Original Article

Application of Statistical Distribution of PM_{10} Concentration in Air Quality Management in 5 Representative Cities of China*

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Abstract

Objective To estimate the frequency of daily average PM_{10} concentrations exceeding the air quality standard (AQS) and the reduction of particulate matter emission to meet the AQS from the statistical properties (probability density functions) of air pollutant concentration.

Methods The daily PM_{10} average concentration in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an was measured from 1 January 2004 to 31 December 2008. The PM_{10} concentration distribution was simulated by using the lognormal, Weibull and Gamma distributions and the best statistical distribution of PM_{10} concentration in the 5 cities was detected using to the maximum likelihood method.

Results The daily PM_{10} average concentration in the 5 cities was fitted using the lognormal distribution. The exceeding duration was predicted, and the estimated PM_{10} emission source reductions in the 5 cities need to be 56.58%, 93.40%, 80.17%, 82.40%, and 79.80%, respectively to meet the AQS.

Conclusion Air pollutant concentration can be predicted by using the PM_{10} concentration distribution, which can be further applied in air quality management and related policy making.

Key words: Statistical distribution; PM_{10} concentration; Lognormal

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INTRODUCTION

The concentration of air pollutants usually varies and is influenced by the local pollutant emission levels and meteorological and topographical conditions[1-2], thus potentially resulting in different distribution patterns in different areas. The probability distribution can be used to predict the number of days when the ambient air quality standard (AQS) is exceeded and the pollutant emission source reduction to meet the specific AQS. It is necessary to use an appropriate type of statistical distribution to compute the exceeding probabilities and percentiles for setting regulatory targets and issuing environmental alerts for public health. Different types of probability distribution have been used to fit the air pollutant concentration, including lognormal[3-6], Weibull[7],

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Gamma[8], Log-logistic[9], type V Pearson[10], and extreme value[11] distributions. It is reported that lognormal distribution is the most appropriate distribution to represent PM_{10} concentrations[4,11]. However, few reports are available on the statistical distributions of daily average PM_{10} concentrations in developing countries' cities in Asia.

China, the largest developing country in the world, has experienced a rapid economic growth and social development since the reform and opening to outside world in the late 1970s. However, severe environmental problems have also appeared, and ambient air pollution is one of the most challenging environmental problems in the cities in China. Of the different air pollutants, particulate matter (PM) often shows the strongest evidence for adverse health effects[12]. PM_{10} (particulate matter less than 10 μm in aerodynamic diameter) is an air pollutant included in the China National Ambient AQS[13]. The total economic loss of PM_{10} pollution in China was US$ 29 178.7 million in 2004, accounting for approximately 1.5% of China's GDP in that year[14]. Therefore, air quality improvement programs are indispensable.

In this study, the statistical distribution characteristics of daily average PM_{10} concentration in Beijing, Guangzhou, Shanghai, Wuhan, and Xi'an during 2004-2008 were described, and the reduction of PM_{10} source emission to meet the AQS was estimated.

MATERIALS AND METHODS

Data

Twenty four-hour average PM_{10} concentra-ons were monitored routinely in Beijing, Guangzhou, Shanghai, Xi'an, and Wuhan of China (Figure 1).

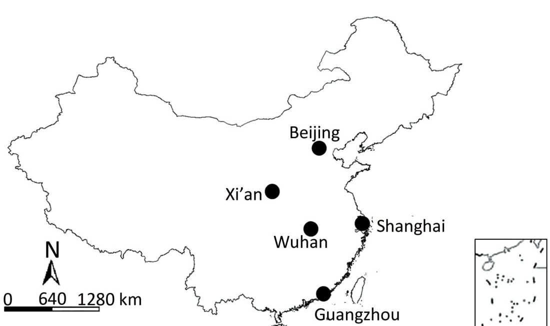

Figure 1. Location of Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an.

To avoid systematic error in maximum likelihood fitting, the PM_{10} concentrations in the 5 cities were measured from January 1, 2004 to December 31, 2008 as previously described[15].

