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Spatial statistics of atmospheric particulate matter in China

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HIGHLIGHTS

- Particulate matter concentrations in China were studied in a multiscale view.
- Logarithm spatial correlation pattern was retrieved experimentally.
- Multifractality nature was identified.

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ABSTRACT

In this paper, the spatial dynamics of the atmospheric particulate matters (resp. PM_{10} and $PM_{2.5}$) are studied using turbulence methodologies. It is found experimentally that the spatial correlation function $\rho(r)$ shows a log-law on the mesoscale range, i.e., $50 \leq r \leq 500$ km, with an experimental scaling exponent $\beta = 0.45$. The spatial structure function shows a power-law behavior on the mesoscale range $90 \leq r \leq 500$ km. The experimental scaling exponent $\zeta(q)$ is convex, showing that the intermittent correction is relevant in characterizing the spatial dynamic of particulate matter. The measured singularity spectrum $f(\alpha)$ also shows its multifractal nature. Experimentally, the particulate matter is more intermittent than the passive scalar, which could be partially due to the mesoscale movements of the atmosphere, and also due to local sources, such as local industry activities.

1. Introduction

In recent decades, many cities in China have experienced heavy air pollution episodes leading to negative impacts on human health (Streets and Waldhoff, 2000; Chan and Yao, 2008; Matus et al., 2012; Chen et al., 2013; Wang et al., 2014b, c; Zhang et al., 2014; Rohde and Muller, 2015), and the air pollution have been one of the biggest problems in urban areas of many megacities in China. The Jing-Jin-Ji region, Yangtze River Delta region, Pearl River Delta region, Central China region and Cheng-Yu region, to list a few, are the major polluted regions in China due to highly densed population and high energy consumption. In order to improve the air quality, China government issued new national ambient air quality standards in 2012 and were to be implemented in Jan. 2016. According to the new standards, air quality indices ranging from 0 to 50 and ranging from 51 to 100 represent excellent and good, respectively. However, air quality index equal to or above 101 means the air quality does not meet the national ambient air quality standards. Hourly observed concentration data for pollutants in numerous cities were released by the government. According to the new ambient air quality standards, only 8 out of China's 74 biggest cities met the government's air quality standards in 2014. Although not by much, the air quality in 2014 was better than in 2013: see http://www.mep.gov.cn/. Particulate matter with an aerodynamic diameter 10 μm or less, or PM_{10} usually dominantes pollution episodes caused by dust storms. Particulate matter with a diameter less than 2.5 μm, or $PM_{2.5}$ usually could lead to more serious health problems for local residents than coarse particle due to easier inhalation. Furthermore, most haze episodes occurring in China are characterized by high concentrations of $PM_{2.5}$ in the ambient air.

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Rohde and Muller (2015) applied the Kriging interpolation to four months of data to retrieve the pollution maps for eastern China and discovered that the greatest pollution occurs in the east. Air pollution episodes can cover a large region and are particularly intense in a northeast corridor that extends from outside of Shanghai to north of Beijing. Particulate matter is a very complicated mixture that comes from numerous emission sources. Industrial process was the dominant local contributor to PM2.5 concentration in the whole city of Shanghai except at the urban center where vehicle emissions contribute slightly more (Wang et al., 2014c). Moreover, haze episode could be caused by the combination of anthropogenic emissions, unusual atmospheric circulation, the depression of strong cold air activities, and weak boundary layer ventilation (Wang et al., 2014a). Wang et al. (2014b) concluded that the response of $PM_{2.5}$ to meteorology possibly changes a feedback loop whereby planetary boundary layer dynamics amplify the initial perturbation of $PM_{2.5}$.

Note that the air pollution occurred in the planetary boundary layer, where atmospheric turbulence is involved, showing a significant impact on the transport and dispersion of pollution matter. However, the effects of atmospheric turbulence are seldom studied in a multiscale view. A common behavior of the turbulence is the multiscaling, or multifractality, of the velocity field (Frisch, 1995). In the view of the hydrodynamic turbulence, a large range of spatial and temporal scales/freedoms are involved, resulting in a cascade process in which the energy transfers from large-scale structures to small-scale ones until the fluid viscosity converts the kinetic energy into heat. This phenomenological Kolmogorov-Richardson energy cascade picture has been widely and successfully applied in multiple disciplinary fields, such as financial activity (Schmitt et al., 1999; Ghashghaie et al., 1996; Li and Huang, 2014), crack of rock surfaces (Schmittbuhl et al., 1995), rainfall patterns (Tessier et al., 1996), etc.

