Atmospheric Research 154 (2015) 191–203

Contents lists available at ScienceDirect

Atmospheric Research

journal homepage: www.elsevier.com/locate/atmos

Multifractal analysis of atmospheric sub-micron particle data

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ARTICLE INFO

Article history:
Received 3 July 2014
Received in revised form 10 November 2014
Accepted 10 November 2014
Available online 20 November 2014

Keywords:
Atmospheric particles
Multifractal analysis
Singularity spectra
Hölder exponent

ABSTRACT

Multifractal analysis was used to describe air pollution by sub-micrometric atmospheric particles. Atmospheric particle concentrations were studied from March 31 to April 21, 2006, as part of the MILAGRO campaign at the Jasso Station by means of an SMPS. Sixteen campaign days were selected to carry out the multifractal analysis of the experimental data through Singularity Spectra f(α). In this work, the roughness/smoothness feature of atmospheric particle distributions was studied by means of the Hölder exponent (α), which can be associated with the intensity of particle emissions through time and the randomness of the external emission sources. Multifractal analysis has been found to be a useful tool to establish intensity fluctuations of atmospheric data.

1. Introduction

Air pollution has been a major concern around the world for some time now. The conduction of air pollution studies is paramount in order to understand this complex phenomenon, which includes different variables such as particle size, particle formation processes, particle sources, and particle size distributions in the atmosphere. These studies in combination with chemical studies enable researchers to elicit the behavior and the hazardous nature of the atmospheric particles and their effect on the human health. In Mexico, some earlier atmospheric particle studies include: 1) the Mexico City Air Quality Research Initiative (MARI), 1990 (Streit and Guzman, 1996), 2) the "Investigación sobre Materia Particulada y Deterioro Atmosférico-Aerosol" and Visibility Evaluation Research (IMADA-AVER), 1997 (Edgerton et al., 1999), 3) Exploratory field measurements in Mexico City Metropolitan Area, MCMA-2003 (Molina and Molina, 2002; Molina et al., 2007), and 4) the Megacity Initiative: Local and Global Research Observations (MILAGRO), 2006. During the MILAGRO campaign, field measurements of atmospheric pollutants were carried out in Tula (Hidalgo State) in order to collect atmospheric data and study the behavior and potential effects of these pollutant emissions on Mexico City (Sosa et al., 2013).

Most atmospheric projects have been focused on the physicochemical study of atmospheric particles and the application of models to describe their behavior over and throughout different geographical areas. Among the models used to study atmospheric particles we find the Fractal Theory, which has proved its capability to describe the dynamics of complex phenomena from the very beginning. In the case of aerosols, Fractal Theory has been applied to describe their behavior (Arizabalo et al, 2011; Sosa, 2002; Guzmán et al., 2002), where the concepts that played a major role were the *Fractal Dimension*, the *Hurst Coefficient* and *Lacunarity*. Through these three concepts, it is possible to decide if a system is fractal (Fractal Dimension), rough or smooth (Hurst Coefficient) and homogeneously or heterogeneously distributed (Lacunarity). Unfortunately, the Fractal Models present some limitations as compared to the more general *Multifractal Models*, which can provide more information to describe a physical system. The methodology of Multifractal Analysis introduces the Hölder exponent (α) and singularity spectra f (α), which in the case of

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http://dx.doi.org/10.1016/j.atmosres.2014.11.008

atmospheric studies are the necessary tools for performing the multiscale analysis of the total concentration of particle distributions. In the multifractal approach the local singularities are characterized by means of the singularity spectrum and the general structure is characterized by the singularity spectra. The multifractal singularity spectra offer general information about the global texture, and in the case of the distribution of the particle concentration with respect to the local time, a more evident irregularity in the time series can be observed, mainly at the hours of highest industrial activity and traffic throughout the day. Multifractals are non-uniform fractals, which in contrast with uniform fractals present local density fluctuations (Korvin, 1992; Posadas et al., 2003).

The study of the multiscale problem regarding atmospheric particles is based on the underlying growing mechanisms of submicron particles. These complex mechanisms depend on several variables such as particle chemical composition, gas phase concentration and meteorological variables, among others. It is known that after the nucleation process, the formation of nanoparticles stems from the coagulation processes. According to González and Rodríguez (2013), the SO_2 emissions from power plants and refineries provoke that the concentrations of submicrometric particles in the air feature peaks remarkably above the average concentrations related to vehicle emissions (Dahl et al., 2006), however, the last ones can represent up to 85% of the concentrations of ultrafine particles in urban zones for 24-h periods (Mbengue et al., 2014). In addition, the submicrometric particles can also contain ammonium sulfate and soot mixtures, which affects directly the shape and growth of the concentrations of cloud drops (Pósfai et al., 2013). At the same time, the secondary particles containing sulfate, ammonium, nitrate, and secondary organic aerosols are present mainly in the vertical concentration profiles, whereas the black carbon and organic carbon particles remain on the ground surface (Ma and Yu, 2014), all of them affecting the integral morphology of aerosols.

