Aerosol and Air Quality Research, 15: 926–934, 2015

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2014.05.0091

Multifractal Processes and Self-Organized Criticality of $PM_{2.5}$ during a Typical Haze Period in Chengdu, China

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ABSTRACT

Scaling and multifractal properties of the hourly $PM_{2.5}$ average concentration series at the four air monitoring stations of Chengdu (southwestern China) were explored by using a multifractal detrended fluctuation analysis method, during a typical haze episode (from 1 March to 17 March, 2013). Using shuffling procedure and phase randomization procedure, the major sources of multifractality in these $PM_{2.5}$ series are studied. The results show that the multifractality nature of $PM_{2.5}$ series is mainly due to long-range correlation. At the same time, the non-Gaussian probability distributions also partly contribute to the multifractal behaviour. The scale-free power laws behaviours are found to govern the cumulative distributions statistics for $PM_{2.5}$ concentration fluctuations. The temporal evolutions of the multifractality were investigated by the approach of a sliding window. Further, we attempt to find the answers to the following questions: how does long-range correlation and power-law distribution in $PM_{2.5}$ evolution emerge? It is inviting to do it in a self-organized criticality (SOC) framework, which was specially designed to model the dynamics of complex systems. A novel $PM_{2.5}$ evolution model is developed on the bases of SOC theory. The model displays robust power law behaviour in certain dynamical region. The self-organized criticality properties of $PM_{2.5}$ evolution are discussed. This SOC behaviour is related to a statistically steady state that implies the presence of long-range correlation and power-law distribution in $PM_{2.5}$ evolution during the haze period. It is the stability of SOC that causes the haze period to be sustained for a long time in Chengdu.

Keywords: $PM_{2.5}$; Multifractal; Long-range correlation; Power-law distribution; Self-organized criticality.

INTRODUCTION

Chengdu (30.67°N, 104.06°E) is the largest city in southwestern China, with a population of about 11 million and an area of about 12,000 km^2. It is located in the western portion of the Sichuan Basin. Chengdu city is surrounded by Longquan Mountain to the east and Qionglai Mountain to the west of the city. The unique topographical condition directly affects meteorological condition of pollutant diffusion. The prevailing wind direction in Chengdu is from the north. However, the change of surface wind speed is small and the annual average wind speed is only 1.2 m/sec, with high static wind frequency, up to 45%–50% (Yin *et al.*, 2013). With rapid economic growth and increasing energy consumption, particulate matter (PM) pollution has become the primary environmental problem in Chengdu. It is considered to be one of the four regions in China most seriously affected by haze (Wang *et al.*, 2013). In the unique meteorological conditions of Chengdu, haze pollution is strongly correlated with the emissions of pollutants from anthropogenic sources and gas-to-particle conversion (Wang *et al.*, 2013). Thus, it is extremely important to understand the dynamic characteristics and mechanisms of the long-term variation the of $PM_{2.5}$ pollution.

At last ten years, increasing attention has been paid to the scaling behaviour of air pollution process. Environmentalists come to realize the importance of investigating scaling properties of air pollution series. It is accepted that air pollution can be considered as a complex nonlinear dynamic system (Windsor and Toumi, 2001; Shi *et al.*, 2008), and good understanding of the scaling properties of air pollution series is of great importance for pollution modelling (Lee *et al.*, 2006; Chelani, 2012). Lee *et al.* (2003, 2006) discussed the scaling characteristics of air pollutant concentration by mono- and multi-fractal box counting methods. Anh *et al.* (1997) had interpreted long-range dependence of daily ozone and NO_2 pollution by trend analysis. Windsor and Toumi (2001) examined the statistical characteristics of UK pollution time series by three methods, named Sigma-T, R/S and Kurtosis. But these methods could suffer

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misleading results due to the presence of non-stationary in the data. In order to overcome this problem, in 1994, Peng *et al.* (1994) introduced detrended fluctuation analysis (DFA) that detects the long range correlations in the nonstationary time series. Afterwards, in 2002, Kantelhardt *et al.* (2002) introduced multifractal detrended fluctuation analysis (MF-DFA), an extended version of DFA, to analyze the multifractal properties of time series. These methods have been widely applied in the detection of persistence and multifractality of trends in the nonstationary time series, such as precipitation (Kantelhardt *et al.*, 2006), acid deposition (Zhu *et al.*, 2003) and air pollution (Shi *et al.*, 2008). It is now known that multifractal is the appropriate framework for scaling fields in time series and thus can provide the natural framework for analysis and modelling various air pollution processes (Lee *et al.*, 2006). But what are the sources of the long range correlation and multifractality of air pollution? The little research has been found on this topic, especially those of $PM_{2.5}$ during a typical haze episode. In our previous study, we found that the presence of scaling behaviour of SO_2, NO_2, PM_{10} pollution series can be linked to self-organized criticality (SOC) of air pollution (Shi and Liu, 2009). However, this study data were daily pollution indexes data in Shanghai, from July 2000 to July 2006. The smallest time interval was one day. One cannot certain whether this dynamical law prevails at the shorter timescales.

In this paper, we first study the long range correlation and multifractal scaling behaviour of $PM_{2.5}$ during a typical haze period in Chengdu, China. Secondly, we investigate the sources of multifractality of $PM_{2.5}$. Furthermore, we study the probability distribution functions of $PM_{2.5}$ concentrations. At last, we put forward a numerical sandpile model to illuminate SOC of $PM_{2.5}$ pollution, based on SOC theory. We found that the presence of persistence and concentration fluctuations of $PM_{2.5}$ can be attributed to SOC mechanism.

