MODELING AND PREDICTION OF PM_{10} CONCENTRATION DURING HIGH PARTICULATE EVENTS IN MALAYSIA

by

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α	shape parameter
β	scale parameter
δ	location parameter
f(x), pdf	probability density function
F(x), cdf	cumulative distribution function
L	likelihood function
Γ	gamma function
$x_1, x_2, \ldots x_n$	random variables
$\bar{x}$	mean
s	variance
ff	frequency factor
cfd	central fitting distribution
evd	extreme value distribution
MLE	maximum likelihood estimation
MoM	method of moments
MoP	method of percentile
ProbPlot	probability plot
MAE	mean absolute error
NAE	normalised absolute error
PA	prediction accuracy
IA	index of agreement
R^2	coefficient of determination
n	total number of measurement of particular site

P_i	Predicted values for one set of annual monitoring records
O_i	Observed values for one set ofannual monitoring records
$\overline{P}$	Mean of the predicted values for one set of annual monitoring records
$\overline{O}$	Mean of the observed values for one set of annual monitoring records
S_{pred}	Standard deviation of the predicted values for one set of annual monitoring records
S_{obs}	Standard deviation of the observed values for one set of annual monitoring records
ED1	extreme data 1
ED2	extreme data 2
ED3	extreme data 3
SP	Seberang Perai
NL	Nilai
MK	Melaka
KU	Kuching
JT	Jerantut

PEMODELAN DAN PERAMALAN KEPEKATAN PM_{10} KETIKA KEADAAN PARTIKEL TINGGI DI MALAYSIA

ABSTRAK

Partikel merupakan pencemar utama ketika jerebu berlaku. Kajian mengenai bahan pencemar ini adalah penting kerana kesannya yang mengganggu kesihatan manusia dan juga persekitaran. Objektif utama kajian ini adalah untuk menghasilkan model bagi meramal kepekatan PM_{10} ketika keadaan partikel di udara tinggi dengan menggunakan kaedah taburan kebarangkalian. Kajian ini menggunakan rekod pemantauan PM_{10} dari tahun 1997 hingga 2006 yang merangkumi tahun berjerebu dan tidak berjerebu di lima stesyen pemantauan. Tiga daripada stesyen tersebut iaitu Seberang Perai, Nilai dan Kuching mewakili kawasan perindustrian. Stesyen yang terletak di Melaka pula mewakili kawasan perumahan manakala stesyen di Jerantut merupakan stesyen rujukan. Selain daripada punca tempatan, kepekatan PM_{10} di semua kawasan kajian berada pada tahap tertinggi ketika monsun kering (monsun barat daya – Jun hingga September) disebabkan pencemar yang diterbangkan oleh angin daripada Indonesia. Tiga taburan penyuaian tengah iaitu taburan Weibull, gamma dan lognormal digunakan untuk disesuaikan dengan rekod pemantauan PM_{10}. Taburan terbaik dipilih berdasarkan petunjuk prestasi. Bagi kepekatan PM_{10} yang tinggi, taburan nilai ekstrem (taburan Gumbel dan Frechet) digunakan untuk disesuaikan dengan data ekstrem. Taburan lognormal didapati sesuai untuk semua rekod pemantauan kecuali rekod pemantaun PM_{10} yang mempunyai nilai kepencongan yang tinggi di Seberang Perai untuk tahun 2001, 2003, 2004 dan 2005 yang mana ianya lebih baik disesuaikan dengan taburan gamma. Keputusan yang diperoleh untuk taburan penyuaian tengah menunjukkan bahawa nisbah ramalan tempoh ulangan dengan tempoh ulangan sebenar menghampiri nilai 1 untuk tahun

