MULTIFRACTAL CROSS-CORRELATION OF ATMOSPHERIC POLLUTANTS AND TEMPERATURE IN DIFFERENT ENVIRONMENTS

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This paper presents a multifractal detrended correlation and cross-correlation analysis between PM_{10}, CO and temperature. Data have been collected for 11 months in three types of sites: urban background, urban- traffic and regional. For the analyzed time-series multifractal characteristics with persistent cross-correlations are observed. The source of the multifractality is found to be the long-range type correlations.

Keywords: multifractal, PM_{10}, CO, temperature, long range correlations, Hurst surface, cross-correlation

1. Introduction

The atmosphere is a complex system, composed from aerosols and gases which are continuously interacting which each other [1]. The study of atmospheric constituents is an important task because they have a direct and an indirect effect (acting as cloud condensation nuclei) on the Earth radiative budget, by scattering and absorbing radiation. Thus, on a long term, they can influence the climate, and on a shorter time scale they can have an impact on air quality and on human health [2].

Experimental investigations on atmospheric aerosols can be performed using high precision mass measurements for micro and nanoparticles [3]. New techniques are continuously reported to improve the accuracy of detection of the atmospheric particles and aerosols, and to facilitate on-site and on-line detections and monitoring [3,4]. Multipole Paul traps could represent versatile tools for environment monitoring working independently or coupled to Aerosol Mass Spectrometers [5-7].

The physical and chemical properties of the aerosols are strongly influenced by the meteorological and ambient conditions. The correlations

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between the aerosol, gases concentrations and the meteorological parameters determined by the highly-nonlinear dynamics involved in their interactions are reflected in the apparent erratic recorded time-series. A complete and relevant analysis, cannot be achieved by the classical (conventional) statistics mainly due to the non-stationary behavior of the atmospheric data. In the case of a stationary data set, the statistical properties do not change over time and a sliding widow with the same number of points has the same characteristic distribution. That is one reason why conventional statistics cannot depict the nonlinear characteristics of non-stationary data series.

Over the last few decades an increasing interest in the development of new methods for analysis of complex, non-stationary data is manifested. Contrary to the traditional descriptive analysis with all the limitations involved, new statistical theories and mathematical formalism such as those based on fractal and multifractal analysis, complex network and visibility graphs, etc., have a strong applicability in the investigation of atmospheric data [8-10].

Multifractal method assesses data over a wider range of temporal scales and spectrum of fluctuation exponents and are based on fragmentation of the time series in self-similar segments and on the investigation of the scaling capacity derived from a power law behavior. A first important step in the evaluation of the cross-correlation between two non-stationary time series was reported by Podobnik and Stanley [11].

Several studies have investigated the link between the atmospheric constituents and meteorological parameters. For example, the multifractal cross-correlation between PM_{25} and four meteorological parameters (temperature, wind speed, relative humidity, pressure) were proven to have multifractal characteristics [12]. Also, the correlation between global CH_4 and temperature showed wide multifractality due to long term correlations on long and short scales [13].

In the present work we investigate the multifractal multiscale detrended correlation and cross-correlation (MM-DCCA) between each pair of combination of PM_{10}, CO and temperature in three different types of natural environments.

The structure of the paper is as follows: the methodology for site selection, data analysis and MM-DCCA are presented in subsections of section 2. The results obtained for different environments and the source of multifractality are presented in section 3, while the main conclusions of the article are summarized in section 4.

2. Methodology

2.1 Sites and data

The multifractal multiscale analysis presented in this paper is based on the environmental data provided by the National Air Quality Monitoring Network

(NAQMN, www.calitateaer.ro). Three types of sites were chosen for the analysis, depending on their location and characteristics:

1. B1 station - an urban background station placed in the Western part of Bucharest (44.45; 26.04) next to Morii Lake.
2. B6 station - a traffic station placed in the center of the city (44.44, 26.10, 5 km away from B1) with a high density of cars (more than 10000/day [14])
3. E3 station (47.32; 25.13) - situated at Poiana Stampei, in the Northern part of Romania, at an altitude of 912 m.

Bucharest and its surrounding area are one of the most polluted sites in Romania [15]. The major pollution sources have been previously investigated: traffic, industry, residential heating, waste and landfill management, dust intrusions, pollen [16-22]. For a final comparison with the urban stations, a regional EMEP E3 station - was chosen.

