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PM_{10}-$PM_{2.5}$ TIME SERIES AND FRACTAL ANALYSIS

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ABSTRACT
Several epidemiological studies have shown an association between particulate air pollution and health effects. Suspended particulates and more specifically the inhalable PM_{10} fraction appear to cause respiratory health effects and heart diseases. Furthermore, particulate pollution is of paramount importance in areas with open-pit mines and especially when it is combined with raw lignite transfer and combustion in power stations through the suspension of particles and stack emissions, respectively. The penetration of particles into respiratory track is a function of the size of the particles and thus, it is more likely for the finer $PM_{2.5}$ fraction to reach the deepest of the lugs.
The fast economic growth the last decades has resulted in an increase of the sources of pollution not only in large metropolitan areas but also in medium-sized urban areas like the city of Kozani, Greece. It is the most densely populated city in the area of West Macedonia affected by urban particulate matter originated from local and stationary sources, from regional and long-range transport, and from street dust resuspension. Kozani is located a few kilometers away from lignite power stations that contribute to about 70% of the total electrical energy produced in Greece. Dust emissions seem to be the most serious problem in the area, as the measured ambient concentrations of suspended particles are at high levels and exceed local and international standards.
In this study PM_{10} and $PM_{2.5}$ concentrations are presented. The measurements have been carried out, from April to December 2002, by the Lab of Atmospheric Pollution and Environmental Physics (LAPEP) of Technological Education Institute of West Macedonia in the commercial centre of the city of Kozani. The temporal variation of PM_{10} and $PM_{2.5}$ concentrations was studied and allowed a further insight on the factors affecting the measured ambient particulate levels. $PM_{2.5}$ – PM_{10} correlation and $PM_{2.5}/PM_{10}$ ratios were investigated and compared to those in the literature together with the factors affecting their diurnal variation. The pollution levels were also detected in process of the experimental time series data by fractal dimension. Generally, fractal analysis is able to detect the data set complexity by scaling empirical data using threshold values. These values define the levels of air pollution episodes. The method presented in this study, is the transformation of PM_{10} and $PM_{2.5}$ concentrations into a set of points whose dimension was estimated by box counting. This technique has estimated the fractal dimension of both the time series by the relationship between data variance and time scale.

KEYWORDS: Urban air pollution, power plants, timeseries analysis, PM_{10}, $PM_{2.5}$, $PM_{2.5}/PM_{10}$ ratio, fractal dimension

1. INTRODUCTION
Several epidemiological studies have shown an association between particulate air pollution and health effects (Dockery *et al.,* 1993; Schwartz, 1994; Pope, 2000). The penetration of

particles into respiratory track is a function of the size of the particles and thus, it is more likely for the finer $PM_{2.5}$ fraction to reach the deepest of the lugs. $PM_{2.5}$ fraction is now widely measured in Europe and is considered to represent the alveolar fraction of the ambient particles (ISO, 1995).

The consideration of $PM_{2.5}/PM_{10}$ ratio is important as high ratios attribute particulate air pollution to vehicle emissions and secondary particles, formed in the atmosphere from gases. On the other hand smaller ratios are related to strong dust emissions and re-suspension due to high traffic volume. (EPA,2001). The fluctuation of $PM_{2.5}/PM_{10}$ ratio during the day describes the higher precipitation rate of coarser particles in contrast with the finer particles (Querol *et al.,* 2001).