MATERIAL AND METHODS

Data Sets

The hourly average $PM_{2.5}$ concentrations data used in this study are supplied by the China National Environmental Monitoring Centre (http://113.108.142.147:20035/emcpublish/). There are eight automatic monitoring stations. In this study, only four stations are examined because of missing data. The selected sites are the urban stations, including the Shilidian, Sanwayao, Liangjiaxiang and Shahepu, from 1 March to 17 March, 2013. These urban stations are located in a heavily populated area in Chengdu city and are intended to provide information pertaining to human exposure. It is noteworthy that although the studied time period consists of 408 h, only 390–405 readings for each station were collected due to instrument calibration and maintenance. However, the missing observations seemed to be evenly distributed throughout the time period. The missing data have been replaced by the mean between two of nearest data. The mean values of $PM_{2.5}$ in all examined stations are with the order: Shilidian (171 $\mu g/m^3$), Shahepu (170 $\mu g/m^3$), Sanwayao (166 $\mu g/m^3$) and Liangjiaxiang (162 $\mu g/m^3$).

Fig. 1 shows the geographical location of Chengdu in the Sichuan Basin. Meteorological data were obtained from Weather Underground (http://www.underground.com/). During the investigated time period, the city's average temperature is 17.1°C. The prevailing wind direction in Chengdu is from the south. The average wind speed is 1.6 m/sec. The change of surface wind speed is small at the main period and the highest wind speed is 8.1 m/sec. The average relative humidity is 49.7%. During the period of March 1 to March 17, 2013, precipitation is zero. This time period is a typical haze period in Chengdu.

Multifractal Detrended Fluctuation Analysis Method

Detrended fluctuation analysis (DFA) can obtain the famous Hurst exponent, or scaling exponent, which detects the long range correlations in the nonstationary time series (Peng *et al.*, 1994). But for multifractal time series, a single

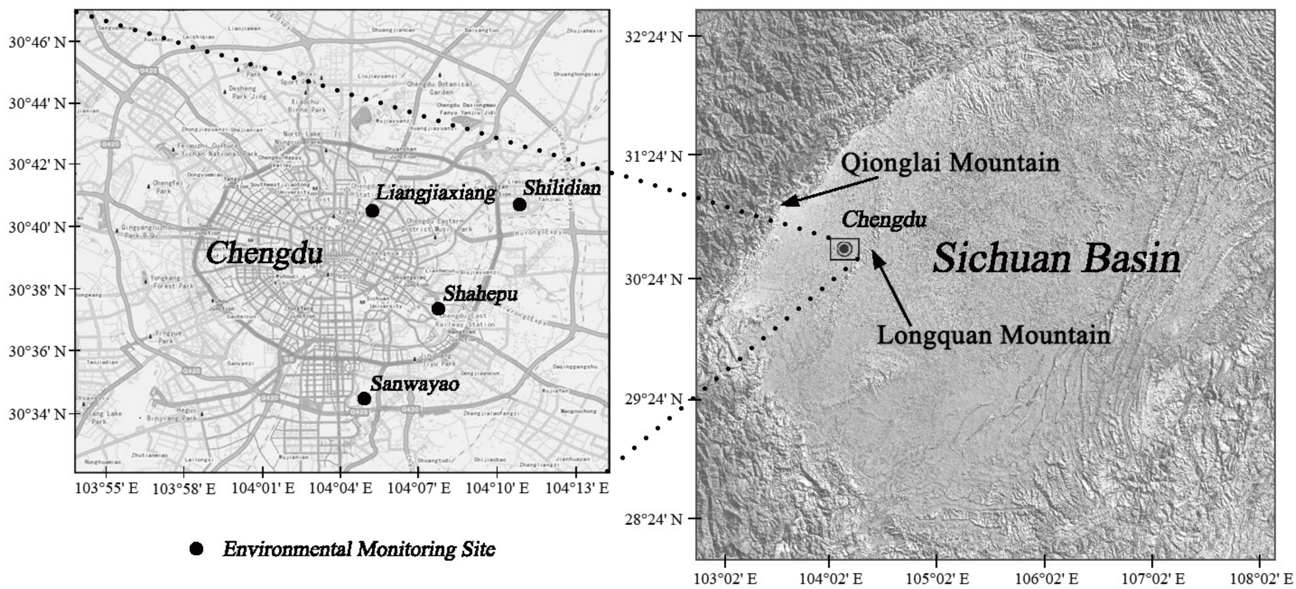

Fig. 1. Location of Chengdu in Sichuan Province, China, and topography of the Sichuan Basin.

scaling exponent does not completely characterize the series, since many subsets of the series have different scaling behaviour. This suggests different types of correlations at small and large time scales. Multifractal detrended fluctuation analysis (MF-DFA) (Kantelhardt *et al.*, 2002) can identify the different long range correlations and multifractal characterization embedded in the nonstationary time series. If the time series are long-range power-law correlated, the detrended fluctuation function.

$$F_q(s) = \left\{ \left[F^2(s) \right]^{q/2} \right\}^{1/q} \sim s^{h(q)} \tag{1}$$

where s is the length of each segments and $F^2(s)$ is the variance of the detrended time series in a given segment after removing a linear trend, while $h(q)$ is the generalized Hurst exponent. When $q = 2$, the scaling exponent $h(2)$ represents the well-known Hurst exponent. The scaling exponent $h(2) = 0.5$ shows a wholly stochastic process lacking correlation. $h(2) > 0.5$ indicates persistent long range correlation, meaning that large (small) values are more likely to be followed by large (small) values. For further detail computation, see Movahed *et al.* (2006). The $h(q)$~q relation is characterized by the typical multifractal form, monotonically decreasing with the increase of q. The singularity spectrum quantifies in details the long range correlation properties of a time series. If $h(q)$ is independent from q, which characterizes monofractal series.

