colours)



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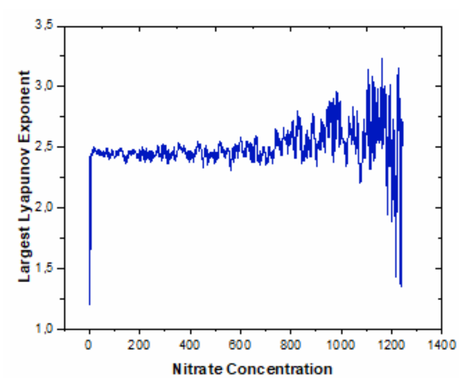
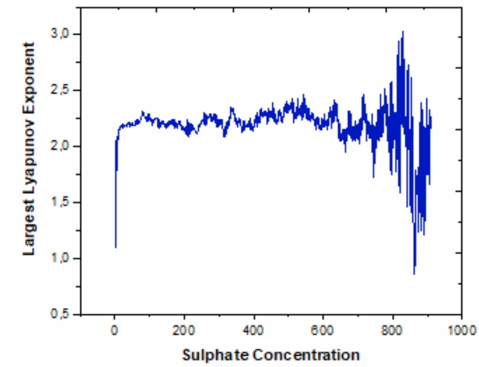


(b)

Figure 3 The largest Lyapunov exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



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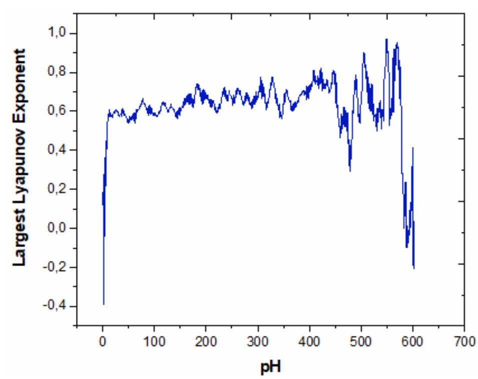

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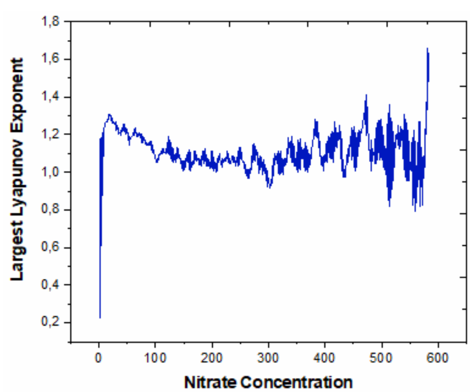

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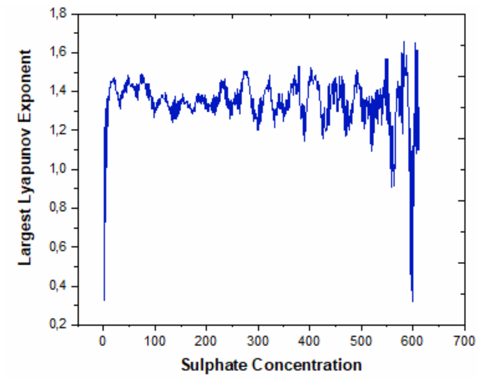

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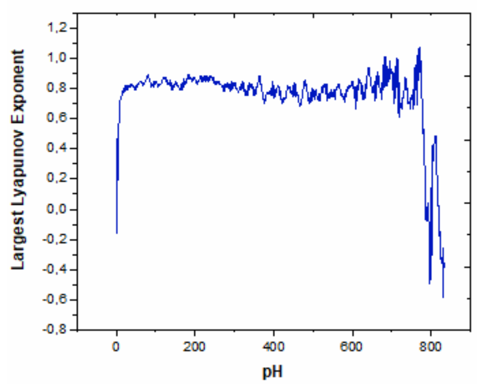

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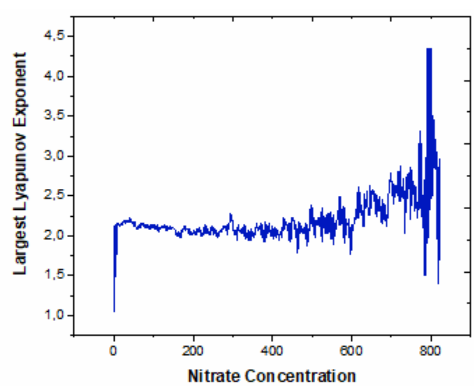

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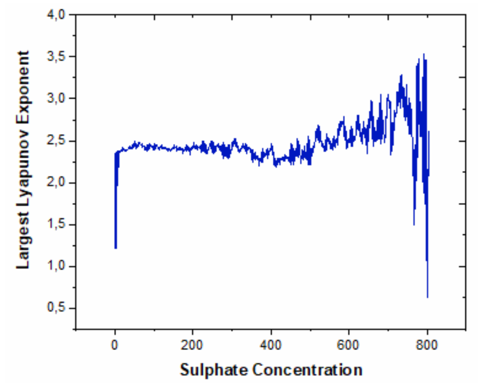
Figure 3 The largest Lyapunov exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