The air pollution problem affects not only the big cities but also medium sized urban areas. A typical example is the city of Kozani in Greece. It has a population of 70,000 inhabitants and recent studies have revealed the nature of the air pollution problem in the city (Triantafyllou *et al.*, 2002). This is due to the fast economic growth in the last decades that resulted in an increase of the sources of pollution in the city of Kozani. This sudden growth in combination with the urban development has resulted to increased traffic throughout the central district of the city, constituting direct sources of PM pollution and causing re-suspension of dust. In addition, particulate pollution is of paramount importance in areas with open-pit mines (Ghose and Majee, 2000) and especially when it is combined with raw lignite transfer and combustion in power stations (PS) through the suspension of particles and stack emissions, respectively (Triantafyllou, 2000). Kozani Basin (KB) is a heavy industrialized area in the northwestern part of Greece, which is characterized by complex terrain. The PS in the basin, contribute to about 70% of the total electrical energy produced in Greece. The lignite used by these PS is mined in the nearby open-pit mines. Dust emissions seem to be the most serious problem in the basin, as the measured ambient concentrations of suspended particles are at high levels and, occasionally, exceed local and international standards (Triantafyllou, 2003). In general, the dust generated in the basin under specific meteorological conditions may affect Kozani. In the present study, an attempt is made by the timeseries analysis of PM_{10} and $PM_{2.5}$ concentrations and $PM_{2.5}/PM_{10}$ ratio in order to assess their origin, relative variability and transformations.

Finally, the data timeseries have been analyzed by a fractal process (Mandelbrot, 1967; Mandelbrot, 1975; Mandelbrot, 1982). This process is characterized by a constant parameter D known as the fractal dimension (Freeborough, 1997) that can be viewed as a relative measure of complexity, or as an index of the scale-dependency of a pattern. A fractal dimension can be assigned to a set of time series data by plotting it as a function of time, and calculating a box dimension. The scale-invariant behavior of $PM_{10/2.5}$ concentrations time structure has been investigated by the application of the box counting method (Lee *et al.*, 2003). The $PM_{10/2.5}$ series of hourly average concentrations were transferred into a useful compact form through this method, namely, the box-dimension (D)-threshold (T_h) and critical scale (C_S)-threshold (T_h) plots, which could be interpreted in terms of traditional statistical parameters. Since the dependences of both D and C_S on the T_h values were closely related to the variation of concentrations in time, they were used to characterize the temporal distribution of concentrations.

2. INSTRUMENTATION

Particulate matter measurements have been performed from April to December 2002 in the city of Kozani at a monitoring station located at Aristotelous square, which is one of the central squares characterized by heavy traffic. For the PM_{10} and $PM_{2.5}$ measurements, a beta adsorption monitor was used (FAG, Germany), equipped with a specific sampling head for particles with an aerodynamic diameter less or equal to 10 μm and less or equal to 2.5 μm. The tube between the head of the monitor and the filter is heated to 50^o C. The measuring efficiency of the low-volume design of this PM_{10} monitor has been tested during inter-comparison experiments with a dichotomous sampler instrument. The principle of its operation is based on a beta ray adsorption by particles collected through the instrument on a fiberglass filter tape. Calibration is performed by means of special foils with known adsorption rates. Zeroing of the instruments was carried out automatically at each measuring cycle and dust average concentrations were calculated every five minutes.

3. RESULTS AND DISCUSSION

3.1 Description of PM_{10} and $PM_{2.5}$ time series

During the period under consideration, the mean daily concentration of PM_{10} was 47 µg m^{-3} with a standard deviation (sd) of 18 µg m^{-3}. The average daily values of PM_{10} exceeded the European Union limit for the year 2002 of 65 µg m^{-3} in percentage of 12% (EU-Commission, 1999). The $PM_{2.5}$ mean daily concentration was 19 µg m^{-3} with a sd of 7 µg m^{-3}. This has surpassed the United States National Ambient Air Quality $PM_{2.5}$ annual standard of 15 µg m^{-3}. Fig. 1 shows the diurnal variation of PM concentrations during the sampling period for the whole period. It was concluded, that the maximum concentrations occurred in the morning and during the evening. The heavy morning traffic volume favour resuspension of the coarser particles and then, they steadily decrease towards next morning. The high proportion of finer particles indicates that the coarser particles are settled generally quickly, leaving finer particles ($PM_{2.5}$) suspended in the air. The finer particles begin to increase with time delay while their concentration remains high during the day due to the vehicles' emissions. However, the background $PM_{10/2.5}$ concentrations are day independent and developed at 15 µg m^{-3} and 30 µg m^{-3} levels for $PM_{2.5}$ and PM_{10}, respectively.