A significant dependence of $h(q)$ on q indicates the different scaling behaviour of small and large fluctuations. For positive q, $h(q)$ describes the scaling behaviour of the segments with large fluctuation. In generally, large fluctuations are characterized by a smaller scaling exponent $h(q)$ for multifractal time series. For negative q, the scaling exponent $h(q)$ describes the scaling behaviour of segments with small fluctuations. In generally, small fluctuations are characterized by a larger scaling exponent $h(q)$ for multifractal time series. So the multifractality gives information about the relative importance of various fractal exponents present in the time series. The strength of the multifractality of a time series can be characterized by

$$\Delta h = h_{max}(q) - h_{min}(q) \tag{2}$$

Where Δh is the range of generalized Hurst exponents $h(q)$. The greater are Δh, the stronger is the degree of multifractality, and vice versa (Luciano and Vincenzo, 2006).

The Model of Self-Organized Criticality for $PM_{2.5}$

We attempt to find the answers to the following questions: how does long-range correlation and statistical distribution in $PM_{2.5}$ evolution emerge? It is inviting to do it in a self-organized criticality (SOC) framework which was specially designed to model the dynamics of complex systems. We conjecture the evolutions of $PM_{2.5}$ are similar to that of the sand-piles, and are governed by similar SOC dynamics.

Based on the SOC theory, we constructed a modified version of the original BTW sandpile model by Bak *et al.* (1987) for $PM_{2.5}$ evolution. This simplified model contains three dynamical mechanisms. These mechanisms include driving mechanism (the formation of primary and secondary $PM_{2.5}$), redistribution and relaxation mechanism (diffusion and transportation of atmospheric pollutants), temporal degradation mechanism (self-purification of atmospheric environment).

The model is defined here on a square lattice of size $L \times L$. Variables $h(i, j)$ represent amounts of the $PM_{2.5}$ pollutants at the site(i, j). We use open boundary conditions.

Driving mechanism: Some studies (Tao *et al.*, 2013) showed that the four sources, including secondary sulfate and nitrate, motor vehicle emissions, soil dust and biomass burning, contributed 24.6%, 18.8%, 23.6% and 33.0% to the total $PM_{2.5}$ mass in Chengdu. We consider that a "pollutant" sand, including primary and secondary $PM_{2.5}$ mass, is randomly added to a site. This process corresponds to the formation and emission of primary and secondary $PM_{2.5}$ at atmospheric system. The continuously emitted pollutants serve as the sands continuously dropped on a lattice. This is the direct driving force of the dynamical system.

$$h(i, j) = h(i, j) + \Delta h \tag{3}$$

where Δh is the amount of the formation and emission of primary and secondary $PM_{2.5}$ at site (i, j).

Redistribution and relaxation mechanism: when the amounts of PM_{10} pollutants at some site (i, j) reach some threshold magnitude h_c, the site becomes unstable or critical. Thus the pollutants at some site (i, j) will diffuse outwards into the surrounding sites by diffusion or convection. So the site relaxes by a toppling process. Of course, in this process, some PM_{10} pollutants will stay at the original site. The proportion of the pollutants at (i, j) is set 1/5. In our case, the prevailing wind direction in Chengdu is from the south. The average wind speed is 1.6 m/sec. By the effect of wind, pollutants at (i, j) will be transported to the downwind sites, including $(i + 1, j)$, $(i + 1, j + 1)$ and $(i + 1, j – 1)$, with each for about a third. So the redistribution and relaxation rules embody downwind transport of air pollutant. The rules are

$$h(i, j) \rightarrow \frac{\Delta h}{5} \tag{4}$$

$$h(i+1, j) \rightarrow h(i+1, j) + [h(i, j) + \Delta h - \frac{1}{5}\Delta h] \times \left(\frac{1}{3} - \sigma\right) \tag{5}$$

$$h(i+1, j+1) \rightarrow h(i+1, j+1) + [h(i, j) + \Delta h - \frac{1}{5}\Delta h] \times \left(\frac{1}{3} - \sigma\right) \tag{6}$$

$$h(i+1, j-1) \rightarrow h(i+1, j-1) + [h(i, j) + \Delta h - \frac{1}{5}\Delta h] \times \left(\frac{1}{3} - \sigma\right) \tag{7}$$

where σ is the loss proportion of $PM_{2.5}$ mass, owing to precipitation, adsorption and chemical action during transportation and diffusion of pollutants. This rule will be circulating running in accordance with the above method until a new stable configuration is reached, namely all $h(i, j) < h_c$. Avalanche size (s) is measured as the total number of toppling during an avalanche. In our model, the σ value is set to 0.01. So this model is local non-conservative.