Meteorological data (temperature, pressure, relative humidity RH) are provided by the NAQMN for sites B1 and EMEP, while for B6, measurements reported at the Filaret meteorological station (situated at 2 km distance) and provided by Weather Graphics [23] were used.

The analysis was performed for PM_{10} and CO hourly measurements from 01 January 2018 to 21 November 2018. In this study we have used only the measurements that passed the quality check criteria and that are validated by the NAQMN. The missing points have been replaced by the average of the neighbor points, resulting in a total number of points of 6500 for each atmospheric parameter.

2.2 Multifractal Multiscale Detrended Cross-Correlation Analysis (MM-DCCA)

The multifractal formalism consists in the evaluation of Hurst exponent for different scales (frequencies) and different order of fluctuations. Taken a time series with N data points, the main steps are [24]:

- a profile is created by subtracting the mean ($\bar{x}$) from each data point (x_i) and integrating the time series

$$X(j) = \sum_{i=1}^{j}(x_i - \bar{x})\,; j = 1,2,\ldots,N \tag{1}$$

- *X(j)* is divided into $N_s = Int(N/s)$ non-overlapping intervals, where s is the number of points (window length) in the time interval. If *N* is not exactly divided by s, in order not to exclude some points, the deviation of *X(j)* is performed twice, from beginning to end and reverse.
- for each interval, the local trend (obtained as a second order fit, $\hat{X}_\nu(k)$) is subtracted from the profile and then the variance is computed, $F_x^2(s,\nu)$:

$$F_x^2(s,\nu) = \frac{1}{s}\sum_{k=1}^{s}\left[X_\nu(k) - \hat{X}_\nu(k)\right]^2 \quad ,\nu = 1,2,\ldots N_s \tag{2}$$

1. a q-order fluctuation function is defined as:

$$F_x(q,s) = \left\{\frac{1}{2N_s}\left[\sum_{\upsilon=1}^{s} F_x^2(s,\nu)\right]^{q/2}\right\}^{1/q} \quad ,\nu = 1,2,\ldots N_s. \tag{3}$$

2. for self-afine series, the q order fluctuation function is described by a power law dependence on s as:

$$F_x(q,s) \approx s^{H_X(q)}, \tag{4}$$

where the generalized Hurst exponent, $H_X(q)$, is computed as the slope of the fluctuation function versus s, in log-log scale.

If $H_X(q)$ does not depend on q, the time series is monofractal and conversely, if $H_X(q)$ depends on q, then the time series is multifractal. The scaling behavior of the data can differ on different ranges of fluctuations: for $q < 0$, the $H_X(q)$ is specific for small fluctuations, while for $q>0$ the $H_X(q)$ is characteristic for large fluctuations.

Also, depending on the $H_X(q)$ value, the time series can be characterized as long range correlated (persistent behavior) for a value greater than 0.5; non-correlated for a value of 0.5 (specific for Gaussian white noise); and anti-correlated (anti-persistent) for $H_X(q)$ less than 0.5. The persistent behavior means that a large (small) rise in the concentration of the data series is more likely to be followed by a large (small) value. For the anti-persistent behavior, a decreasing in the data series is more likely to be followed by an increment, and vice-versa.

In the case of two time series (X and Y) with the same number of points, a generalized Hurst exponent can be defined similarly, the only difference consisting in the computation of the variance function:

$$F_{xy}^2(s,\nu) = \frac{1}{s}\sum_{k=1}^{s}\left|X_\nu(k) - \hat{X}_\nu(k)\right|\left|Y_\nu(k) - \hat{Y}_\nu(k)\right| \tag{5}$$

where $\hat{Y}_\nu(k)$ is the local trend for the second time series.

It was proved that a single scaling exponent does not describe the whole properties of the time series and it possibly not reveals the internal dynamics of signals. Gierałtowski [25] proposed a multiscale multifractal analysis for the computing $H_X(q)$ dependence versus the range of the scale s, without any initial time-scale assumptions. An extension of this formalism was proposed by Shi et al. [26] to the analysis of cross-correlation properties between two time series. Such investigation of the cross-correlation can be visualized as a Hurst surface, each point representing the generalized dependence H_{XY} (q, s). If there is a dependence of H_{XY} on the fluctuation parameter q for a fixed s, the two time-series are multifractal cross-correlated, otherwise, if no dependence is observed, the two time series are mono-fractal.