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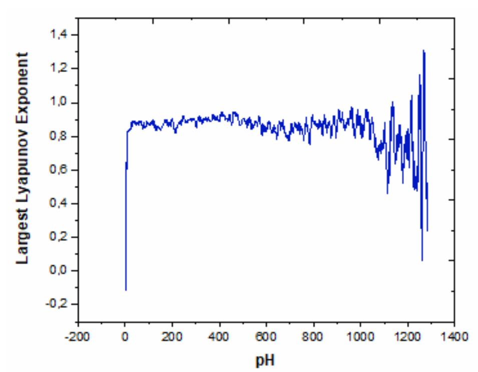

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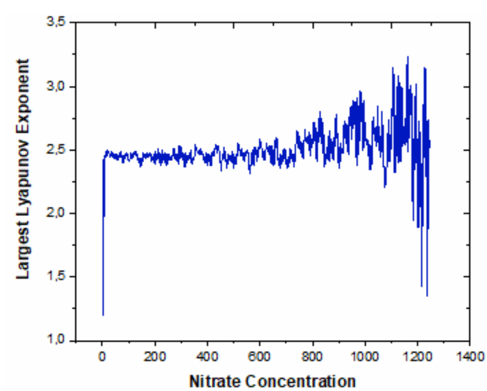

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Figure 3 The largest Lyapunov exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)

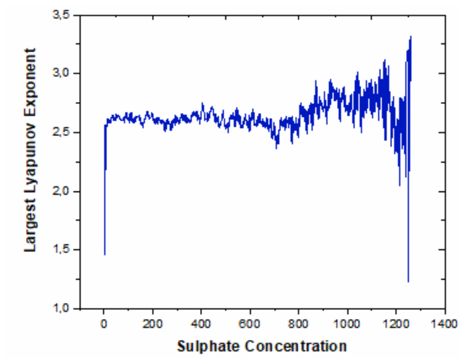

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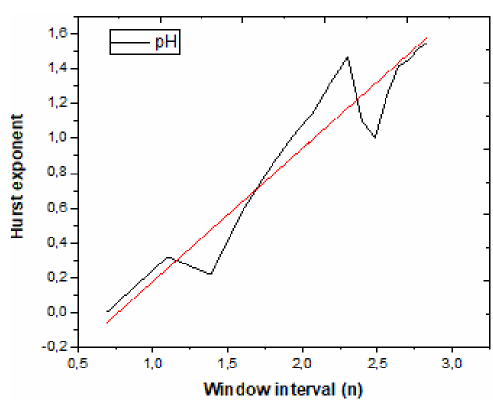
Figure 4 Hurst exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (see online version for colours)



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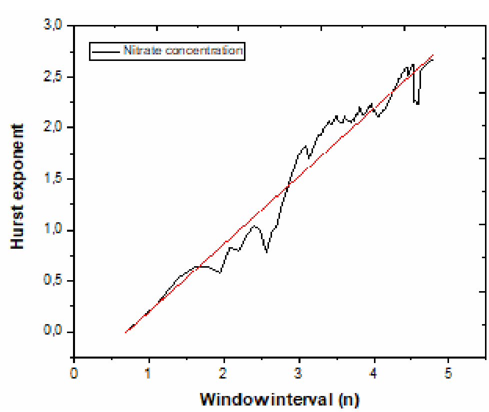

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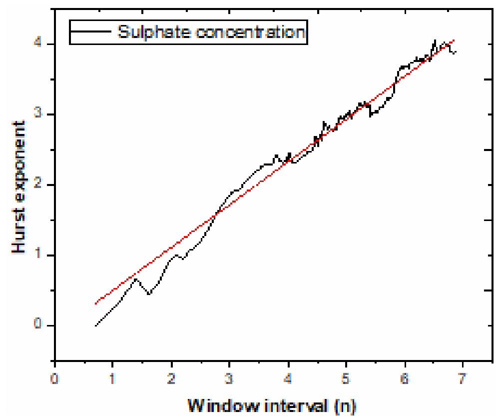
Figure 4 Hurst exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



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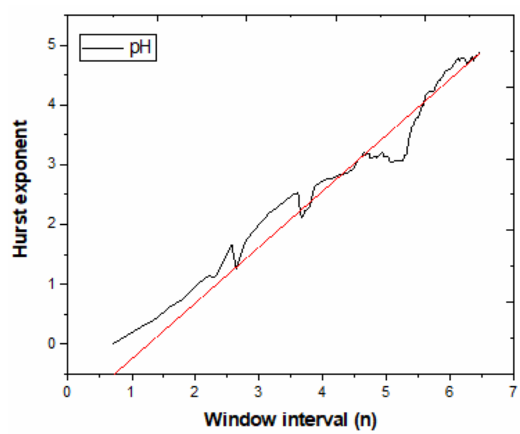

