



The statistical characters of PM₁₀ in Belgrade area

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ARTICLE INFO

Article history:

Received 10 March 2008

Received in revised form 5 December 2008

Accepted 4 January 2009

Keywords:

PM₁₀

Probability distribution

Extreme theory

Source reduction

ABSTRACT

The concentrations of air pollutants depend on meteorological conditions and pollutant emission level. From the statistical properties of air pollutants the number of times the daily average concentrations exceed the assigned air quality standard (AQS) can be estimated, as well as the level of reduction of particle matter emission sources required to meet the AQS. In this paper three statistical distributions (lognormal, Weibull and type V Pearson distribution) were used to fit the complete set of PM₁₀ data for the Belgrade urban area during a three-year period (2003–2005). The method of moments and the method of least squares were both used to estimate the parameters of the three theoretical distributions. The type V Pearson distribution represented the PM₁₀ daily average concentration most closely. However, the parent distributions sometimes diverged in predicting a high PM₁₀ concentration and therefore asymptotic distributions of extreme values were used to fit the high PM₁₀ concentration distribution more correctly. This method can successfully predict the return period and exceedances over a critical concentration in succeeding years. The estimated emission source reduction of PM₁₀ to meet the assigned standard varied from 53% to 63% in the Belgrade urban area. The results provide useful information for air quality management and could be used to examine the similarities and differences among air pollution types in diverse areas.

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1. Introduction

Atmospheric aerosols are of major scientific interest due to their confirmed role in climate change (IPCC, 2001) and their effect on human health (Schwartz et al., 1996; Dockery and Pope, 1994) and local visibility. The impact of atmospheric aerosols on the radiative balance of the Earth is of comparable magnitude to greenhouse gas effects (Anderson et al., 2003). In order to protect public health and the environment i.e. to control and reduce particulate matter (PM) levels, air quality standards (AQS) were issued and target values for annual and daily mean PM₁₀ (particles below 10 μm in diameter) mass concentrations were established. For the first stage the EU Directive (EC, 1999) required an annual limit of 40 μg m⁻³ and a 24 h limit of 50 μg m⁻³ (not to be exceeded more than 35 times in a calendar year) to be met by 2005 and, in the second stage, an annual limit of 20 μg m⁻³ and a 24 h limit of 50 μg m⁻³ (not to be exceeded more than 7 times in a calendar year) to be met by 2010.

Although PM is of great concern for public health, studies related to the influence of suspended PM₁₀ on air quality in the Belgrade urban atmosphere were not initiated until 2002. Some results of preliminary investigations concerning PM₁₀ have been published (Rajšić et al., 2004).

The obtained data for PM₁₀ mass concentrations have been subjected to statistical processing in order to determine the frequency distribution. Similarly to other air pollutants, PM concentrations are random variables influenced by the emission level, meteorological conditions and topography. Each area is specific and the required emission reduction to meet AQS is different. Information about the frequency distribution of pollutants is useful for developing air pollution control strategies. When the specific probability function of an air pollutant is known, it is easy to predict the required emission reduction, the frequency of exceedance of the AQS, as well as the return period. Many types of probability distributions have been used to fit air pollutant concentrations. These include lognormal distribution (Mage and Ott, 1984; Kao and Friedlander, 1995; Lu, 2002; Lu and Fang, 2002), pseudo-lognormal distribution (Vukmirović, 1990), Weibull distribution (Georgopoulos and

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Seinfeld, 1982), gamma distribution (Lu, 2004) and type V Pearson distribution (Morel et al., 1999).

Lognormal, Weibull and type V Pearson distributions were chosen to fit the data for PM₁₀ in Belgrade. The distributional parameters were estimated by the moments and maximum likelihood methods. The reduction in emission sources of PM₁₀ required to meet AQS was estimated employing a rollback equation. Moreover, since the tail of theoretical distributions diverged in the high concentration region, a two-parameter exponential distribution and asymptotic distribution of extreme value were used to fit high PM₁₀ concentrations and estimate exceedances and return period more precisely.

Thus, the goal of this work was to present some statistical characteristics of PM₁₀ mass concentrations measured in the Belgrade urban area during the period 2003–2005, and to estimate the reduction of average PM₁₀ source emission required to meet AQS.