Temporal degradation mechanism: All primary and secondary $PM_{2.5}$ mass will decay with time owing to self-purification of atmospheric environment at last. We simplify this process and presume that degradation of $PM_{2.5}$ follow the first level of decaying kinetics. So when a new stable configuration is reached after each relaxation rule, $PM_{2.5}$ mass at all sites will decay, namely.

$$h(i, j) \rightarrow h(i, j) \times e^{-k} \tag{8}$$

where k is related to $PM_{2.5}$ mass' own characteristics in atmospheric environment. After all lattice sites are stable, another grain of sand is added.

Continuing this adding and toppling processes and then measuring the cumulative probability distribution function $P(s)$ of avalanche sizes (s). If $PM_{2.5}$ evolution is an example a SOC process, the avalanche size distribution will follow power-law distribution. Note that what the avalanche threshold value represents in the sandpile model is not important (Bak e*t al.*, 1987; Turcotte and Malamud, 2004). The more important thing is its dynamics, in which an infinitesimally slow external driving of sand particles associated with a threshold rearrangement dynamics lead to a stationary state with activity (avalanches) distributed on all length scales. Thus, in a nondimensional formalism, we select $\Delta h = 1$ and $h_c = 4$ referring the classical BTW sandpile model.

RESULTS AND DISCUSSION

Results for Multifractal Characterization

Fig. 2 shows the q-dependence of the generalized Hurst exponent $h(q)$ for the $PM_{2.5}$ series of Shilidian, Sanwayao, Liangjiaxiang and Shahepu stations. All $h(q)$~q relations are characterized by the monotonically decreasing trends when q varies from –10 to 10. $h(q)$ are not a constant, indicating multifractality in these time series. The $h(2)$ values of the $PM_{2.5}$ series of these four stations are 1.17 (Shilidian), 1.15 (Sanwayao), 1.16 (Liangjiaxiang) and 1.20 (Shahepu) respectively. The $h(2)$ values are all larger than 0.5, implying the original $PM_{2.5}$ series are nonstationary signals with long range correlation. The long range correlation property signifies that the $PM_{2.5}$ concentration fluctuations in these stations, from small time intervals (one hour) to larger ones (up to 17 days at least) are positively correlated in a power-law fashion. For example, there is an increase tendency in $PM_{2.5}$ concentration to be followed by another increase in $PM_{2.5}$ concentration at a different time in a power-law fashion. In contrast, a randomly forced first-order linear system with the classical Markov-type stochastic behaviour should have fluctuations whose autocorrelation decays exponentially with lag time. This suggests that the correlations between the fluctuations in $PM_{2.5}$ concentration do not obey the classical Markov-type stochastic behaviour (exponential decrease with time), but display more slowly decaying correlations.

The width Δh is a measure of how wide the range of fractal exponents found in the series is. The Δh values of these four stations are 0.788 (Shilidian), 0.867(Sanwayao), 0.766 (Liangjiaxiang) and 0.739 (Shahepu) respectively. The high Δh values show that the probability distributions of the fluctuation are wider and the multifractality are very powerful.

Sources of Multifractality

Furthermore, we are interested in the nature of the multifractal behaviour of the $PM_{2.5}$ series. In general, there are two major sources of multifractality which can be found in the time series (Kantelhardt *et al.*, 2003; Shi *et al.*, 2013). One is different long-range correlations for small and large fluctuations. The other is fat tailed probability distribution of variations. Shuffling procedure (shuffled data) and phase randomization (surrogated data) are the main procedures to find the contributions of two sources of multifractality (Kwapien *et al.*, 2005). Shuffling procedure can destroy the temporal correlations and preserves the distribution of the original data. Therefore the shuffled time series will show mono-fractal scaling because all long-term correlations are destroyed by the shuffling procedure. So shuffling procedure can be applied to study the contributions of long-range correlation on the multifractality. In order to study the contributions of the fat tailed probability distribution on the multifractality, the surrogate data are used. Phase randomization procedure can eliminate the non-Gaussianity of the distributions, preserving only the linear properties of the original time series.

The $h(q)$ spectra have been shown for shuffled and surrogated data in Fig. 2. One can see that the q dependence of $h(q)$ for the original $PM_{2.5}$ time series is higher than the shuffled and surrogated data for these four stations respectively. The main feature of these plots is that the q dependence of $h(q)$ is lowest for the shuffled time series. The phenomenon indicates that the multifractality nature of the original $PM_{2.5}$ time series is mainly due to long-range correlation. At the same time, the q dependence of $h(q)$ for the surrogated data is much lower than the original time series. This shows the non-Gaussian probability distribution of $PM_{2.5}$ time series also partly contribute to the multifractal behaviour.

Time Dependent Multifractal Detrended Fluctuation Analysis of $PM_{2.5}$ Series

The multifractal parameters calculated over the whole series are not informative about the dynamical changes in the behaviour of $PM_{2.5}$ time series. Thus, what we need is a time-dependent analysis of multifractality. The temporal evolution of the multifractality can be calculated by the approach of a sliding window. In our case, we considered a sliding window of 72 data (namely 72 hours). The number of events in each window was chosen in order to have sufficient amount of points to perform the estimates. The shift between two successive windows was set to 1 event (namely 1