In order to better quantify the fractal characteristics of the time series, we proposed two parameters: the strength of multifractality ($\Delta_s H(q)$) and variation of cross-correlation ($\Delta_q H(s)$) [9]

$$\Delta_s H(q) = |H(q_{max}, s) - H(q_{min}, s)| \tag{6}$$

$$\Delta_q H(s) = |H(q, s_{max}) - H(q, s_{min})|. \tag{7}$$

The strength of multifractality is a measure of the change in multifractality, while the second parameter is used to identify the multifractality variation across the different scales analyzed.

3. Results

3.1 Correlations in the atmospheric parameters (PM_{10}, CO, temperature)

The analysis of the correlation in the time-series of the investigated atmospheric parameters was performed for two types of urban environments.

After preliminary tests similar as in [9], we choose as the starting width of the window scales s ∈ (30, 150), corresponding to (1.25 days, 6.25 days), with a slide length of 1 point. Then the window is moved and expanded until reaching the final width s ∈ (40, 200) corresponding to the time interval of around (1.6 days, 8.33 days).

The individual Hurst surfaces for PM_{10}, CO and temperatures corresponding for the two environments are shown in Fig. 1.

As observed, all the parameters present specific dependence of Hurst exponent, demonstrating the multifractal characteristics. The values of the Hurst exponent greater than 0.5 suggest that the fluctuations are persistent.

Looking on each column, a significant modification of characteristics the Hurst surfaces can be seen, with an increasing departure from horizontality in the specific order PM_{10}, CO and temperature. This is equivalent with an increasing in multifractality, which in turn is an indicator of the stability of the dynamics of the time-series at different scale and order of fluctuations. Consequently, the most active dynamics dependent by the fluctuations are those involved in temperature variation. It follows by CO data-set and by PM_{10} - that seems to have a more stable behavior.

Looking on each line, the Hurst surfaces in the two environments are similar, but for the traffic station, the Hurst values at all the scales are smaller at large fluctuations. The Hurst surfaces for CO are different in the urban background and traffic region: for small fluctuations, the urban site has higher values than in the traffic station, while in the large fluctuations zone, the values of the Hurst exponent decrease to approximately the same value (0.7).

For temperature time-series, Hurst surfaces in the two environments have different characteristics compared to the Hurst surfaces for PM_{10} and CO. However, the similarities between their characteristics for the two location are obvious, as expected. For the temperature measured at the two city sites, the lower values (~0.5) of H are obtained when q is 0 and the highest values (1.5) are obtained when q=-5.

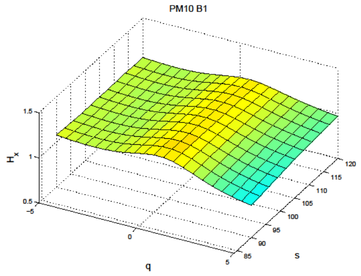
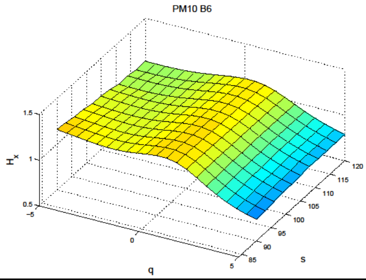
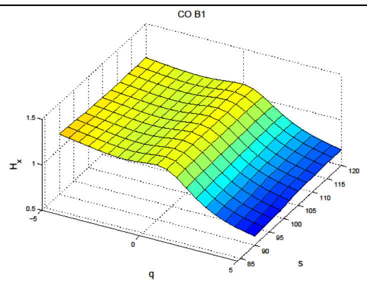
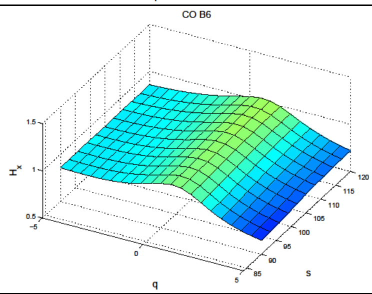
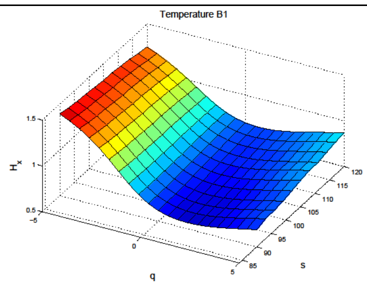
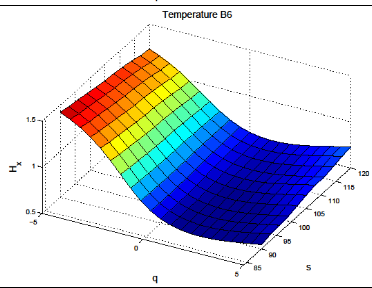
	B1	B6
PM_{10}		
CO		
temp		