Statistical Methods

Probability Density Functions Used in Representing PM_{10} Concentrations The lognormal, Weibull, Gamma, Log-logistic and type V Pearson distributions of the daily PM_{10} concentration were detected as previously described[3-9]. The values of lognormal, Weibull, and Gamma distributions were smaller than those of other distributions. In order to determine the best representative distributions of the daily PM_{10} concentrations in the 5 cities, 3 distributions were further analyzed. The parameters of lognormal, Weibull and Gamma distributions were estimated according to the daily average PM_{10} concentrations (Table 1).

Estimation of Parameters If a set of data follows a specific distribution, the parameters can be estimated from the measured data. The parameters of different distributions were estimated using the maximum likelihood method. If the probability density function computed from the measured data is p (x_i), the probability density function of theoretic distribution is p (x_i, y_1, y_2) where y_1 and y_2 are the parameters of the distribution, then the sum of the squares of errors (SSE) is:

$$SSE = \sum_{i=1}^{n} (p(x_i) - p(x_i, y_1, y_2))^2 \quad (1)$$

When the $\frac{\partial \ln SSE}{\partial y_1} = 0$ and the $\frac{\partial \ln SSE}{\partial y_2} = 0$, a maximum probability estimation equation of y_1 and y_2 can be obtained. The parameters estimated with the maximum likelihood method for theoretical lognormal, Weibull and Gamma distribution functions are presented in Table 2.

Goodness-of-fit In this study, sets of fit statistical data to measure how good the distribution fits the input data for all available functions were recorded after each dataset was simulated. The most commonly used goodness-of-fit tests are chi-square (χ^2), Kolmogorov-Smirnov (K-S), and Anderson-Darling (A-D) tests.

Table 1. Probability Density Function, Complementary Cumulative Distribution Functions (CCDF), and Lognormal, Weibull, and Gamma Distribution Functions

Distribution Function	Probability Density Function (PDF)	CCDF
Lognormal	$p_l(x_i) = \frac{1}{x_i \ln\sigma_g \sqrt{2\pi}} \exp\left[-\frac{(\ln x_i - \ln\mu_g)^2}{2(\ln\sigma_g)^2}\right]$	$\bar{F}_l(x) = 1 - \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\frac{\ln x - \ln\mu_g}{\ln\sigma_g}} e^{-t^2/2} dt$
Weibull	$p_w(x_i) = \frac{\lambda}{\sigma_w}\left(\frac{x_i}{\sigma_w}\right)^{\lambda-1} \exp\left[-\left(\frac{x_i}{\sigma_w}\right)^{\lambda}\right]$	$\bar{F}_w(x) = \exp\left[-\left(\frac{x_i}{\sigma_w}\right)^{\lambda}\right]$
Gamma	$p_g(x_i) = \frac{x_i^{a-1}}{b^a \Gamma(a)} \exp^{-(\frac{x_i}{b})}$	$\bar{F}_g(x) = 1 - \gamma(\frac{x}{b}, a)$

Note. For the lognormal distribution, μ_g is a location parameter and σ_g is a shape parameter. For Weibull distribution, λ is a shape parameter, σ_w is a location parameter and γ is an incomplete gamma function. For the Gamma distribution, a is a shape parameter and b is a location parameter.

Table 2. Parameters Estimated Using the Maximum Likelihood Method for Lognormal, Weibull, and Gamma Distribution Functions

Distribution Function	Estimate of Parameters
Lognormal	$\ln\mu_g = \frac{1}{n}\sum_{i=1}^{n} \ln x_i, \quad (\ln\sigma_g)^2 = \frac{1}{n}\sum_{i=1}^{n} (\ln x_i - \ln\mu_g)^2$
Weibull	$\lambda = \left[\left(\sum_{i=1}^{n} x_i\right)\right]$
Gamma	$\frac{d\ln\Gamma(a)}{da} = \ln\left(\frac{a}{n}\sum_{i=1}^{n} x_i\right) - \frac{1}{n}\sum_{i=1}^{n}(\ln x_i) \quad b = \frac{1}{na}\sum_{i=1}^{n} x_i$

Note. For the lognormal distribution, μ_g is a location parameter and σ_g is a shape parameter. For Weibull distribution, λ is a shape parameter, σ_w is a location parameter and γ is an incomplete gamma function. For Gamma distribution, a is a shape parameter and b is a location parameter.