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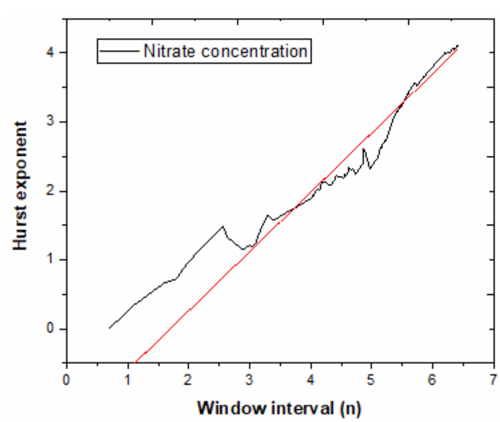

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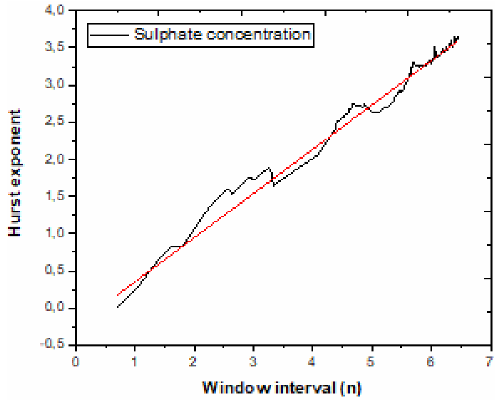

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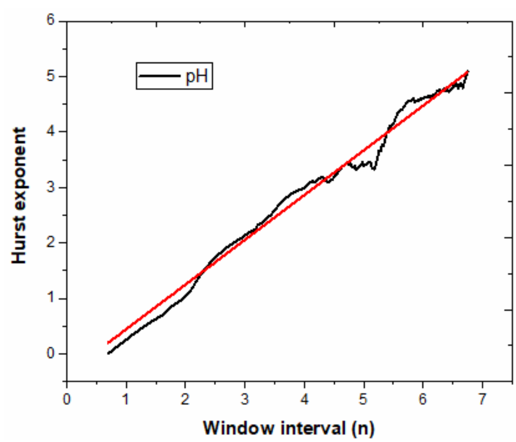
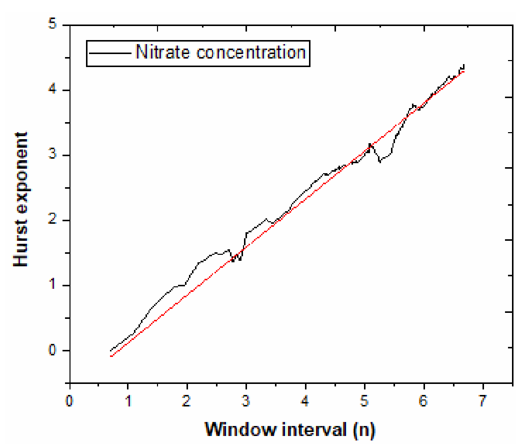
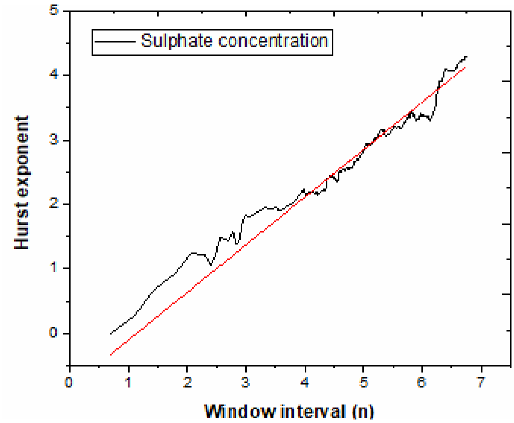


(g)

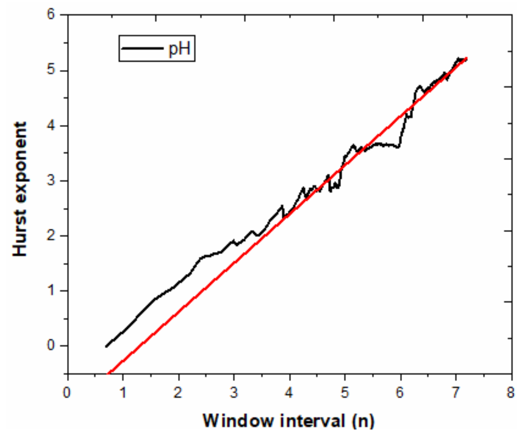


(h)