3.2 $PM_{2.5}/PM_{10}$ ratio distribution mass

The $PM_{2.5}/PM_{10}$ mass ratios have been used for the evaluation of the daily seasonal variation. The average $PM_{2.5}/PM_{10}$ mass ratio was 0.42±0.1 that indicates that coarser particles have participated by a larger portion in PM content. Similar ratios 0.42 and 0.44 were found, at two case studies in the Mexico City (Vega *et al.*, 2002). Furthermore, annual means of $PM_{2.5}/PM_{10}$ ratios measured in a large number of urban and semi-rural US areas (EPA, 2001) have taken values within the range 0.39-0.69. Fig.2 has shown similar diurnal variation for mass ratios during weekdays due to the governing and similar urban activities throughout the period. The maximum ratio was observed in December because the most of the days were rainy. The minimum values (0.29-0.35) occurred during traffic hours (9:00-12:00) as a consequence of the re-suspended coarse road dust. Then, the ratios lied within the range 0.35-0.54 due to PM_{10}'s increased precipitation rate (Querol *et al.*, 2001).

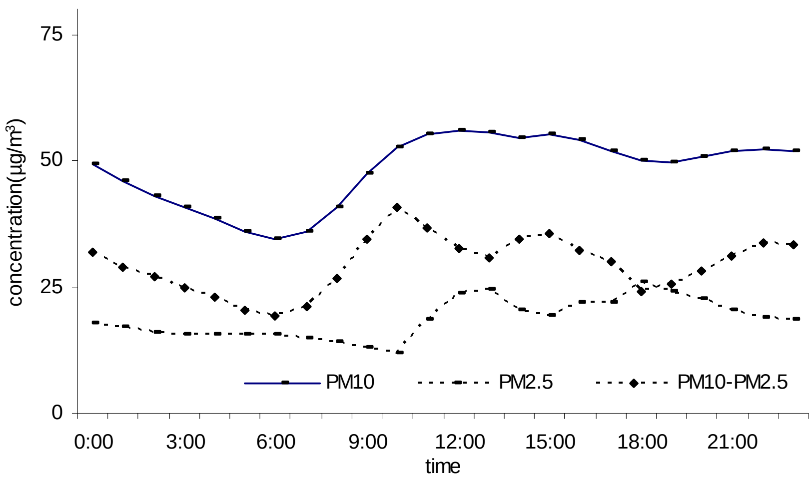

Figure 1. PM diurnal variation

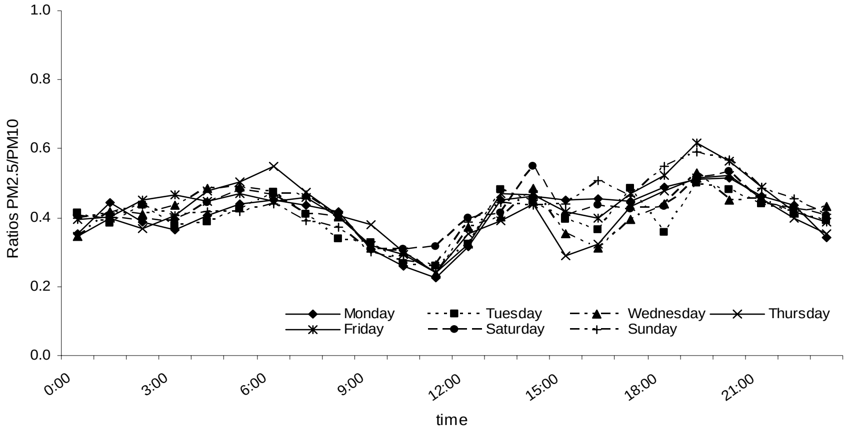

Figure 2. Diurnal variation of the $PM_{2.5}/PM_{10}$ ratios

4. FRACTAL ANALYSIS

In the present study the fractal analysis method can be applied in particulate matter concentrations according to a study carried out by Lee (Lee *et al.,* 2003). This method calculates the number of boxes that include at least one concentration above a characteristic value (threshold value T_h). The space of observation is divided into non overlapping boxes of characteristic size *L* (in hours) and the number of boxes *N*(*L*) needed to cover the time period of the dataset is counted. When applying this method to a time series, the boxes represent time intervals and the space of observation is equal to the total length of the series. In this study, we convert the values of the measurements into sets of points (indicating value above T_h) by using different T_h levels. For this conversion, a zero T_h means that all hours with registered measurements in the series are considered a point. If scale invariance exists in the data set, the expression $N(L)=L^{-D}$ will hold, with D as the box dimension. From the time series we can generate a plot of log[*N*(*L*)] vs. Log(*L*) and the exponent *D* can be calculated from the slope of a linear regression to the values obtained (see Fig. 3). The time scale *L* denotes a time interval within which $PM_{10/2.5}$ exceeding occurs. The curve is composed of two distinctly different sections; one with slope equals to -1 (Log(*L*)>2.6) and the other with –*D* (Log(*L*)<2.6). The value of *L* at the intersection of the zero T_h straight line and a different T_h line depicts the critical scale C_S. When time scale L is greater than or equal to the critical scale C_S, the values events exceeding the threshold T_h must occur.

In Fig. 3 T_h is double the mean concentration. Similar to this, if T_h takes several different values then Fig. 4a and 4b describe the box counting of PM_{10} and $PM_{2.5}$ concentrations, respectively.