2. Methods

Daily average mass concentrations of PM₁₀ were taken from 2003 to 2005 in order to estimate the parameters for three theoretical distributions. The measurements were performed in the central urban area of Belgrade.

Suspended particles were collected on preconditioned (48 h at 20 °C and constant relative humidity around 50%) and pre-weighed Pure Teflon filters (Whatman, 47 mm diameter, 2 μm pore size) using two MiniVol air samplers (Airmetrics, Co. Inc., 5 l min⁻¹ flow rate) provided with PM₁₀ cutoff inlets and positioned at 2 m height. The sampling time was 24 h yielding a sample volume of 7.2 m³. Routine maintenance of the samplers and calibration of the flow meters were conducted in order to ensure the sampling quality (Airmetrics, 2001). After particle collection, the filters were sealed in plastic bags and kept in portable refrigerators, in a horizontal position during transportation back to the laboratory where there were reconditioned for another 48 h.

The samples were handled and processed in a Class 100 clean laboratory, at the Institute of Physics. Particle matter mass concentration was determined by weighting of the filters using a semi-micro balance (Sartorius, R 160P), with a minimum resolution of 0.01 mg. Loaded and unloaded filters (stored in Petri dishes) were weighed after 48 h conditioning in a desiccator, in a clean room at a relative humidity of 45–55% and temperature of 20 ± 2 °C. Quality assurance was provided by simultaneous measurements of a set of three “weigh blank” filters that were interspersed within the pre- and post-weighing sessions of each set of sample filters and the mean change in “weigh blank” filter mass between weighing sessions was used to correct the sample filter mass changes.

The daily average concentrations of PM₁₀ were used to estimate the parameters of the three theoretical distributions whose probability density functions are:

lognormal

$$p_l(x) = \frac{1}{x\sigma_g(2\pi)^{\frac{1}{2}}} \exp\left[-\frac{(\ln x - \ln \mu_g)^2}{2\sigma_g^2}\right], \quad x > 0; \sigma_g > 0; \mu_g > 0 \quad (1)$$

where x is the pollutant concentration and μ_g and σ_g are the parameters of distribution;

Weibull

$$p_w(x) = \frac{\lambda}{\sigma} \left(\frac{x}{\sigma}\right)^{\lambda-1} \exp\left[-\left(\frac{x}{\sigma}\right)^\lambda\right], \quad x \geq 0; \sigma, \lambda > 0 \quad (2)$$

where λ and σ are the parameters of distribution;

type V Pearson

$$\phi(x_i; x_i^{\text{eq}}) = \frac{x_i^{\text{eq}}(\rho_i)^{\rho_i+1}}{\Gamma(\rho_i+1)} e^{-\rho_i \frac{x_i}{x_i^{\text{eq}}}} \left(\frac{x_i}{x_i^{\text{eq}}}\right)^{-(\rho_i+2)}, \quad x_i > 0; x_i^{\text{eq}} > 0; \rho_i > 0 \quad (3)$$

where Γ is the gamma function and ρ_i and x_i^{eq} are the parameters of type V Pearson distribution (Morel et al., 1999).

The appropriateness of each distribution was assessed by the Kolmogorov–Smirnov (K–S) and chi-squares tests. The K–S statistic is defined as the maximum difference between the sample cumulative probability and the expected cumulative probability i.e. $D = \max|f_n(x) - F(x)|$ where $f_n(x)$ and $F(x)$ are the expected and observed cumulative frequency functions, respectively. The D value declines with increasing goodness of fit.

2.1. Parameter estimation

Distribution parameters were estimated from the measured data by the method of moments and the method of maximum likelihood.

2.1.1. Method of moments

Parameters for lognormal distribution can be estimated by calculating the first (M_1) and second (M_2) moments and using the following relations (Georgopoulos and Seinfeld, 1982; Lu, 2002):

$$\ln \mu_g = 2 \ln M_1 - \frac{1}{2} \ln M_2 \quad (4)$$

$$(\ln \sigma_g)^2 = \ln M_2 - 2 \ln M_1 \quad (5)$$

The relations between the estimated parameters and moments for Weibull distribution are:

$$\left(\frac{\Gamma(1+2/\lambda)}{\Gamma^2(1+1/\lambda)} - 1\right)^{\frac{1}{2}} = \frac{\left[(1/n-1) \sum_{i=1}^n (x_i - M_1)^2\right]^{\frac{1}{2}}}{M_1} \quad (6)$$