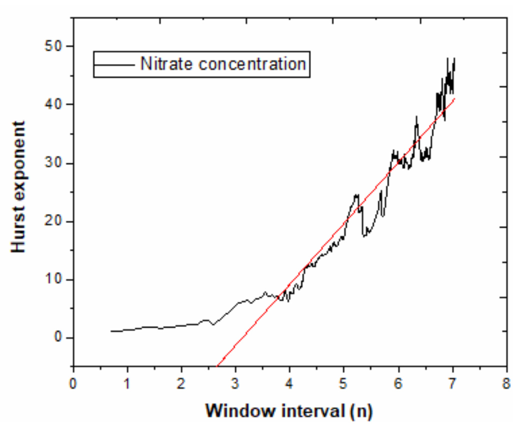
Figure 4 Hurst exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



(i)

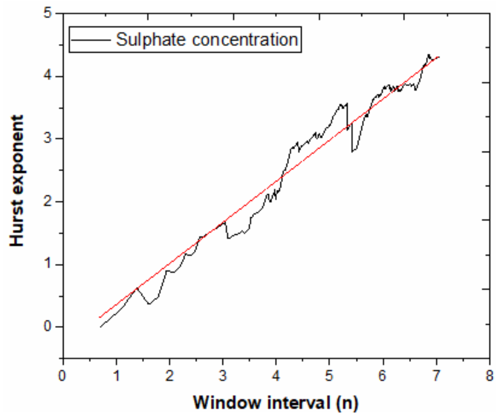


(j)



(k)

Figure 4 Hurst exponent graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



(l)

An H value ranging from 0.5 to 1 indicates a persistent series with long-term positive autocorrelation. This means that a high value in the series tends to be followed by a higher value, and the time series will likely continue to show an increasing trend. On the other hand, a value in the range of 0 and 0.5 indicates an anti-persistent series with a long-term transition between high and low values in adjacent pairs. This means that a low value will likely follow a high value, and the value will tend to increase after that. The tendency to switch between high and low values persists for a long time and also obeys a power law. A value of $H = 0.5$ denotes short memory in which (absolute) autocorrelations rapidly fall to zero exponentially.

3.1.3 Mutual information function

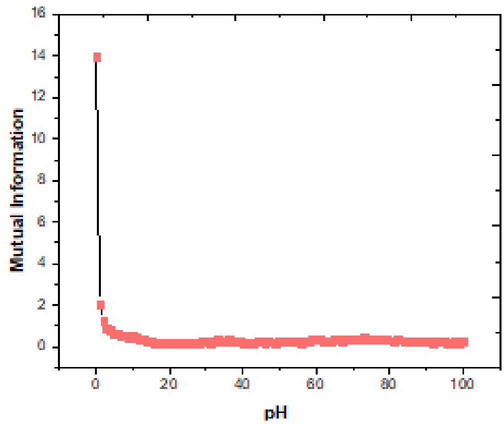
It is a useful method for achieving the delay time for reconstructing an attractor. Figure 5 gives the mutual information regarding the time delay ($m = 5$) and time lags ($m = 0, 1, 2, \ldots, 20$). Their corresponding surrogates are presented in Figure 5.

Mutual information is used to measure dependencies without bias for relationships in the dataset. It is a method for estimating time-dependent correlation with an empirically biased estimation of time-delayed common information for a time series. As we have observed in Figure 5, the bias of time-delayed common information is equivalent to shared information between two point distributions separated from the same system by infinite time.

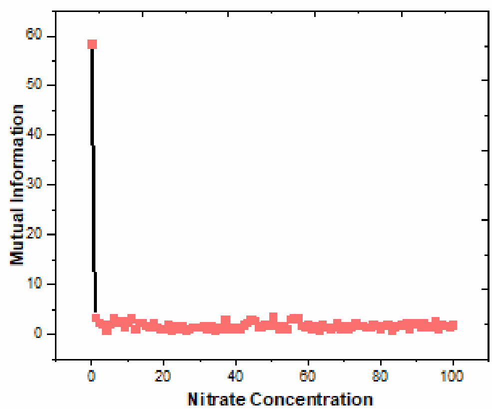
3.1.4 Power spectrum

Power spectrum analysis is another method for detecting chaotic signals of a dynamic system. The continuity of the peaks in the power spectrum indicates the presence of chaotic behaviour in the time series. Power spectrum graphics were drawn on MATLAB. Power spectrum graphics of pH, nitrate, and sulphate concentrations for all stations are shown in Figure 6. According to the power spectrum graphics, it can be said that this dynamic system has chaotic behaviour due to significant peaks.