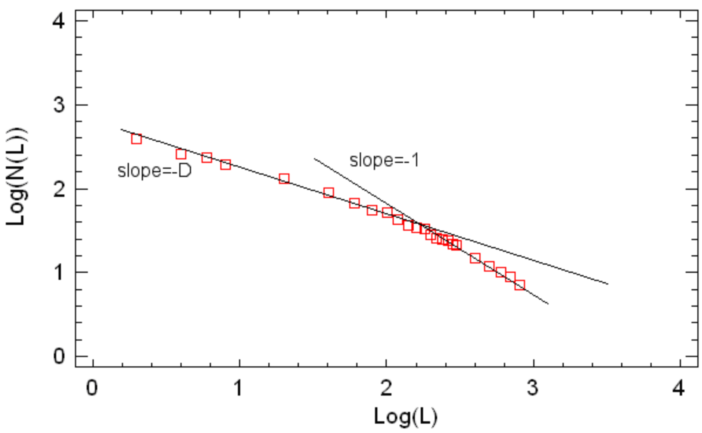

Figure 3. Box counting for T_h=2*mean

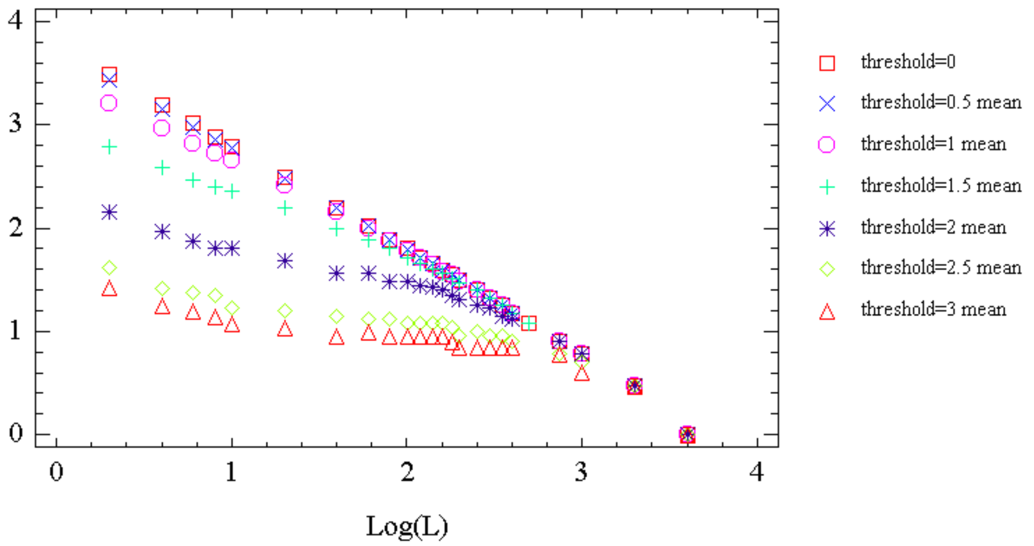

Figure 4a. Box counting graph for PM_{10} measurements

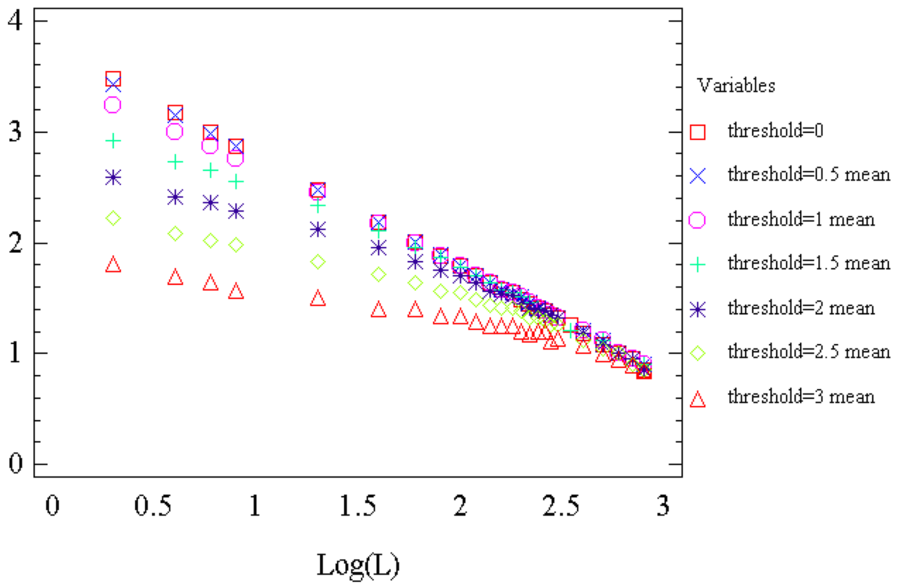

Figure 4b. Box counting graph for $PM_{2.5}$ measurements

As demonstrated in Fig. 5 and 6, at low T_h (0–1.5 * mean) the D and C_S values of PM_{10} are equal with that of $PM_{2.5}$. Then, increasing T_h value, D and C_S values of PM_{10} become smaller and larger, respectively, than those of $PM_{2.5}$. That may indicate the same sources for background measurements and different sources for high pollution episodes with coarser particles.

The calculation of the number of exceedances N(L) for each T_h has been carried out by a simple computer program created for the purposes of the present work. It has been developed in visual basic and is available to the users at the URL:http://airlab.teikoz.gr.