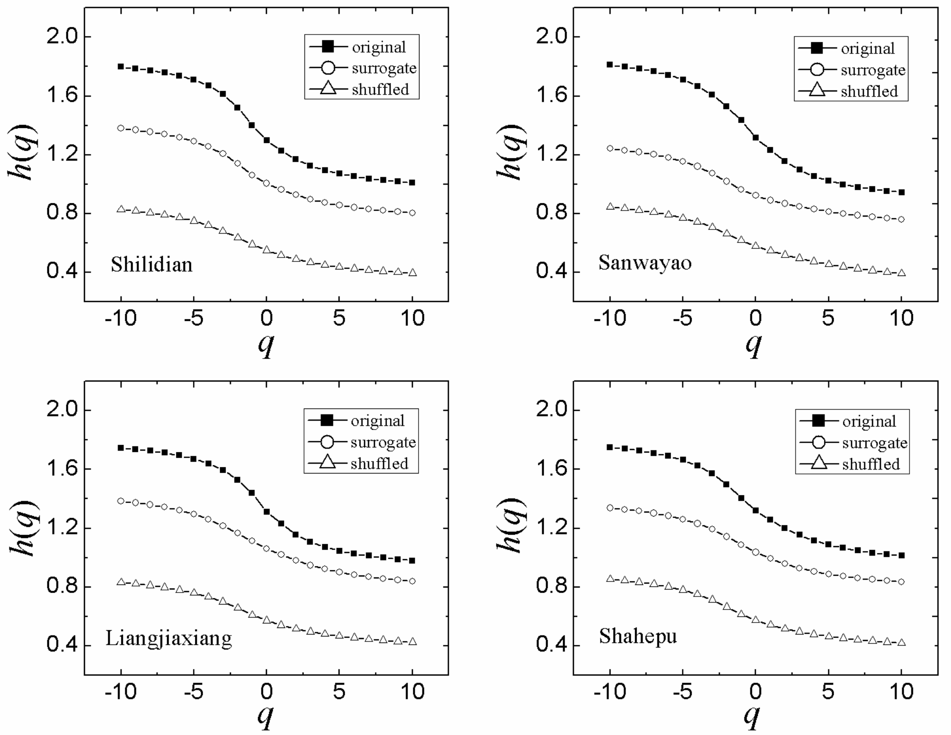

Fig. 2. Generalized Hurst exponent $h(q)$ as a function of q for the original, shuffled and surrogated $PM_{2.5}$ series of Shilidian, Sanwayao, Liangjiaxiang and Shahepu stations.

hour), in order to evaluate the variation of multifractality with a good time resolution. The multifractal parameters Δh can be calculated for each window. Thus each Δh value was associated to the occurrence time of the last $PM_{2.5}$ concentration in the sliding window.

Fig. 3 shows the time variation of the multifractal parameters Δh for the four stations. For all the four stations, they look like very low amplitude "ripples" propagating along the mean values. There are no significant differences among these time dependence of Δh during the whole haze period. It implied that the underlying mechanism of $PM_{2.5}$ fluctuations in the formation of multifractality is consistent during the haze period.

Scaling Relationships for $PM_{2.5}$ Series

The cumulative distributions for the relative $PM_{2.5}$ concentration fluctuations,

$$\Delta c = c_{n+1} - c_n \tag{9}$$

where c is the hourly average $PM_{2.5}$ concentration data and n is time order, are plotted in Fig. 4.

Fitting by the least square method, the laws shows the relative $PM_{2.5}$ concentration fluctuations follow power-law distributions. They have the similar form

$$N(\Delta c \geq \Delta c_0) \propto \Delta c^{-\tau} \tag{10}$$

where $N(M)$ is the cumulative number of $PM_{2.5}$ concentration events with a magnitude greater than Δc, τ is the scaling exponent. The τ values of these four stations are 1.677 (Shilidian), 0.983 (Sanwayao), 1.796 (Liangjiaxiang) and 2.166 (Shahepu) respectively. The departure from Gsussian behaviour is evident. This result implies that the $PM_{2.5}$ concentration fluctuations follow scale invariance pattern, and the typical $PM_{2.5}$ concentration fluctuation value does not exist. For the different stations, power law scaling is ubiquitous and robustness. The different scaling exponents for the four stations indicate the spatial variability of $PM_{2.5}$ concentration.

The Simulation of Self-Organized Criticality of $PM_{2.5}$

The SOC state is stationary when the constant average height of the sandpile at which the current of influx of sand to the system is equal to the current of outflux of the sand at the open boundary. In order to achieve this steady state, the first 10^6 avalanches are skipped. The simulated data are generated on a square lattice of size 50 × 50 in runs of 10^7 avalanches. The simulations are performed for 50 × 50 lattice sizes. We found that this model exhibit SOC behaviour for a wide range of k values. In a wide range of k values, the cumulative probability distribution function $P(s)$ of avalanche sizes (s) follows a power-law relation $P(s > s_0) \propto s^{-\alpha}$, where α is the scaling exponent.

The simulated results of avalanche size distributions are shown in Fig. 5. We have found that when $k = 2.51 \times 10^{-4}$, 7.49×10^{-5}, 2.57×10^{-4}, 2.76×10^{-4}, the simulated α values of these four stations are 1.677 (Shilidian), 0.983 (Sanwayao), 1.796 (Liangjiaxiang) and 2.166 (Shahepu) respectively. These results are in agreement with the observations. Thus, the model can generate the highly significant ($R^2 > 0.99$) power law distributions of $PM_{2.5}$ fluctuations. The ability of a simple cellular automata model of $PM_{2.5}$ evolution to generate SOC behaviour suggests that real-world $PM_{2.5}$ evolution might be an SOC system. The simulated α values depends on k. The dependence was shown in Fig. 6. The relation between α and k is a simple exponential relation, i.e., $\alpha \propto 0.013e^{16527.4k}$. In Fig. 6, the arrows indicate the actual measured scaling exponent values ranges for $PM_{2.5}$ fluctuations in the four stations.