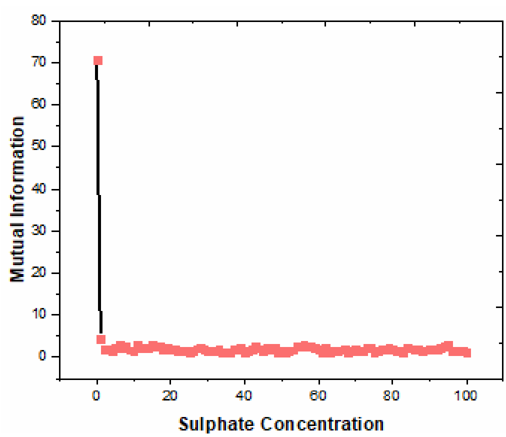
Figure 5 Mutual information graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (see online version for colours)



(a)

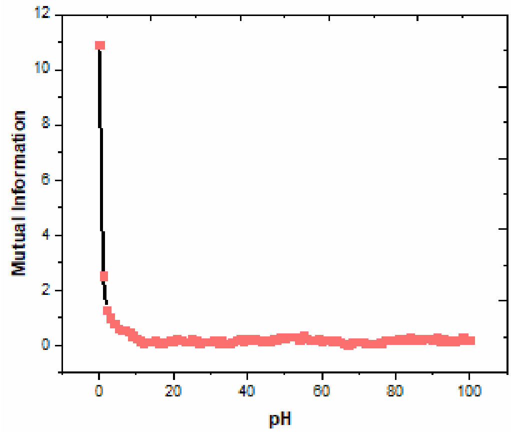


(b)

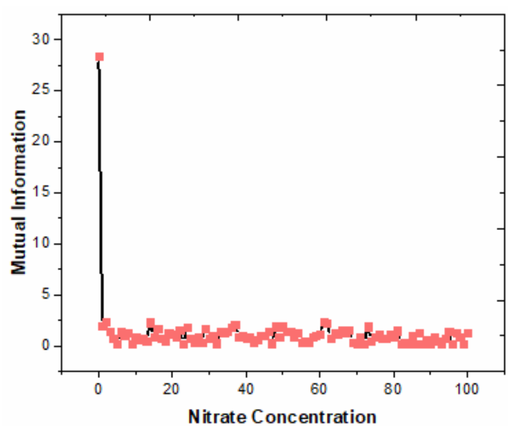


(c)

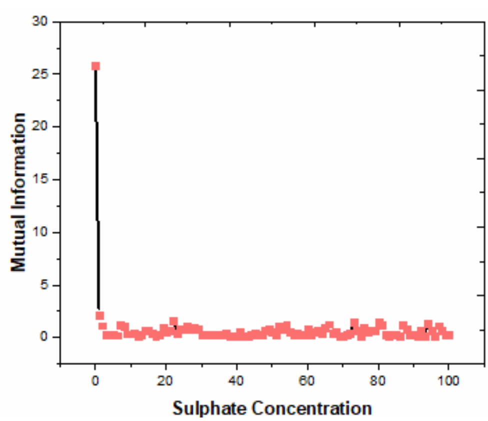
Figure 5 Mutual information graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



(d)

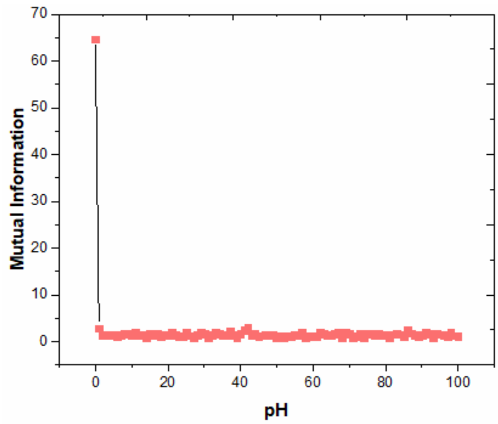


(e)

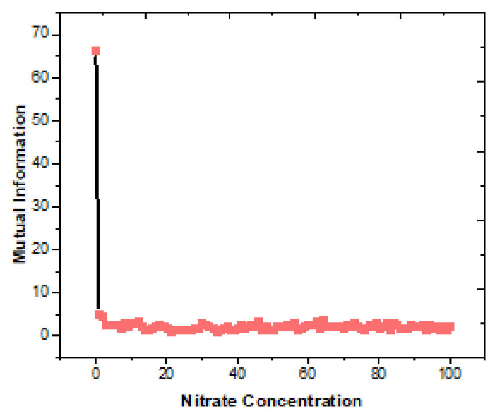


(f)

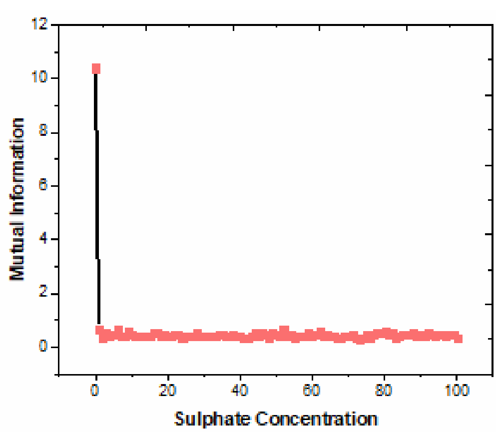
Figure 5 Mutual information graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