$$\sigma = \frac{M_1}{\Gamma(1+1/\lambda)} \quad (7)$$

In addition, the parameters of the type V Pearson distribution can be computed from Eqs. (8) and (9):

$$x_i^{\text{eq}} = M_1 \quad (8)$$

$$\rho_i = \frac{M_2}{M_2 - M_1^2} \quad (9)$$

2.1.2. Method of maximum likelihood

For lognormal distribution the estimated values of the parameters are given by:

$$\ln \mu_g = \frac{1}{n} \sum_{i=1}^n \ln x_i \quad (10)$$

$$(\ln \sigma_g)^2 = \frac{1}{n} \sum_{i=1}^n (\ln x_i - \ln \mu_g)^2 \quad (11)$$

For the Weibull distribution the estimated values of λ and σ_w are:

$$\lambda = \left[\left(\sum_{i=1}^n x_i^\lambda \ln x_i \right) \times \left(\sum_{i=1}^n x_i^\lambda \right)^{-1} - \frac{1}{n} \sum_{i=1}^n \ln x_i \right]^{-1} \quad (12)$$

$$\sigma_w = \left(\frac{1}{n} \sum_{i=1}^n x_i^\lambda \right)^{\frac{1}{\lambda}} \quad (13)$$

For the type V Pearson distribution the estimated values of parameters are:

$$x_i^{\text{eq}} = \frac{\rho_i + 1}{\rho_i} \frac{1}{\left(\frac{1}{n} \sum_{i=1}^n \frac{1}{x_i} \right)} \quad (14)$$

$$\frac{d \ln(\Gamma(\rho_i + 1))}{d\rho_i} = \ln(\rho_i x_i^{\text{eq}}) - \frac{1}{n} \sum_{i=1}^n \ln(x_i). \quad (15)$$

In order to calculate parameters of the distributions all equations can be solved numerically.

2.2. Estimation of emission source reduction

Assuming unchanged spatial distribution of emission sources, meteorological conditions and nonreactive species, according to the rollback equation (Georgopoulos and Seinfeld, 1982; Seinfeld and Pandis, 1998) the emission source reduction R (%) required to meet AQS can be estimated by:

$$R = \frac{E\{C_p\} - E\{C_s\}}{E\{C_p\} - C_b}. \quad (16)$$

In this equation $E\{C_s\}$ is the mean (expected) concentration of distribution when the extreme value equals C_s (the concentration of the AQS), $E\{C_p\}$ the mean concentration of the actual distribution and C_b the background concentration. If $C_s = 125 \mu\text{g m}^{-3}$, the PM_{10} daily average concentration is not exceeded more than once per year ($P\{\text{PM}_{10} > C_s\} = 1/365 = 0.00274$), then $E\{C_s\}$ is the expected daily PM_{10} average concentration of a distribution where the probability of a concentration exceeding $125 \mu\text{g m}^{-3}$ equals 0.00274.

2.3. The distribution of high PM_{10} concentrations

Previous theoretical distributions can give good result for estimating the mean concentration and required reduction of source emission. However, the fitted results of the parent distributions are not accurate enough in the high concentration region. Therefore, a two-parameter exponential distribution and asymptotic distribution were applied to predict the return period and exceedances of the critical PM_{10} concentration.

2.3.1. Two-parameter exponential distribution

A two-parameter exponential distribution derived from extreme value theory (Lu and Fang, 2003) represents the cumulative frequency distribution of high concentrations over a specific percentile.

$$F_L = 1 - e^{-y_n} \quad (17)$$

$$y_n = b_m(x_n - \phi) \quad (18)$$

where y_n , b_m and ϕ are the variate and the parameters of the distribution and x_n is the chosen PM_{10} concentration exceeding the specific percentile. From the complete data set of PM_{10} (2003–2005) concentrations exceeding 80th percentile were chosen to fit the two-parameter exponential distribution. The estimated cumulative probability can be calculated from the chosen high PM_{10} concentration, x_n , using Eq. (19):

$$\bar{F}_L(x_n) = \frac{N_1 - r + 1}{N_1 + 1} = P_{rN_1} \quad (19)$$

where N_1 is the size of the chosen high PM_{10} concentration and P_{rN_1} is the probability of a value that is ranked r out of N_1 values. The relation between variate y_n and P_{rN_1} is:

$$y_n(r) = -\ln(1 - P_{rN_1}) \quad (20)$$

and the parameters b_m and ϕ can be estimated by the least-squares method. In addition, the return period $R(x_c)$, defined as the average number of averaging periods (or observations) between exceedances of a given critical concentration, x_c , can be calculated as:

$$R(x_c) = \frac{1}{(1-f)(1 - F_L(x_c))} \quad (21)$$

where f is the chosen specific percentile. After determining return period the days of exceedances within one year can be calculated in order to develop air control strategy.

2.3.2. Asymptotic distribution of the extreme value theory

The cumulative distribution of m th largest value x_m out of samples of size n is denoted as $G_{mn}(x)$. If the parent distribution of the random variable X is exponential type, then the extreme values are themselves random variables and the statistical distribution of the series of N extremes approaches Gumbel's type I asymptote (Gumbel, 1958; Roberts, 1979). For type I asymptotic distribution the approximation is shown as:

$$G_{mn}(x_m) = \exp[-\exp(-y_m)] \quad (22)$$

where $y_m(x_m) = a_m(x_m - u_m)$ is the asymptote variate of Gumbel's type I asymptote and α_m and u_m are the parameters of type I asymptotic distribution. The cumulative probability of measured extremes can be calculated by Eq. (23):

$$\bar{G}_{mn}(x) = \frac{N - r + 1}{N + 1} = P_{rN} \quad (23)$$

where N is the number of extreme values, P_{rN} is the probability of a value that is ranked r out of N extremes. In addition, the relation between the asymptotic variate y_m and P_{rN} is:

$$y_m(r) = -\ln(-\ln P_{rN}) \quad (24)$$

and the appropriate parameters can be estimated (Surman et al., 1987). The maximum values of daily average PM_{10} concentrations of each month for 2003–2005 were chosen to obtain the asymptotic distribution $G_{mn}(x)$. Then, the expected return period can be calculated as:

$$R(x_c) = \frac{1}{1 - G_{mn}(x_c)}. \quad (25)$$

3. Results and discussion

The fitted results of three theoretic distributions and the measured data for PM₁₀ in Belgrade from 2003 to 2005 are presented in Fig. 1.