Fig. 1 Hurst surfaces for PM10 (first row), CO (second row) and temperature (third row) in suburban (B1 station, left) and traffic (B6 station, right) environment

Table 1 shows the values of the strength of multifractality ($\Delta_s H(q)$) and of the variation of correlation ($\Delta_q H(s)$). The highest strength of multifractality is for temperature and the lowest for PM_{10}. Also, for these two data sets, $\Delta_s H(q)$ is higher for all the parameters in the traffic conditions, which could be explained by the more complex processes that take place in such locations. For example, in the traffic environments there are more sources that produce different aerosols and gases, thus the chemical and physical reactions between the aerosols and gases are more complex. This is not the case for CO, whose dynamics seems to be strongly biased and quite similar at large and small fluctuation order (q), consequently less multifractal.

For all data set investigated, $\Delta_s H(q)$ decreases while s increases, different from parameter $\Delta_q H(s)$ that have two distinct behavior: increases with the increasing of q for the suburban environment, while, conversely, for the traffic station, decreases when q increases. Consequently, as expected, at traffic station the dynamics at small fluctuations is more dominant than at large fluctuations.

Table 1

Strengths of multifractality and variation of correlation

Single-Correlation	$\Delta_s H(q)$		$\Delta_q H(s)$	
	s_min	s_max	q_min	q_max
PM_{10} B1	0.22	0.09	0.02	0.11
PM_{10} B6	0.42	0.24	0.13	0.05
CO B1	0.56	0.45	0.04	0.07
CO B6	0.21	0.14	0.00	0.07
TEMP B1	0.71	0.38	0.14	0.18
TEMP B6	0.88	0.50	0.19	0.18

3.2 Cross-correlations of pairs PM_{10}, CO and temperature

We search now for the multifractal characteristics of the cross-correlated pairs PM_{10}, CO and temperature, and investigate if there are new information that can be revealed.

Figure 2 illustrates the Hurst surfaces of the cross-correlation PM_{10}-temperature and PM_{10}-CO at the two stations. As observed, for all the analyzed cases, the correlations are long range persistent and the Hurst values are almost similar in both environments.

However, specific differences can be noted. The range of the Hurst values is small for the cross-correlation PM_{10}-temperature (between 0.6 and 1.1) across all the fluctuations interval, while for the cross-correlation PM_{10}-CO, the range of Hurst values is different on the q scale: lower for high fluctuations and higher for small fluctuations.