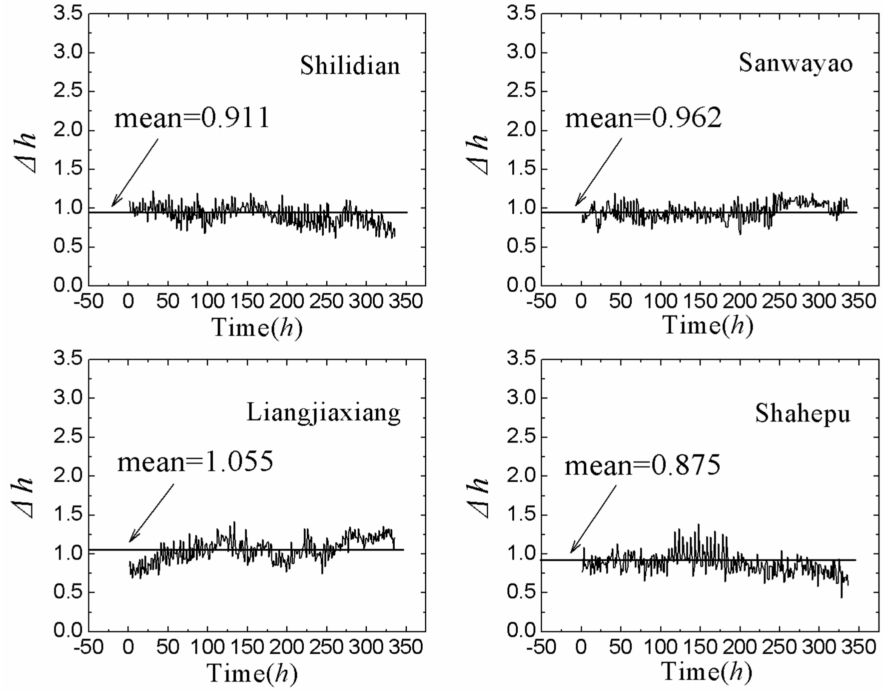

Fig. 3. The time variation of Δh for Shilidian, Sanwayao, Liangjiaxiang and Shahepu stations.

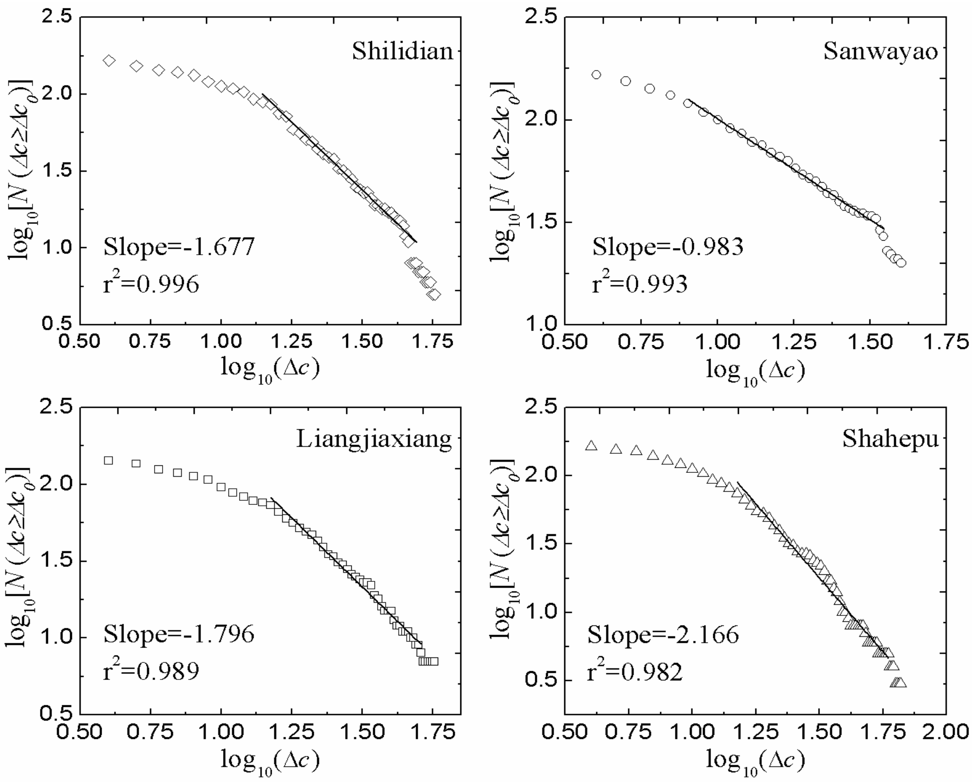

Fig. 4. The cumulative distributions for the relative $PM_{2.5}$ concentration fluctuations for Shilidian, Sanwayao, Liangjiaxiang and Shahepu stations.