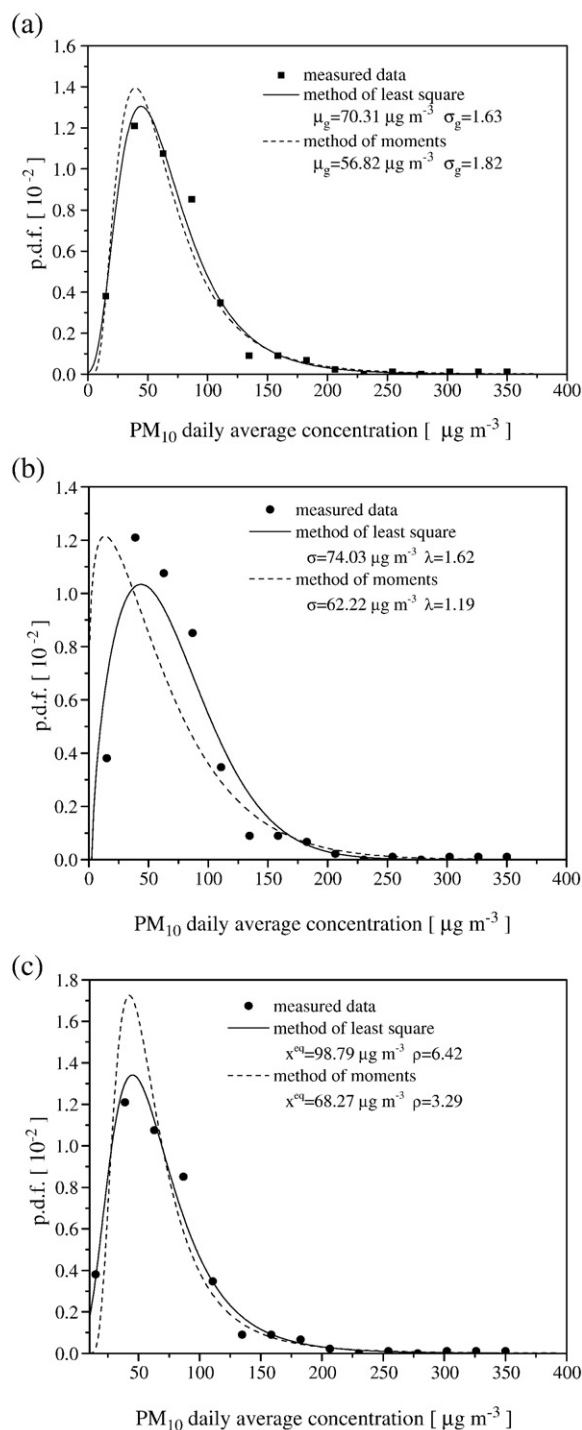


Fig. 1. Comparisons of two methods for estimating the daily average concentration distribution of PM₁₀ for (a) lognormal distribution, (b) Weibull distribution and (c) type V Pearson distribution.

Table 1

Estimated parameters for three theoretical distributions.

| | Type V Pearson | Lognormal | Weibull |
|--------------------|----------------|-----------|---------------------------------------|
| Method of moments | x^{eq} | ρ | μ_g σ_g λ σ |
| K-S | 68.275 | 3.293 | 56.826 1.823 1.192 62.226 |
| χ^2 | 0.077 | 0.051 | 0.226 |
| Maximum likelihood | x^{eq} | ρ | μ_g σ_g λ σ |
| K-S | 51.83 | 6.420 | 34.77 136.50 1.621 74.032 |
| χ^2 | 98.790 | 0.041 | 0.073 |
| | 0.038 | 22.34 | 47.21 |

Using the method of moments the values of the estimated parameters are $x^{eq} = 68.2 \mu\text{g m}^{-3}$, $\rho = 3.29$ for type V Pearson distribution; $\mu_g = 56.82 \mu\text{g m}^{-3}$, $\sigma_g = 1.82 \mu\text{g m}^{-3}$ for lognormal distribution and $\lambda = 1.19$, $\sigma = 62.22$ for Weibull distribution. With the method of maximum likelihood the estimated parameters are $x^{eq} = 98.7 \mu\text{g m}^{-3}$, $\rho = 6.42$ for type V Pearson distribution; $\mu_g = 70.31 \mu\text{g m}^{-3}$, $\sigma_g = 1.63 \mu\text{g m}^{-3}$ for lognormal distribution and $\lambda = 1.62$, $\sigma = 74.03$ for Weibull distribution. Once the distribution parameters were determined, the chi-square and K-S tests could be used to assess which type of distribution is more appropriate for representing the PM₁₀ distribution. The results of both tests are presented in Table 1.

Smaller values of chi-square as well as D_{max} were obtained by the method of least squares and this method yields a better fit with the actual data. It is clear that the Weibull distribution is inappropriate for representing PM₁₀ distribution, while type V Pearson distribution is the most suitable one (Fig. 2).

In addition, the probability of exceeding the AQS can be predicted using predetermined distribution parameters by least square methods and integrating probability distribution functions. Fig. 3 shows the relation between exceeding probability and PM₁₀ concentration for different distribution functions.

It was found that the probabilities of exceeding the AQS ($x_{PM_{10}} > 125 \mu\text{g m}^{-3}$) were 0.12, 0.096 and 0.088 for lognormal, Weibull and type V Pearson distribution, respectively. This means that the number of days exceeding the AQS in the following year would be 43, 35 and 32, respectively. The actual probability for this period was 0.075 (27 days) and it is clear that all three distributions overestimated the exceeding probability but type V Pearson distribution most closely represented the true PM₁₀ data.