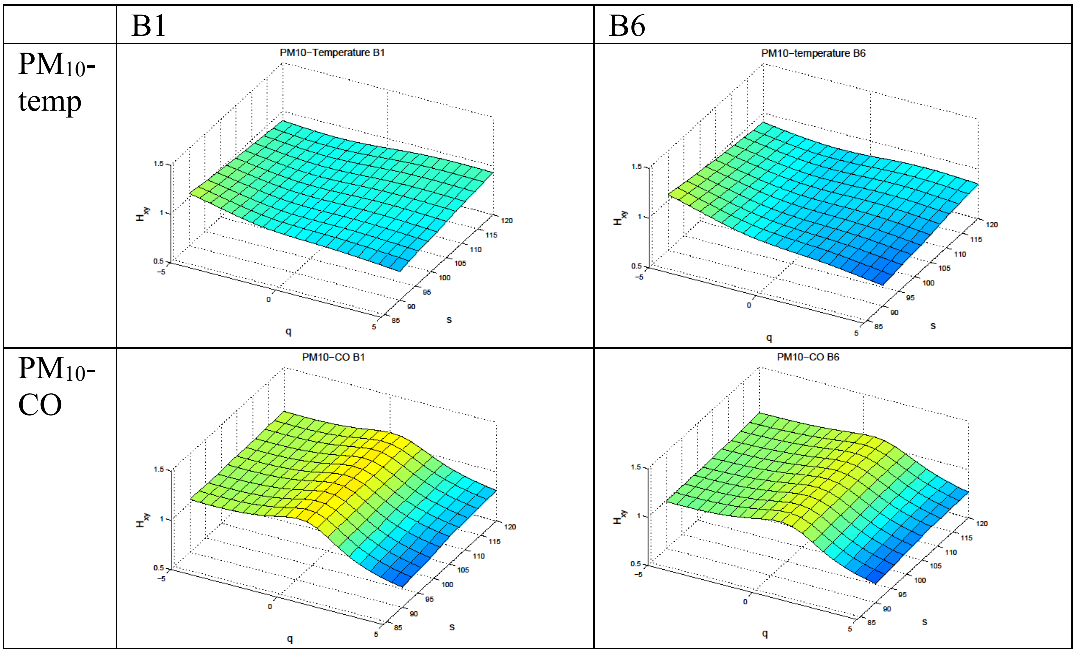


Fig. 2 Hurst surfaces for cross-correlation PM_{10}-temperature (first row), PM_{10}-CO (second row) in suburban (B1 station, left) and traffic (B6 station, right) environment

Hurst exponents for the cross-correlation PM_{10}-CO pair correlation decreases, at large fluctuations, towards 0.5. We can assume that, at short term, pollution can accumulate while at a larger scale of time a quasi-stationary state is reached.

Table 2

Strengths of multifractality and variation of cross- correlation

Cross-correlation	$\Delta_s H(q)$		$\Delta_q H(s)$	
	s_min	s_max	q_min	q_max
PM_{10}-temp B1	0.24	0.03	0.13	0.14
PM_{10}-temp B6	0.35	0.06	0.15	0.14
PM_{10}-CO B1	0.33	0.25	0.02	0.10
PM_{10}-CO B6	0.29	0.26	0.04	0.07

From the data of Table 2, it is observed that $\Delta_s H(q)$ is higher for the smallest scale of investigation (around 1 day) for all cross-correlated time-series and decreases at the maximum scale (around 8 days). Consequently, the strongest interactions manifest at low time-scale.

The strength of multifractality is higher for the cross-correlation PM_{10}-CO than PM_{10}-temperature only at B1 station, while the variation of cross-correlation is high for the PM_{10}-temperature at both stations.

3.3 Cross-correlations of atmospheric parameters in remote environments

The Hurst surfaces for the cross-correlation between the CO from the different city environments (urban background and traffic) and the CO measured at the regional station (EMEP) are illustrated in Fig. 3. Both show multifractal characteristics and persistent correlations. This means that an increasing in one data series will be followed by also an increasing of concentrations in the second data series, or if the values in one data set is decreasing, a decrease will be observed also in the second data set.

The Hurst surfaces for the comparison of the city station with the regional station are similar, with higher values at small fluctuations, while at the large fluctuation regime, a decrease is observed. By comparing the two analyzed cases, the Hurst values are higher for the cross-correlation between urban background and regional station than for the cross-correlation between traffic and the regional station.

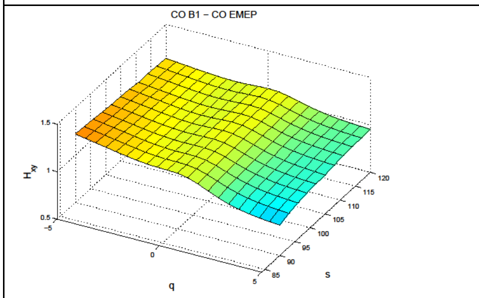
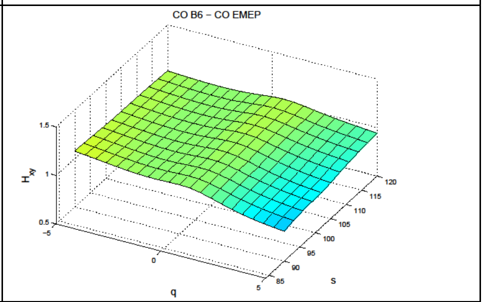
CO B1-CO EMEP	CO B6-CO EMEP
	