(g)

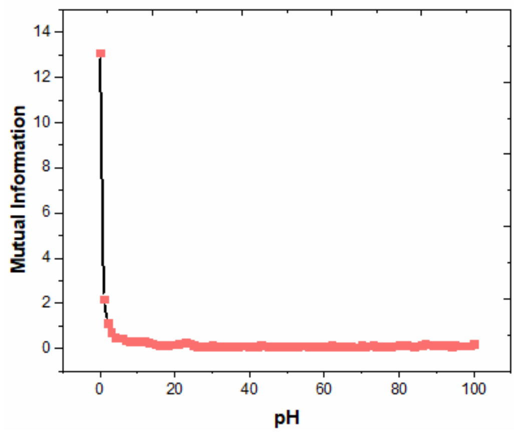


(h)



(i)

Figure 5 Mutual information graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



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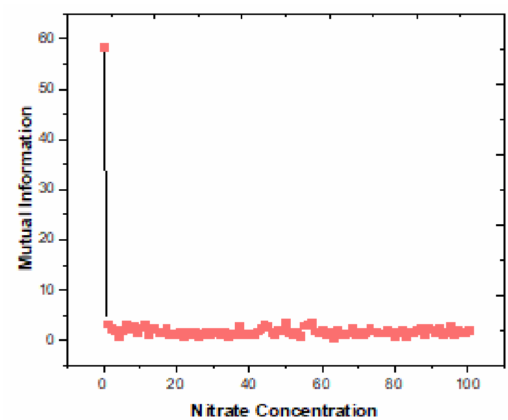

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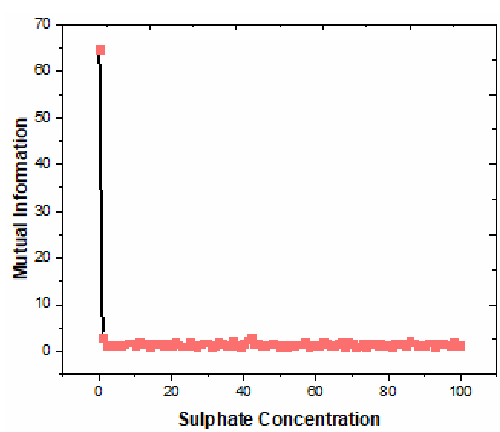
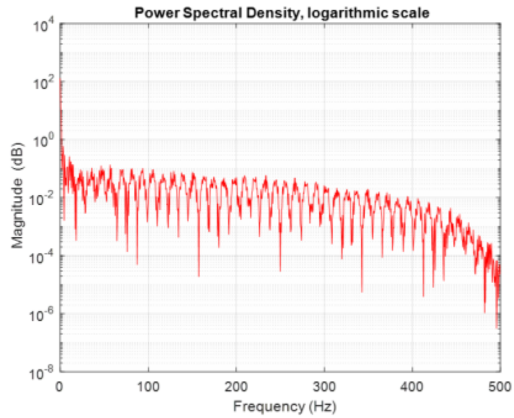
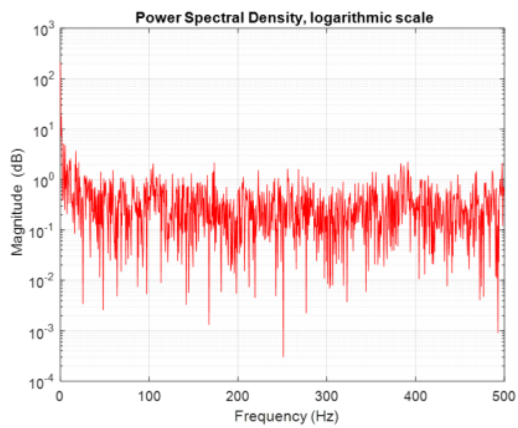


(l)

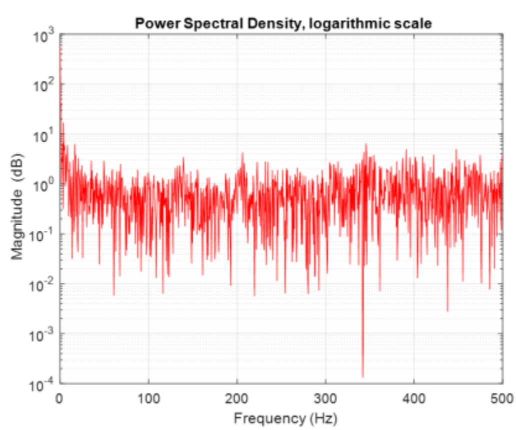
Figure 6 Power spectrum graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (see online version for colours)



(a)