3.1. Estimating the emission source reduction

After determining the most appropriate distribution function for PM₁₀ the emission source reduction required to meet AQS can be predicted from a rollback equation. Parameters of distribution which control the size of fluctuation are not influenced by pollution emission level, unlike the mean (expected) value of PM₁₀ daily average concentration ($E\{C_p\}$). The complementary distribution function for calculating the probability of variable x_i exceeding the critical value x for lognormal and type V Pearson distribution can be found in a statistical textbook. The actual mean concentration for the measured period was $68.4 \mu\text{g m}^{-3}$ with a standard deviation of $46.3 \mu\text{g m}^{-3}$. In order to meet AQS, mean values of PM₁₀ should be reduced. The relation between μ_g and mean concentration

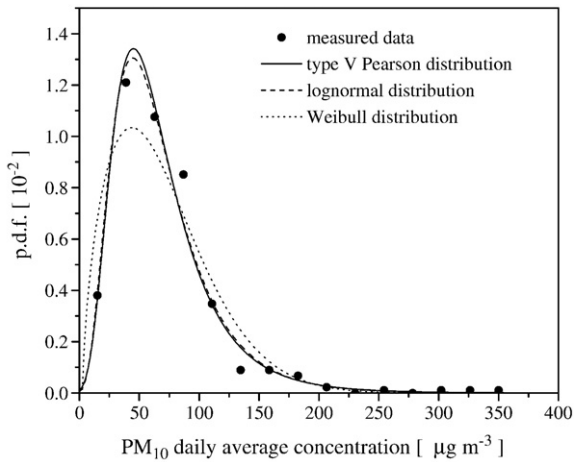


Fig. 2. Comparison of measured data with the theoretical distributions by the method of least squares.

for lognormal distribution is: $\ln E\{C_p\} = \ln \mu_g + \frac{1}{2} (\ln \sigma_g)^2$ (Georgopoulos and Seinfeld, 1982). The parameter $\sigma_g = 1.63$ was used to calculate the relationship between mean (expected) PM_{10} concentration and the probability of exceeding AQS for lognormal distribution. It was found that the value of $E\{c\}_s$ (the average concentration of lognormal distribution where the probability of a concentration exceeding $125 \mu\text{g m}^{-3}$ equals 0.00274) was $32.1 \mu\text{g m}^{-3}$. Therefore, the mean PM_{10} concentration should be reduced from the current value of $68.4 \mu\text{g m}^{-3}$ to $32.1 \mu\text{g m}^{-3}$ and the estimated emission reduction can be calculated as follows:

$$R = \frac{E\{C_p\} - E\{C\}_s}{E\{C_p\} - C_b} = \frac{68.4 \mu\text{g m}^{-3} - 32.1 \mu\text{g m}^{-3}}{68.4 \mu\text{g m}^{-3}} = 0.531$$

i.e. 53.1%. For type V Pearson distribution the estimated source reduction required to meet AQS is 58.6%. Therefore, the source emissions should be controlled much more to reduce PM_{10} concentration and meet AQS in the future period. However, the

tail property of distributions is important for estimating emission reduction and the probability of exceedance. Two-parameter exponential distribution and asymptotic distributions were employed to provide better fit for the high region of PM_{10} concentrations.

3.2. High PM_{10} concentration analyses

Fig. 4 shows the fitted theoretical line of the variate y_n and PM_{10} concentration over 80th percentile, x_n . The fitted equation for the theoretical two-parameter exponential distribution is $F_L(x_n) = 1 - \exp[-0.016(x - 75.37)]$. The coefficient of determination, $R^2 = 0.94$, indicates that this theoretical distribution fits the high-concentration region quite successfully. From Eqs. (17) and (21) for critical concentration, $x_c = 125 \mu\text{g m}^{-3}$, F_L and the return period can be calculated as: $F_L(x_c) = 1 - \exp[-0.016(125 - 75.37)] = 0.548$ and $R(x_c) = 1 / [(1 - 0.8)(1 - 0.548)] = 11$ days. From this calculation predicted exceedances over AQS are estimated to be 33 days in the following year. The prediction of return period for a critical concentration can also be used for estimating source emission reduction. If the AQS of PM_{10} is $125 \mu\text{g m}^{-3}$ and the allowed exceeding probability is once per year, then the expected return period is 365 days and $F_L(125 \mu\text{g m}^{-3}) = 0.986$. In addition, the PM_{10} concentration expected to be equaled or exceeded once per year can be calculated from Eq. (17). The expected concentration is $337 \mu\text{g m}^{-3}$ and it can be seen that a reduction of $212 \mu\text{g m}^{-3}$ needs to be achieved in Belgrade in order to meet AQS ($125 \mu\text{g m}^{-3}$). The standard deviation of the expected PM_{10} concentration S_{x_0} can be obtained from Eq. (26) (Kleinbaum et al., 1988)

$$S_{x_0} = S_y \sqrt{1 + \frac{1}{n} + \frac{(x_0 - \bar{x})^2}{(n-1)S_x^2}} \tag{26}$$

where S_y^2 is the estimate of variance of y ; \bar{x} the average and S_x^2 the variance of x . The calculated value is: $S_{x_0} = 2 \mu\text{g m}^{-3}$, and, if we assume direct proportionality between emission level and high PM_{10} concentration, the needed reduction can be expressed as $(62.9 \pm 0.6)\%$.