The chi-square goodness-of-fit test can be applied to any univariate distribution for which the cumulative distribution function can be calculated and to binned data (i.e., data put into classes). Its statistical value which depends on how the data is calculated as:

$$X^2 = \sum_{i=1}^{r} \frac{(O_i - E_i)^2}{E_i} \quad (2)$$

Where O_i is the observed frequency of bin i and E_i is the expected frequency of bin i. In this study, how bins were defined for the chi-square test was fully controlled through bin setting in the fit distributions in data dialog. Bins were arranged in accordance with the equal probabilities, and the number of bins was produced autonomically.

Unlike the chi-square test, the K-S test is based on the empirical distribution function (EDF) and its statistical data are defined as:

$$D = \max_{1 \le i \le N} \left\| F(Y_i) - \frac{i}{N} \right\| \quad (3)$$

Where F is the theoretical cumulative distribution tested, which must be a continuous distribution, and N is the total number of data points. The K-S test tends to be more sensitive near the center of the distribution than at the tails. The A-D test is a modified K-S test and gives more weight to the tails than the K-S test. Its statistical data are defined as

$$S = \sum_{i=1}^{N} \frac{2i-1}{N}\left[\ln F(Y_i) + \ln(1 - F(Y_{N+1-i}))\right] \quad (4)$$

Where F is the cumulative distribution function of the specified distribution, Y_i is the ordered data.

Reduction of Pollutant Emissions to Meet the AQS

Assuming an unchanged spatial distribution of emission sources, meteorological condition and no chemical reactions in the particulate matter, the pollutant emission source reduction R (%) to meet the AQS is often calculated according to the rollback equation[7]:

$$R = \frac{E\{C_p\} - E\{C\}_s}{E\{C_p\} - C_b} \quad (5)$$

Where $E\{C\}_s$ is the mean (expected) distribution concentration when the extreme value equals Cs (the concentration of the AQS), $E\{C_p\}$ is the mean actual distribution concentration and C_b is the background concentration (often neglected). If Cs=150 μg/m^3, the PM_{10} daily average concentration is not exceeded more than once per year [(PM_{10}>Cs) =1/365=0.00274] then E{C}s is the expected daily average PM_{10} distribution concentration, where the probability of a concentration exceeding 150 μg/m^3 equals 0.00274[16].

To estimate the emission source reduction of a pollutant according to the rollback equation, one needs to know the type of distribution that best fits the pollutant concentration.

RESULTS

Variability of Measured Data with Time

The basic statistical data of PM_{10} concentration in the 5 cities are listed in Table 3. During the study period, the mean daily PM_{10} concentration was 145.09 μg/m^3, 98.32 μg/m^3, 81.21 μg/m^3, 122.65 μg/m^3, and 131.03 μg/m^3, respectively, in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an. The estimated parameters for the 3 theoretical daily PM_{10} concentration distributions in the five cities from 2004 to 2008 are listed in Table 4.

In this study, the properties of daily air quality data in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an were compared. The daily average PM_{10} concentration varied with time (2004-2008) is shown in Figure 2A-E. Figure 2 indicates that the daily PM_{10} concentration varied with the seasons. The daily average PM_{10} concentration limit is 150 μg/m^3 in China National Ambient Air Quality Standard. The frequency of exceeding the AQS in Guangzhou was

Table 3. Descriptive Statistical Data of Daily PM_{10} Concentrations in the 5 Cities (μg/m^3)

City	Number	Mean	Std	Minimum	P25	Median	P75	Maximum
Beijing	1796	145.09	91.42	9.00	80.00	128.59	183.50	600
Shanghai	1805	98.32	58.72	15.00	56.50	85.57	124.00	632
Guangzhou	1767	81.21	45.02	8.00	47.00	72.18	106.00	355
Wuhan	1796	122.65	52.59	23.00	82.00	115.54	152.00	350
Xi'an	1808	131.03	55.72	31.00	94.00	122.03	150.00	456