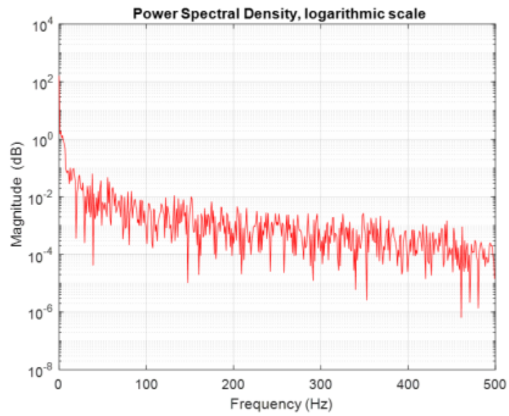


(b)



(c)

Figure 6 Power spectrum graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



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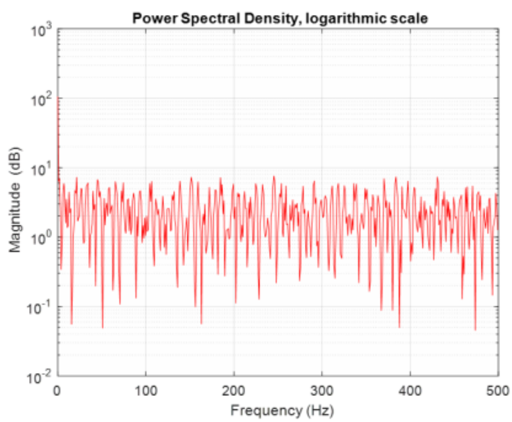

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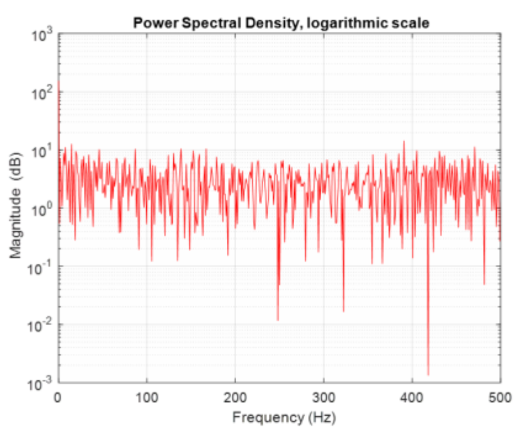
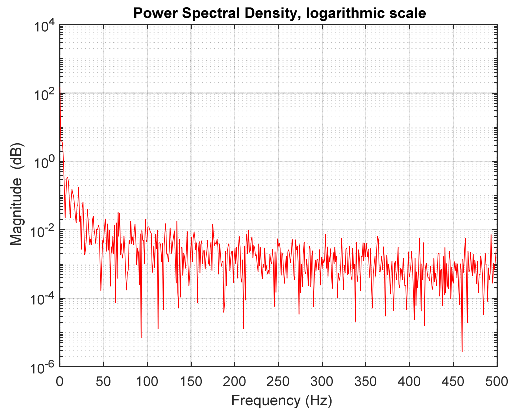


(f)

Figure 6 Power spectrum graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



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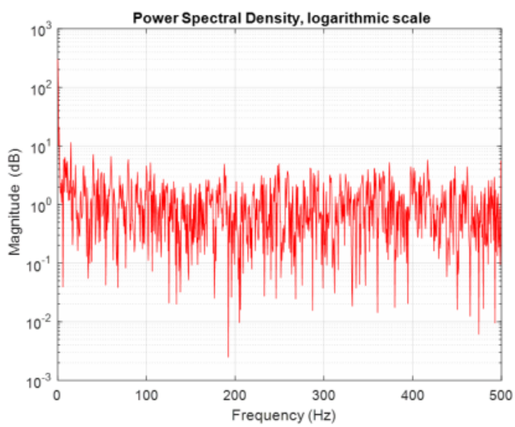