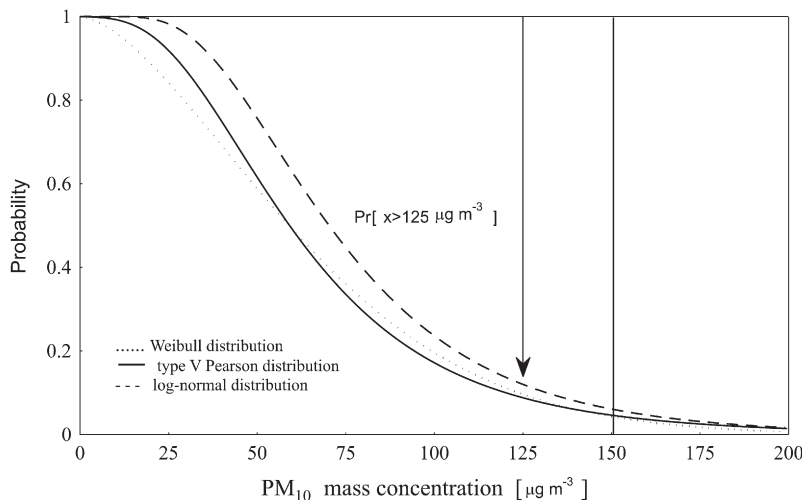


Fig. 3. Relation between PM_{10} mass and probability for different distribution functions.

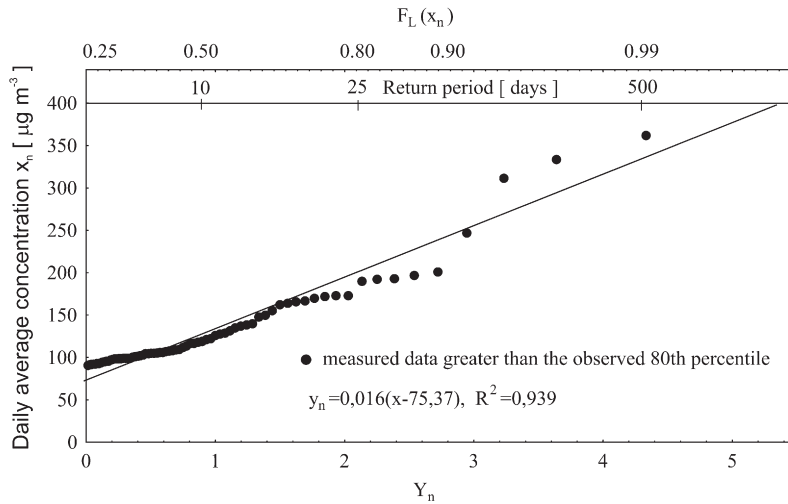


Fig. 4. The fitted theoretical line of variate and PM₁₀ concentration over a specific percentile by type I two-parameter exponential distribution.

PM₁₀ monthly extremes (x_m) and the asymptotic variate (y_m) fitted by type I asymptotic distribution are shown in Fig. 5 together with the return period and cumulative probability of extremes. As the extreme data were selected as the monthly maximum concentration, the asymptotic distribution G_{mn} can be written as $G_{1,30}$. The fitted asymptotic distribution is $G_{1,30} = \exp[-\exp[-0.013(x - 74.84)]]$. If the same AQS is assumed as earlier, it is easy to calculate $G_{1,30}(x_c = 125 \mu\text{g}^{-3}) = \exp[-\exp[-0.013(125 - 74.84)]] = 0.594$ and from Eq. (25) a return period of 2.46 months is obtained. In this case the return period represents the average period within the monthly maximum exceeding AQS will occur again and the unit of the return period is in months.