Fig. 3 Hurst surfaces for cross-correlation CO from the suburban station (left) and traffic station (right) with the CO measured at the regional station

3.4 Source of long-range correlations

There are two sources of multifractality: long range correlations (at small or large fluctuations) and broad probability mass distribution. A shuffling procedure, which involves the reordering of the values in the time series, destroys the long-range correlations and does not affect probability mass distribution. So, if the source of the multifractality is the long-range correlation, the shuffled data will have a *H* value close to 0.5, specific for uncorrelated gaussian noise [26,27]. If the multifractality is derived from the fat-tailed distribution, the shuffling procedure will not affect the Hurst surfaces, and another procedure is required, which involves the calculation of surrogate data from the original data by randomizing the phase in the Fourier space.

We evaluated the source of multifractality for the cross-correlated data and illustrate the case between the CO from the urban background station and the one from the traffic station in order to find hidden influences in the data sets.

In Fig. 4 are shown the Hurst surfaces for this cross-correlation and the Hurst surface after the shuffling procedure. Before the shuffling procedure, the surface shows multifractality characteristics and long-range correlations. After the shuffling, the H values for each independent time-series (H_X, H_Y) and for the correlated one (H_{XY}) are close to 0.5, which indicates that the source of multifractality is of long-range type for small and large fluctuations at all the scale analyzed.

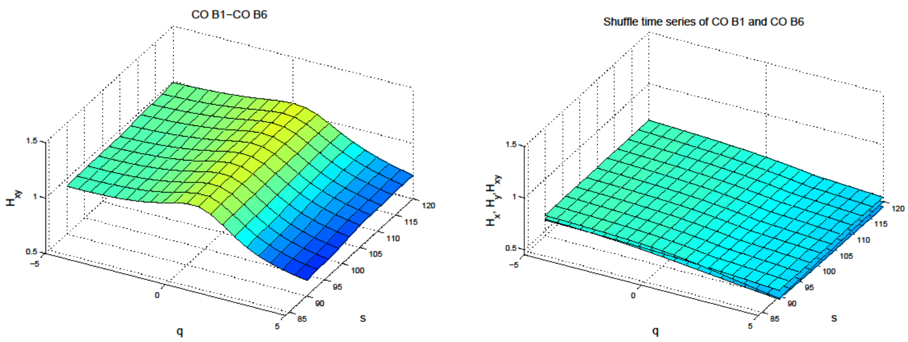


Fig. 4 The cross-correlation between CO measured at the suburban station and CO measured at the traffic station (left) and the Hurst surfaces after the shuffling procedure

4. Conclusions

In this paper, correlation, and cross-correlation for PM_{10}, CO and temperature have been investigated. The data analyzed were collected for almost one year in three different types of environments: city- background, city-traffic and regional.

The time series exhibit multifractal and nonlinear features for single and cross-correlated data and the fluctuations are long range correlated for all the analyzed cases. The highest multifractal strength is for the temperature at the traffic station, followed by CO and the most stable dynamics is characteristic for PM_{10}. The cross-correlated analysis of pair PM_{10}-temperature at the traffic station shows a more sensitive variation at small temporal scale (around 1 day). After the shuffling procedure of the data for CO measured at the background station and at the traffic station and for their correlated set, the Hurst surfaces approaches close to 0.5, which demonstrates the long-range correlations as the source of multifractality for the whole range of small and large fluctuations.

Analysis of the long-term correlations offers new important information related to the complexity of air pollutants dynamics, useful for improving the investigation and forecasting the air quality, mainly in how the future might be affected by the present and past values of specific atmospheric parameters/pollutants, at different temporal scales.

Further studies are planned for the analysis of the data from a network of stations in different environmental locations in order to map the results of MM-DCCA methods and to define new quantitative descriptors that can signal sudden changes in the dynamics of atmospheric processes and providing a better forecasting of air pollutant time-series.

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