Table 4. Estimated Parameters of 3 Theoretical Distributions

City	Lognormal		Weibull		Gamma	
	μ_g	σ_g	σ_w	λ_w	σ_{ga}	λ_{ga}
Beijing	145.29	4.535	152.32	1.53	56.96	2.43
Shanghai	98.59	4.109	92.51	1.45	36.73	2.28
Guangzhou	81.38	3.849	83.01	1.69	26.36	2.85
Wuhan	131.51	3.996	112.09	1.44	29.76	3.54
Xi'an	122.80	4.029	115.30	2.02	26.80	4.06

Note. For the lognormal distribution, μ_g is a location parameter and σ_g is a shape parameter. For the Weibull distribution, λ is a shape parameter, σ_w is a location parameter and γ is an incomplete gamma function. For the Gamma distribution, a is a shape parameter and b is a location parameter.

lowest and the air pollution was worst in Beijing compared with the other 4 cities. Generally, the PM_{10} concentrations are higher in winter and lower in summer.

Best Distribution Detection

The A-D, K-S and Chi-square values and the best fit probability distribution of PM_{10} concentrations in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an are shown in Table 5. The smaller values indicated a better fit with the actual data. The A-D value was considered as the main criterion for goodness-of-fit and the lognormal distribution was most suitable for K-S and A-D values.

The best fit probability distribution of PM_{10} concentrations in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an from 2004 to 2008 are shown in Table 6. The fit quality could be quantified according to the A-D test criteria. The distribution with the smallest A-D test value represents the daily average PM_{10} concentration in the whole single year and the lognormal distribution was still the most suitable value while the Gamma distribution was a common value of A-D test.

The fitted results of 3 theoretic distributions and the measured data of PM_{10} in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an from 2004 to 2008 are presented in Figure 3.

Table 5. Probability Distribution of Best Fit for Daily PM_{10} Concentrations in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an

City	Distributions	K-S value	A-D value	Chi-square
Beijing	Lognormal	0.058*	2.350*	236.18
	Gamma	0.068	2.714	207.974*
	Weibull	0.084	8.783	303.94
Shanghai	Lognormal	0.026*	1.111*	59.985
	Gamma	0.027	1.427	56.090*
	Weibull	0.049	9.048	124.68
Guangzhou	Lognormal	0.038*	2.309	93.196
	Gamma	0.039	1.746*	86.664*
	Weibull	0.042	5.579	127.915
Wuhan	Lognormal	0.036	2.335	160.797
	Gamma	0.032*	1.442*	163.517
	Weibull	0.038	2.349	116.793*
Xi'an	Lognormal	0.082*	5.232*	271.062*
	Gamma	0.094	7.770	298.908
	Weibull	0.127	20.789	350.948

Note. *The best fit probability distribution under this criterion.