The collision and coalescence processes are fundamental for determining the shape and size of the particles for at high temperatures, coalescence prevails over the collision processes, producing large and spherical particles. On the other hand, at low temperatures, the frequency of the particle collisions prevails over coalescence, which leads to the formation of small particle clusters. As for nanoparticles, the diffusion processes control the collision–coalescence mechanisms, which occur mainly when small particles collide and get adhered to large ones. These processes are faster when the size difference among the coalescent particles is higher (Seinfeld and Pandis, 1998, Zeng et al., 1998, Hawa and Zachariah, 2006).

Once the primary particles are formed, mainly by Brownian movement, the collision among such particles occurs, which are kept together by Van der Waals forces, leading to the formation of clusters or agglomerates (Matsoukas and Friedlander, 1991).

At high supersaturation levels, condensation processes take place, which prompt the spontaneous formation of Aitken nuclei. However, in the absence of supersaturation, these nuclei can develop themselves efficiently when the molecules of low volatility organic compounds, present in the air, interact with solar radiation or gases such as NO_2, which promotes the condensation among themselves or on surfaces (Went, 1966). Since 1966, it was already known that the Aitken nuclei can be formed spontaneously when there are molecule supersaturation conditions or when there are volatile organic compounds (VOCs) with low volatility that cannot stay as individual molecules, which condensate either on surfaces or on other molecules (Went, 1966). Equally, in zones close to refineries like the oil refinery and the thermoelectric power plant in Salamanca City (Guanajuato State, Mexico), provoke high VOC concentration events, which consist mainly of 11% halogenated, 13% aromatics, 40% alkanes and 11% olefins, among others (Vega et al., 2011).

Pöschl et al. (2010) showed that in rain forests, the submicrometric particles consist mainly of secondary organic material, which has been formed by the oxidation of gaseous biogenic precursors. In addition, the concentrations of secondary organic aerosols can be affected mainly by oxidation products of isoprene, sugar, sugar alcohols, trace gases such as SO_2, NO_x, O_3, NH_3, the acidity of aerosols and organic carbon (Li et al., 2013), which affect the size and shape of particles. In urban zones, the formation of submicrometric particles that contain sulfates is related to conversion processes from gas to particle by means of

Table 1
Month day, local mean particle distribution, average particle diameter, particle formation process, α_{min} and α_{max}, $\Delta\alpha$ and figure locations.

Month day 2006	Local mean particle distribution [particle/cm^3]	Average particle diameter [nm]	Particle formation process	α_{min}	α_{max}	$\Delta\alpha$	Figures
March 31 (Friday)	77,333	38.5	Aitken	0.0272	0.7307	0.7035	1, 2(a), (b)
April 1 (Saturday)	52,998	15.7	Nucleation	0.0127	0.2677	0.2550	1, 2(a), (b)
April 4 (Tuesday)	61,602	24.1	Aitken	−0.2234	1.3217	1.5451	1, 2(a), (b)
April 6 (Thursday)	11,433	16.8	Nucleation	−0.6412	−0.3277	0.3135	1, 2(a), (b)
April 7 (Friday)	14,455	21.7	Aitken	−0.3156	0.1594	0.4750	3, 4(a), (b)
April 8 (Saturday)	14,868	76.4	Aitken	−0.2989	0.0419	0.3408	3, 4(a), (b)
April 9 (Sunday)	23,868	35.9	Aitken	−0.0734	0.4706	0.5440	3, 4(a), (b)
April 10 (Monday)	22,762	21.7	Aitken	−0.1279	−0.0504	0.0775	3, 4(a), (b)
April 11 (Tuesday)	23,072	46.1	Aitken	−0.4029	0.1703	0.5732	5, 6(a), (b)
April 13(Easter Thursday)	17,876	15.7	Nucleation	−0.9051	−0.0725	0.8326	5, 6(a), (b)
April 15 (Saturday)	15,675	18.8	Nucleation	−0.7095	0.5936	1.3031	5, 6(a), (b)
April 17 (Monday)	10,382	37.2	Aitken	0.0077	0.0899	0.0822	5, 6(a), (b)
April 18 (Tuesday)	11,984	15.7	Nucleation	0.2332	0.7558	0.5226	7, 8(a), (b)
April 19 (Wednesday)	14,100	16.3	Nucleation	0.0497	0.4091	0.3594	7, 8(a), (b)
April 20 (Thursday)	11,585	41.4	Aitken	−1.0241	0.0454	1.0695	7, 8(a), (b)
April 21 (Friday)	19,790	21.7	Aitken	−0.0131	0.1046	0.1177	7, 8(a), (b)