In addition, predicted exceedances over AQS during the sampling period is 14 days. After determining the return period one can estimate the required source emission reduction in a similar way. The expected return period for the AQS in this case is 12 months i.e. $G_{1,30} = 0.917$ and from Eq. (22) the expected PM₁₀ concentration is $263 \mu\text{g} \text{m}^{-3}$. The corresponding standard

deviation, S_c , of the expected PM₁₀ concentration can be calculated according to Chow et al. (1988):

$$S_c = \left[\frac{1}{N} (1 + 1.1396K + 1.1K^2) \right]^{1/2} S \quad (27)$$

where S is the standard deviation of the original sample of size N , and K is the frequency factor given by $K = -\frac{\sqrt{6}}{\pi} \{0.5772 + \ln(\ln(\frac{P}{P-1}))\}$, where P is the return period. The calculated value for S_c is $4 \mu\text{g} \text{m}^{-3}$, so a reduction of $138 \pm 4 \mu\text{g} \text{m}^{-3}$ needs to be achieved. Therefore, a decrease in emission level of $53 \pm 2\%$ should be met in the future years.

Although many assumptions have been introduced, which have caused some inaccuracy in estimating the required source reduction, these results might be useful for air pollution control strategy.

In our previous paper multivariate analysis, principal component analysis and cluster analysis were used to identify the possible emission sources of trace elements (Rajšić et al.,

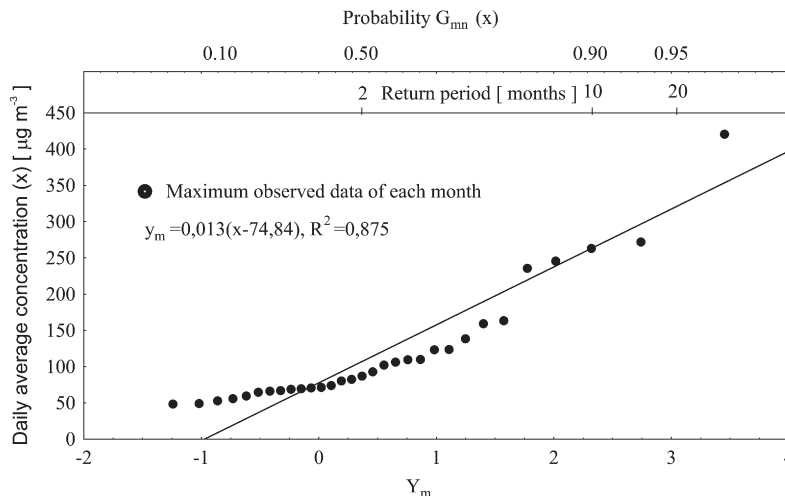


Fig. 5. The fitted theoretical line of the asymptotic variate and PM₁₀ monthly extremes by type I asymptotic distribution.

2008). The results implied that the main source of trace elements in urban suspended particles is traffic, with a considerable portion of resuspended road dust and products of other fossil fuel combustion processes. Finally, the analyses indicated that in the urban area of Belgrade, vehicle-related sources contribute more to pollutant concentration levels than industrial sources, being the major source emission which should be reduced.

4. Conclusion

In this paper three theoretical distributions (lognormal, Weibull and type V Pearson) were used to fit PM_{10} concentrations in the Belgrade urban area for the period 2003–2005. The type V Pearson distribution was found to be the most appropriate for representing PM_{10} daily average concentrations.

The average reduction of PM_{10} emission required to meet the assigned Air Quality Standard was predicted from rollback equation. The calculated values are 53% for lognormal distribution and 58% for type V Pearson distribution.

The tail properties of the distribution are very important for predicting the probability of exceedance, so a two-parameter exponential distribution was used to fit the high PM_{10} daily concentration region, while the monthly maximum level of PM_{10} was fitted by type I asymptotic distribution. The return period and exceedances of the critical PM_{10} concentration were estimated. These methods can reasonably predict the return period and exceedances in the succeeding period and can also be used for predicting source emission reduction. The estimated source emission reduction ranged from 53% to 63%. These results, together with those obtained earlier, confirm the need for further emission source reduction in Belgrade, especially from traffic, and may be useful for developing air control strategy in future years.

Acknowledgment

This work was carried out within the framework of the project No 141012 funded by the Ministry of Science and Technological Development of the Republic of Serbia.

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