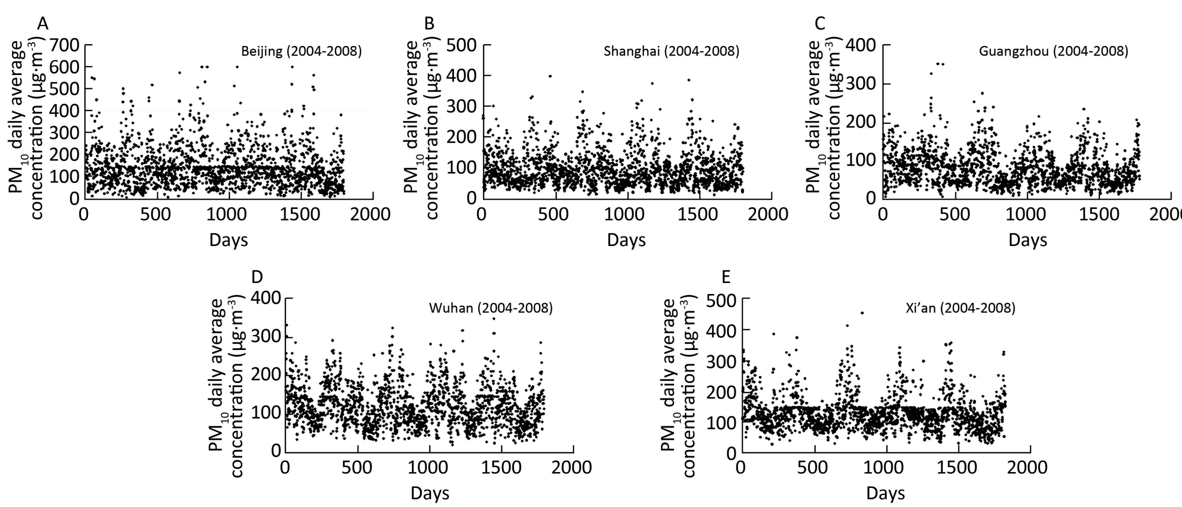

Figure 2. Variation of daily average PM_{10} concentrations in Beijing (A), Shanghai (B), Guangzhou (C), Wuhan (D), and Xi'an (E) from 2004 to 2008.

Table 6. Best Fit Distribution of PM_{10} Concentrations in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an according to the A-D Test Criteria (2004-2008)

City	2004	2005	2006	2007	2008
Beijing	Lognormal	Gamma	Lognormal	Lognormal	Gamma
Shanghai	Lognormal	Lognormal	Gamma	Lognormal	Gamma
Guangzhou	Gamma	Lognormal	Gamma	Gamma	Lognormal
Wuhan	Gamma	Weibull	Gamma	Gamma	Weibull
Xi'an	Lognormal	Lognormal	Lognormal	Lognormal	Gamma