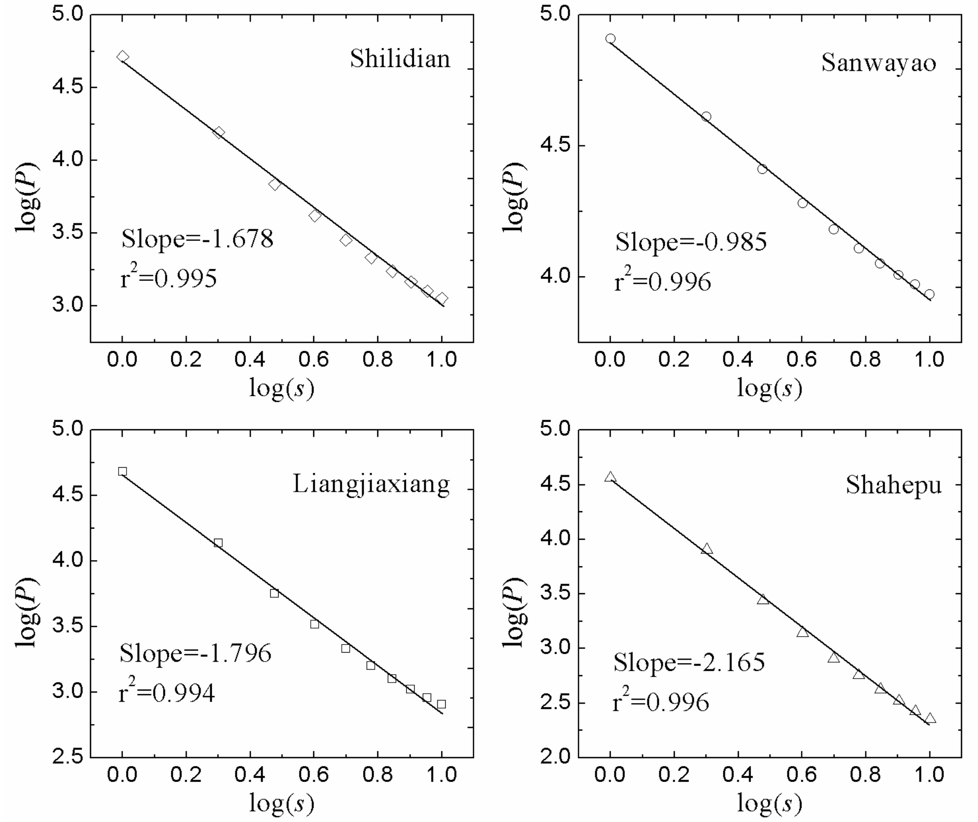

Fig. 5. Avalanche size distribution for $PM_{2.5}$ evolution model.

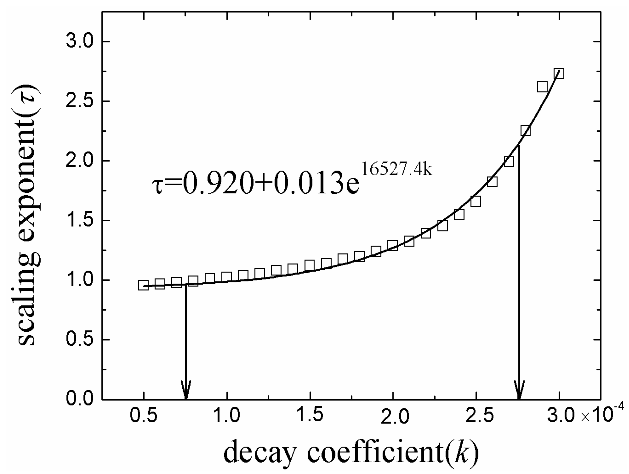

Fig. 6. The relation between the simulated scaling exponent (τ) values and k.

Although the two main sources of multifractality of $PM_{2.5}$ fluctuations, i.e., long-range correlation and non-Gaussian probability distribution have been examined, the origin of long-range correlation and non-Gaussian probability distribution of $PM_{2.5}$ remains unclear. Some studies (Shi and Liu, 2009) suggest that the presence of long-range correlation and scale invariance property in air pollution may be attributed to self-organized criticality.

The concept of self-organized criticality (SOC) has been introduced by Bak *et al.* (1987) to provide a framework of explaining $1/f$ noises and scale invariance, which are widespread in nature. Complex systems, such as sandpile, contain a number of non-linear and short-range interacting elements. The systems may naturally evolve toward a critical state by slow driving or energy input. When a system reaches the critical state, long-range correlation interacting will emerge. The critical state is an ensemble of metastable configurations and the system evolves from one to another event via an avalanche-like dynamics. These avalanche events demonstrate a power law distribution on the temporal-spatial scale (Turcotte and Malamud, 2004). However, there has been no specific investigation into whether $PM_{2.5}$ fluctuations exhibit SOC behaviour.

Naturally, the external conditions, such as meteorological and topographical conditions, have some strong effects on $PM_{2.5}$ fluctuations (Shen *et al.*, 2009; Ram and Sarin, 2011). However, when these external conditions maintain relative stability, the internal dynamical mechanism of $PM_{2.5}$ fluctuations is determining factor. In our case, the chosen time period is a typical haze period in Chengdu. The property of atmospheric boundary layer at Chengdu city (30.67°N, 104.06°E) is relatively stable. Since the changes of meteorological condition and underlying surface are small, these external conditions are inadequate to explain the complex dynamical processes of $PM_{2.5}$ fluctuations.