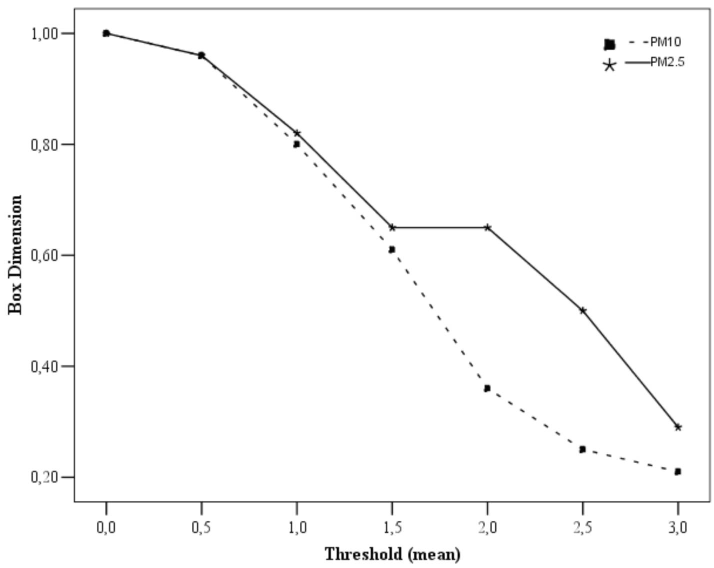

Figure 5. Box dimension vs threshold

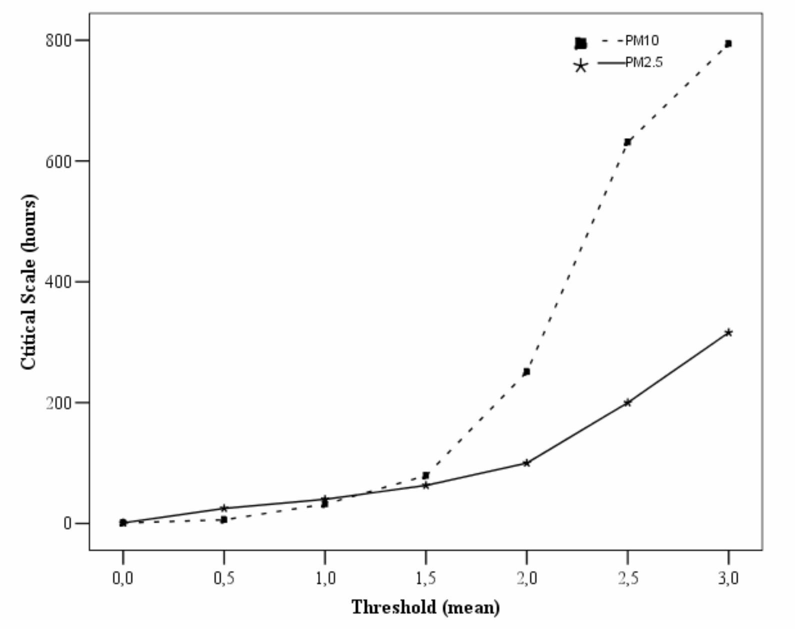

Figure 6. Critical scale vs threshold

4. CONCLUSIONS

The PM_{10}'s mean daily concentration (47 µg m^{-3}) from April to December of 2002 has exceeded the European Union annual limit of 40 µg m^{-3}. The $PM_{2.5}$ mean daily concentration was 19 µg m^{-3} and has exceeded the United States National Ambient Air Quality $PM_{2.5}$ annual standard of 15 µg m^{-3}. The PM_{10}'s maximum concentrations occurred earlier in the morning than for $PM_{2.5}$ indicating the importance of resuspension of the coarser particles. Then, the finer particles are settled slower than PM_{10} leaving $PM_{2.5}$ suspended in the air. In addition, the $PM_{10/2.5}$ concentrations background build up was at relatively high levels.

The average $PM_{2.5}/PM_{10}$ mass ratio was 0.42±0.1 that indicates that coarser particles have participated by a larger portion in PM content. A similar ratio has been found in Mexico City and in urban areas in the US that indicated the same nature of sources. The ratio daily distribution has varied similarly for all weekdays due to the governing and urban activities throughout the period. The maximum ratio has occurred in December because of the high degree of wet precipitation. Moreover, the minimum values occurred during traffic hours (9:00-

12:00) as a consequence of the re-suspended coarse road dust. Then, the ratio decreased due to the faster rate of precipitation of the coarser particles.

Box counting technique has been used to investigate the clustering properties of $PM_{10/2.5}$ time series. This proved to give an interesting description of particulate matter concentrations that could accompany ordinary frequency diagrams. The existence of a straight-line section in the box counting graphs indicates that a box dimension can characterize the temporal structure of $PM_{10/2.5}$. In other words, it displays time scale invariance within a specific time interval. That may indicate the same sources for background measurements and different sources for high pollution episodes.

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