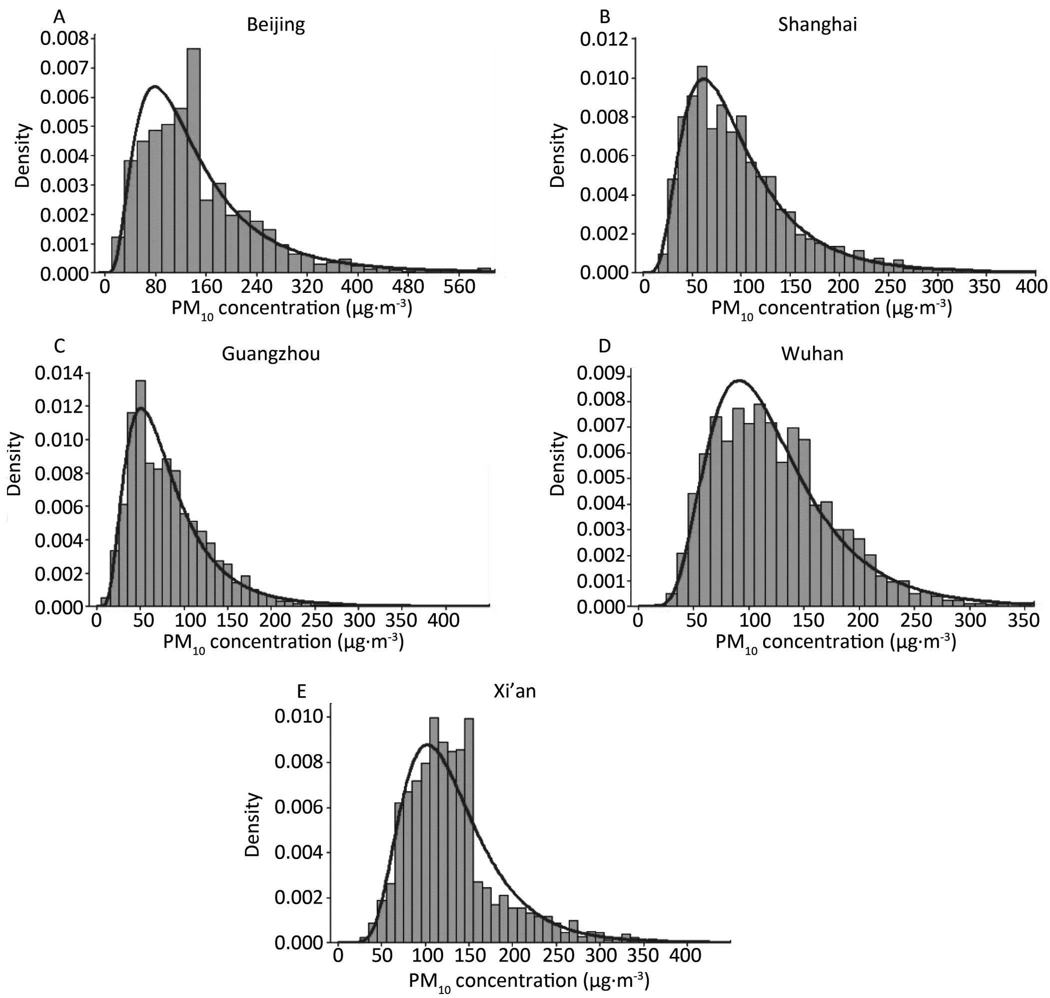

Figure 3. Best fit statistical distributions of daily average PM_{10} concentrations in Beijing (A), Shanghai (B), Guangzhou (C), Wuhan (D), and Xi'an (E).

Estimation of Exceeding AQS Probability

The exceeding AQS probability could be predicted by using the predetermined distribution parameters. The actual times for which the daily average PM_{10} concentration was over 150 μg/m^3 in Beijing, Shanghai, Guangzhou, Xi'an, and Wuhan during 2004-2008 were 586, 262, 136, 392, and 455 days, respectively. The predicted times according to the estimated distribution were 618, 262, 147, 388, and 455 days, respectively. Thus, the lognormal distribution is an appropriate distribution to predict the number of days when PM_{10} concentrations exceed the AQS. The results could be used for predicting the exceeding days in the succeeding year with variation of air quality in each city.

Estimation of Emission Source Reduction

The reduction of pollutant emission sources to meet the AQS in Beijing, Shanghai, Guangzhou, Xi'an, and Wuhan could be estimated by using the best fit statistical distribution of air pollutant concentration. The parameters of distributions controlling the fluctuation sizes were not influenced by the pollutant emission level, unlike the mean (expected) value of the daily average PM_{10} concentration ($E\{C_p\}$).The actual mean PM_{10} concentrations in the 5 cities should be reduced in order to meet the AQS. The relation between μ_g and mean concentration in the lognormal distribution could be expressed as

$$\ln E\{C_p\} = \ln \mu_g + \frac{1}{2}(\ln \sigma_g)^2 \qquad (7)$$

The relation between the mean (expected) PM_{10} concentration and the exceeding AQS probability for the lognormal distribution was studied by using the parameter σ_g. Since the PM_{10} background concentration was not reliably estimated in the 5 cities, it was set as $C_b = 0$.

Taking Guangzhou as an example, the value of $E\{C\}_s$ could be calculated as 35.26 μg/m^3 according to the density function of the lognormal distribution. The mean PM_{10} concentration should thus be reduced from the current value of 81.21 μg/m^3 to 35.26 μg/m^3, and the estimated pollutant emission reduction could be calculated as:

$$R = \frac{E\{C_p\} - E\{C\}_s}{E\{C_p\} - C_b} = \frac{81.21\,\mu g/m^3 - 35.26\,\mu g/m^3}{81.21\,\mu g/m^3} = 0.5658 \qquad (6)$$

For the lognormal distribution, the estimated pollutant source emission reduction required to meet the AQS was 56.58%, 93.40%, 80.17%, 82.40%, and 79.80%, respectively, in Guangzhou. Beijing, Shanghai, Xi'an, and Wuhan, indicating that the pollutant source emissions should thus be more strictly controlled in order to reduce the PM_{10} concentration and meet the AQS.

DISCUSSION

In this study, the statistical distribution of daily PM_{10} concentration during 2004-2008 was investigated in 5 cities of China, the actual data of different distributions were fitted and the parameters of each distribution were estimated by using mathematical method, the best fit distribution in each city was determined by 3 goodness-of-fit tests.