SOC theory in the context of $PM_{2.5}$ fluctuations can provide valuable description and explanation. One can compare $PM_{2.5}$ evolution processes to a sand grain which falls on a sand pile in critical regime. Firstly, the sand grains play the role of the primary and secondary $PM_{2.5}$. $PM_{2.5}$ pollutants forms mainly as a result of first and (or) secondary pollutants produced from the emission of air pollution sources. The continuously $PM_{2.5}$ pollutants emission in atmospheric environment can serve as the grains continuously dropped on a pile. Secondly, the chain of forces among sand grains represents the diffusion and transportation of $PM_{2.5}$ mass. Thirdly, the normal atmospheric environmental capability serves as the critical state. When the amounts of microscopic condensed $PM_{2.5}$ pollutants reach some threshold magnitude, the pollutants can be transported on microscopic scales by diffusion or convection. These masses reach a new location, where the local $PM_{2.5}$ concentration is lower, and can be diluted. Conversely, if the local $PM_{2.5}$ pollutants concentration in the neighborhood is higher, the amount of condensed pollutants will increase. Once the system reaches some critical point, any small perturbation, in principle, can trigger a chain reaction like the avalanches in atmospheric system. If the local $PM_{2.5}$ pollutants concentration is higher than the some critical value, pollutants are assembled and precipitated in the atmosphere. Thus, the system will adapt itself by removing these dissidents to maintain the critical state just as the sandpile adapts itself by avalanching to reach its constant angle of repose. Therefore, we can define the fluctuations of $PM_{2.5}$ concentrations as avalanches events in a SOC sandpile. At the critical state, long range correlation and scale-invariant of $PM_{2.5}$ concentrations will emerges from the dissipative atmosphere system.

This remarkable similarity between $PM_{2.5}$ fluctuations and sand pile is also interesting in terms of theoretical description. Our theoretical results suggest that such complex $PM_{2.5}$ interactions between different areas may underlie the complex temporal architecture of $PM_{2.5}$ fluctuations. The simulated results have shown that branched avalanches of conformational reorganizations, caused by local random perturbations can lead to long-term correlation in the dynamics of the system. This model naturally evolves to the state, where no characteristic scale can be seen, and the system operates in the state of self-organized criticality. $PM_{2.5}$ fluctuations, as critical systems, exhibit invariant scale behaviour meaning that big and small $PM_{2.5}$ fluctuations have the same significance.

In this paper, the average wind speed is small. This is the typical meteorological features in Chengdu city. In the Chengdu city, the change of surface wind speed is small and the annual average wind speed is only 1.2 m/sec, with high static wind frequency, up to 45%–50%. So a haze episode in Chengdu city may be mainly caused by local emissions rather than transport from other cities. Thus, this simple model can provide the basic explanation of how does the long-term correlation and invariant scale in the $PM_{2.5}$ fluctuations dynamics emerge. Of course, in the other region, a haze episode may be caused by pollution transport among cities in the region. For example, the air pollutants of Zhoushan city (29°32′–31°59′N, 121°30′–123°25′E) are contributed mainly by transport from other cities in the Yangtze River Delta rather than local emissions. Shi *et al.* (2014) developed a self-organized criticality (SOC) based trans-boundary PM_{10} model. Simulations showed the self-organized criticality mechanism is still the key physical process governing PM_{10} pollution evolution in higher surface wind speed. So we believe that the self-original criticality will be useful in understanding $PM_{2.5}$ evolution dynamics and the origin of long-term correlation and invariant scale of $PM_{2.5}$.

CONCLUSIONS

Air pollution phenomenon underlying $PM_{2.5}$ concentrations evolution is very complex. During a typical haze period, the scaling and multifractal properties of $PM_{2.5}$ concentrations series of the four monitoring stations in Chengdu have been analyzed by MF-DFA. The empirical evidence from MF-DFA confirmed that there exist long-term correlation and multifractality in $PM_{2.5}$ concentrations evolution. By shuffling procedure and phase randomization procedure, it is found that main sources of the multifractality in these $PM_{2.5}$ series is long-range correlation. At the same time, the non-Gaussian probability distribution also partly contributes to the multifractal behaviour. For all the four stations, there are no significant differences in the time dependence of the multifractal parameters Δh during the whole haze period. It implied that the underlying mechanism of $PM_{2.5}$ fluctuations in the formation of multifractality is consistent during the haze period. The scale-free power laws behaviour are found to govern the cumulative distributions statistics for $PM_{2.5}$ concentration fluctuations. In order to explain how long-range correlation and power-law distribution in $PM_{2.5}$ evolution emerges, a novel $PM_{2.5}$ evolution model is developed on the bases of SOC theory. The main mechanism includes the formation of primary and secondary $PM_{2.5}$, diffusion and transportation of atmospheric pollutants, self-purification of atmospheric environment. The model is a continuous, directional, non-conservative, attenuated cellular automation modelling, which shows robust SOC behaviour. We have shown that a simplest possible mechanism for $PM_{2.5}$ evolution is sufficient to recover essentially long-range correlation and power-law distribution documented in $PM_{2.5}$ concentrations of Chengdu city. The high correspondence of the results to observations indicates that the model provide an effective parameterization of the key physical process that govern $PM_{2.5}$ fluctuation in the whole haze period.

ACKNOWLEDGMENTS

The work is supported by the National Natural Science Foundation of China (No. 41465010), Hunan Provincial Natural Science Foundation of China (No. 13JJB012), Scientific Research Fund of Hunan Provincial Education Department (No. 13B089), and Key Laboratory of Ecotourism Hunan Province (No. JDSTLY1408).

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Received for review, May 23, 2014
Revised, July 12, 2014
Accepted, October 16, 2014