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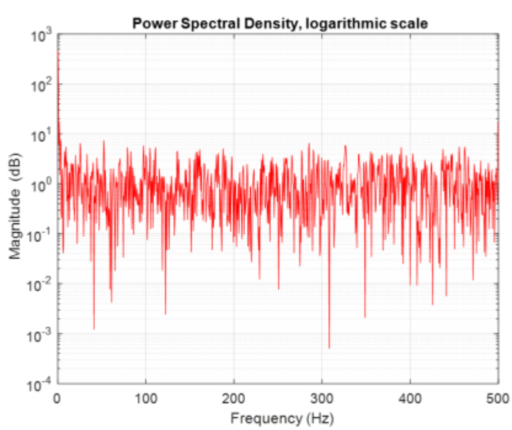
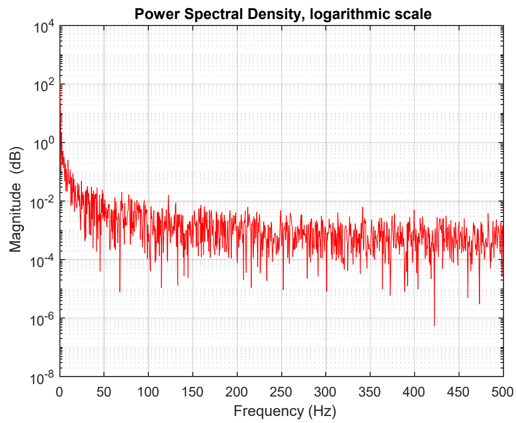
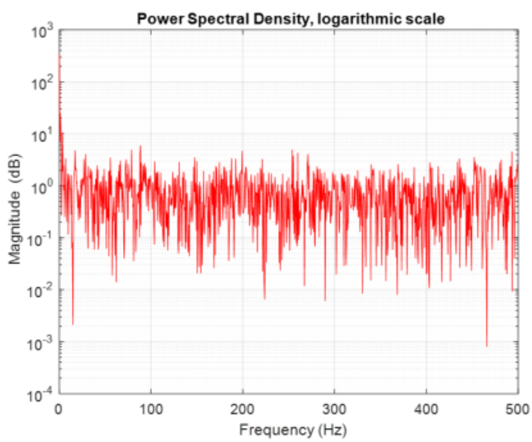


(i)

Figure 6 Power spectrum graphics of pH, nitrate, and sulphate for (a), (b), (c) Amasra, (d), (e), (f) Antalya, (g), (h), (i) Balıkesir, and (j), (k), (l) Çatalca (continued) (see online version for colours)



(j)



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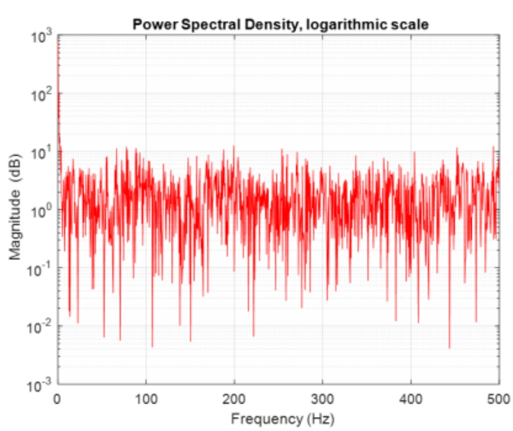

(l)

3.2 Results and discussion

Analysis of chaotic time series is a convenient method to evaluate the behaviour of highly complex dynamic systems. The system is named chaotic if nonlinear time series analysis methods indicate irregular or unpredictable behaviour. Chaotic time series analysis conducted in this study revealed that the acidic components of the precipitation do not show linear behaviour. Furthermore, they do not show any complex or unpredictable behaviour like noise. However, these parameters have been proven to have nonlinear characteristics. We thus conclude that nonlinear analysis methods can be used to calculate the total amount of acid rain falling on the earth and to determine their potential ecological impacts. In this way, it will be possible to accurately predict the negative ecological effects of acid rain by making reliable environmental risk analyses.

As can be seen from the analysis of the computational results, the largest Lyapunov exponents of pH, nitrate, and sulphate are 1.219, 3.236, and 3.036 for Amasra; 0.973, 1.658, and 1.655 for Antalya; 1.076, 4.350, and 3.540 for Balıkesir; 1.315, 3.236, and 3.325 for Çatalca, respectively. The value of the Hurst exponent calculated from the original dataset is determined by taking the slope of the line passing through the dataset, and it means that there is a long-term dependent process for the data that is shown to be greater than zero. We further observed that the pH value, nitrate, and sulphate concentrations in acid rain showed an increasing trend.

4 Conclusions

One of the types of pollution that occurs in the atmosphere is acid rain. Pollution has become a growing problem between countries, and international agreements have been signed to prevent cross-border pollution. To predict air pollution more accurately, there is a need to reveal whether acid rain exhibits chaotic behaviour. In this study, pH, nitrate, and sulphate concentrations in wet precipitation samples from four rainwater collection stations in Türkiye were analysed by chaotic analysis methods. The most important criterion for the existence of chaos is to have at least one positive Lyapunov exponent. It is also the condition for sensitivity to initial conditions. In this study, we first calculated the Lyapunov exponents. According to our calculations, the largest Lyapunov exponents of the all-time series have at least one positive Lyapunov exponent. Power spectrum analysis is another criterion for detecting chaotic behaviour. The continuity of peaks in the power spectrum indicates the presence of chaotic behaviour in the time series. Finally, Hurst exponents and mutual information functions are obtained for the continuously measured dataset. These are other evidence of the existence of chaotic behaviour in acid rain. The implementation results show that it will be possible to obtain more accurate results in the estimation of acid rain by the chaotic structure and to take measures in the long term to prevent possible negative consequences of air pollution. The study can be extended to other geographic regions worldwide as a future research area.

Acknowledgements

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