The best daily PM_{10} concentration distribution in Beijing, Shanghai, Guangzhou, Wuhan, and Xi'an was the lognormal distribution, which has long been considered as the most appropriate distribution type and was most frequently used to represent air pollutant concentrations[17], and is consistent with the results of a number of previous researches[3-6]. The WHO air quality guidelines (AQG) also applied lognormal distribution to compute exceeding probabilities and percentiles for setting regulatory guidelines and interim targets because the lognormal distribution was related with the normal distribution and theoretical and physical mechanistic arguments support its use when air-pollutant data are analyzed[18]. However, the best-fit distribution in this study may differ from those in previous studies. The reason of this inconsistency is still unclear but can potentially be explained as follows. First, the distribution of air pollutant concentrations is a specific case in different areas and influenced by local pollutant emission levels, meteorological and geographical conditions[11]. Second, the simulation software, Oracle Crystal Ball used in this study, has a library of more than 20 types of continuous distribution functions and can provide more alternatives to fit. Third, some distribution functions are of the identical properties and the disparities between them may be considerably small.

Each of the 3 goodness-of-fit tests we used has strengths and weaknesses, thus directly leading to the instability and inconsistency in which the best fit probability distribution may vary with the type of fit test. It was reported that the empirical distribution function (such as K-S and A-D tests) is more powerful than the chi-square test[19]. The χ^2 and K-S tests are commonly used to decide whether the hypothesized distribution can be rejected. The low ambient air pollution level is still associated with adverse health effects[12,20-21] and mostly located in the center of a probability distribution. The tail properties of a distribution are important in predicting the exceeding probability in order to meet a threshold concentration[17]. Nevertheless, the distribution frequency of air pollutants cannot fit the high concentrations and predicting errors may be ignored[5]. Therefore, adding the A-D statistical data into the multiplicative aggregation test can increase the applicability and effectiveness of the selected best statistical distributions.

The problem reflected in this study was highly worthy of attention. In this study, we assumed that the daily average PM_{10} concentration was linear with the emission level and calculated the estimated percentage of pollutant emission source reduction. Even in Guangzhou, the 'cleanest' city in China, the pollutant emission needs to be reduced by over 50% to meet the AQS. China is one of the countries with the highest ambient particle levels in the world due to its rapid economic development and urbanization. Totally 113 cities in China were selected as 'the national key environmental protection cities' in 2007 and measures were taken to monitor the air quality in these cities. The air quality was improved in most cities due to the use of new and clean energy. Nevertheless, the annual average PM_{10} concentration in the 113 cities was 85 μg/m^3 while the annual average PM_{10} concentration in A-D was 70 μg/m^3[23]. It is still a great challenge for China to balance the environmental protection and economic development.

Our study provided a feasible method for estimating the statistical distributions of daily average PM_{10} concentration for a single city in China. More multi-center researches could be conducted in specific regions of China and in different seasons by using the same method to determine the particular statistical distribution types of daily average PM_{10} concentration. The results could help to estimate the exceeding probabilities and percentiles and to issue the environmental alerts for the public. Some potential suggestions were put forward in this study for the control of air pollution. The short-term (daily) air quality standard was defined as the mass concentration with evidence-based averaging times for the lowest pollutant level associated with acute adverse effects during temporary exposure, and the long-term (annual) criteria for the maximum permissible pollutant levels were established according to the lowest adverse effect level following a systematic review of toxicological, clinical and epidemiological studies[24], suggesting that even if the daily average concentration in one city could meet the standard for daily average PM_{10} concentration in a year, the annual average PM_{10} concentration might not meet the standard. The WHO indicated that the concentration-time relationship between short-term and annual limits for an individual pollutant is not fully understood. Our findings suggest that the lognormal distribution can provide a robust estimation for daily PM_{10} concentration. More studies on the statistical distribution of different air pollutants are needed to show the relationship between daily average concentration and annual average concentration and its allowable exceeding level. Nationwide cohort studies on the adverse health effects of air pollutants are also needed to provide suggestions for the control of air quality and emission reduction.

In conclusion, the best fit distribution for daily PM_{10} concentration in the 5 cities of China is the lognormal distribution. Further study is needed to reduce the emission of air pollutants to meet the AQS and to show the statistical distribution of air pollutants in different regions of China.

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