Specifically for atmospheric turbulence, due to the geometrical constrain of the atmospheric movement, there exists several typical spatial scales. These include the microscale (resp. 1 km or less), showing three-dimensional property and synoptic scale (resp. up to 1000 km), showing a two-dimensional feature (Vallgren et al., 2011). Between the microscale and synoptic scale, a large range of scale motion exists in mesoscale structures (resp. from few dozens of km to few hundreds km). The famous Kolmogorov 5/3-law has been observed on the mesoscale range (Nastrom et al., 1984; Nastrom and Gage, 1985) and agrees well with the above mentioned Kolmogorov-Richardson cascade prediction (Vallgren et al., 2011). Therefore, the air pollution indices, such as $PM_{2.5}$ could display a spatial scaling behavior since they are advected mainly by these mesoscale structures. In this paper, we employ the standard structure function analysis to retrieve the multiscale and multiscaling properties of the PM_{10} and $PM_{2.5}$ to show the impact from the mesoscale atmospheric turbulence.

2. Data

The hourly concentrations of PM2.5 and PM10 were released by the government (http://www.cnemc.cn). We processed these data into daily average concentrations to be used in this study. There are 305 monitor stations belonging to different cities. Fig. 1 a) shows the spatial distribution of these monitor stations, which were monitored during the period from 31 Dec. 2013 to 01 Mar. 2015, corresponding to 425 days for most of cities with several missing. In total, there are 82,755 daily averaged data points. The neighbor distance, r, of two cities is calculated via a great circle distance algorithm. The corresponding probability density function (pdf) is shown in Fig. 1b). For convenience, we used the logarithm of r, $x = \log_{10}(r)$. A bin width 0.1 in the logarithm scale was adopted to estimate the pdf. It is interesting to note that the measured pdf agrees well with the Bramwell-Holdsworth-Pinton (BPH) formula (Bramwell et al., 1998), which is:

$$\Pi(y) = K\left(e^{y-e^{y}}\right)^{a},\ y = b(x-s),\ a = \pi/2, \tag{1}$$

where parameters $b = 0.938$, and $K = 2.14$ were obtained numerically (Bramwell et al., 2000). Note that this formula was first introduced to characterize rare fluctuations in turbulence and critical phenomena. The neighbor distance was often chosen based on cities located near a water source. Therefore, this neighbor distance could be used as a proxy of the spatial distribution of water sources. However, this postulate needs to be verified by carefully analyzing neighbor distance statistics for different regions. Fig. 2 shows the recorded $PM_{2.5}$ index with unit $\mu g/m^3$ for three typical cities, Beijing, Shanghai and Xiamen. Visually, the measured index shows similar evolution trends: they are higher during the winter and smaller during the summer, showing an annual cycle. In the following analysis, these database are analyzed by pairing two cities, i.e., $[\theta_i(t), \theta_j(t)]$ with the neighbor distance r_{ij}, where $\theta_i(t)$ is the air quality index of the ith city.

3. Results

3.1. Spatial correlation

We first calculated the spatial correlation function for different neighbor distances r. The spatial correlation $\rho(r)$ is defined by the following equation:

$$\rho(r) = \frac{1}{N(r)}\sum^{N(r)} \frac{\left\langle \tilde{\theta}_i(t)\tilde{\theta}_j(t) \middle| r_{ij} = r\right\rangle_t}{\sigma_i\sigma_j}, \tag{2}$$

where $\tilde{\theta}_i(t) = \theta_i(t) - \langle\theta_i(t)\rangle_t$ is the centered index of the ith city, $\langle\ \rangle_t$ is the time average, σ_i is the standard deviation, r is the neighbor distance; and $N(r)$ is the number of pairs with distance r, where a bin width 0.1 in the logarithm scale is used. The final $\rho(r)$ is then calculated for all pairs of cities with distance r. Fig. 3 a) shows the measured $\rho(r)$ in a semilog plot for the $PM_{2.5}$ (○) and PM_{10} (□). A log-law is observed in the range $50 \le r \le 500$ km, as follows:

$$\rho(r) \propto A - \beta\log_{10}(r), \tag{3}$$

where β is the scaling exponent, which is experimentally $\beta = 0.45 \pm 0.02$. To emphasize the experimental log-law behavior, Fig. 3b) shows the corresponding compensated curve using the fitted parameters. A clear plateau confirms the existence of the log-law. Note that the log-law range is between the microscale (resp. 1 km or less) and synoptic scale (resp. up to 1000 km), corresponding to the mesoscale movement in the atmospheric boundary layer (Vallgren et al., 2011).

3.2. Structure function analysis

Intermittency or multifractality is an important feature of the turbulent-like dynamical systems (Frisch, 1995). More precisely, numerous spatial or temporal freedoms exist simultaneously and interact with each other to transfer energy, momentum, or other physical quantities. To characterize this multiscale interaction, structure function analysis is used to retrieve the scale invariance for high Reynolds turbulent flows (Kolmogorov, 1941). It is then widely used in a variety of fields, including financial activity (Schmitt et al., 1999; Ghashghaie et al., 1996; Li and Huang, 2014), crack of rock surfaces (Schmittbuhl et al., 1995), rainfall patterns