yang tidak berjerubu. Sebaliknya, untuk tahun berjerubu, taburan kebarangkalian ekstreme menggunakan taburan Gumbel dan Frechet memberikan penyuaian yang lebih baik terhadap data ekstrem. Data kepekatan harian maksimum pula merupakan data ekstrem yang terbaik dan membolehkan tempoh ulangan ditentukan dalam unit hari. Selain itu, kepekatan PM_{10} di semua stesyen kajian telah direkod menggunakan E-sampler. Data yang diperoleh menggunakan E-sampler dihubungkaitkan dengan data BAM menggunakan regresi linear. k-faktor untuk Seberang Perai dikenalpasti untuk lima kumpulan data berdasarkan peratus dan digunakan untuk stesyen lain. Keputusan menunjukkan ramalan E-sampler untuk stesyen lain adalah bagus menandakan bahawa E-sampler boleh digunakan sebagai alat alternatif yang berkos rendah bagi menggantikan pemantauan PM_{10} di peringkat tempatan. Kerja-kerja memprofail yang telah dijalankan dalam lingkungan 5 km daripada setiap stesyen juga telah membantu untuk mengenalpasti punca-punca yang menyebabkan pencemaran udara di setiap stesyen kajian.

MODELING AND PREDICTION OF PM_{10} CONCENTRATION DURING HIGH PARTICULATE EVENTS IN MALAYSIA

ABSTRACT

Particulate matter is the major pollutant during haze events. Study on this air pollutant is crucial due to its detrimental effects to human health as well as the environment. The aim of this research is to model and predict PM_{10} concentrations during high particulate events (haze event) using probability distribution methods. PM_{10} monitoring records from 1997 to 2006 that include years with and without haze for five stations were used in this research. Three stations represent the industrial areas of Seberang Perai, Nilai and Kuching. One station represents residential area located in Melaka and another station is the reference station in Jerantut. Apart from local sources, PM_{10} at all sites were found to have the highest concentrations during the dry season (south west monsoon - June to September) as the effect of transboundary pollution carried by winds from Indonesia. Three central fitting distributions, Weibull, gamma and lognormal distributions, were used to fit PM_{10} observation records. The best distribution was selected based on performance indicators. For high PM_{10} concentrations, extreme value distributions (Gumbel and Frechet distributions) were applied to fit the extreme data. The lognormal distribution fits the observed data well in all sites except for PM_{10} concentrations with high skewness in Seberang Perai for 2001, 2003, 2004 and 2005, which were better fit with the gamma distribution. The results for central fitting distribution show that ratios of predicted return periods to actual return periods were close to 1 for years without haze events. However, for years with haze events, extreme value distribution using Gumbel and Frechet distributions give a better fit for the monitoring records. Daily maximum data is the best extreme data that allows return

period to be estimated in unit days. In addition, PM_{10} concentrations at all research sites were monitored using E-sampler. The results obtained from E-sampler (*y*) were correlated with PM_{10} monitoring records from BAM (*x*) using linear regression. K-factors for Seberang Perai were identified for five different groups based on percentile and were used for other stations. The result shows that predicted E-sampler for other sites is also good, which indicates that monitoring using E-sampler can be used as a low cost alternative for PM_{10} monitoring at local governance levels. Profiling work conducted within a 5 km radius of every monitoring station helped to identify the possible sources of pollution at all sites.

CHAPTER 1

INTRODUCTION

1.1 AIR POLLUTION IN MALAYSIA

Air pollution is the presence of undesirable material in air, in quantities large enough to produce harmful effects to human health, vegetation, human property, or the global environment, as well as create aesthetic insults in the form of brown or hazy air or unpleasant smells (de Nevers, 2000). In Pennsylvania, air pollution is defined as any form of contaminant including but not limited to the discharging from stacks, chimneys, opening, buildings, structures, open fires, vehicles, processes, or any other source of any smoke, soot, fly ash, dust, cinders, dirt, noxious or obnoxious acids, fumes, oxides, gases, vapors, odors, toxic or radioactive substances, waste or any other matter in such place, manner or concentration inimical or which may be inimical to the public health, safety, or welfare or which is, or may be injurious to human, plant or animal life, or to property, or which unreasonably interferes with the comfortable enjoyment of life or property (Heinsohn and Kabel, 1999). In the Malaysian Environmental Quality (Clean Air) Regulations, 1978, air pollution is termed as air impurities, which includes smoke, soot, dust, ash (including flyash), cinders, grit, solid particles of any kind inclusive of particulates, gases, fumes, mist, odours and radioactive substance which are generated as a result of combustion of fuel and the like, or a result of the use of electricity as a heat source, or a result of synthesis, resolution or any other treatment and any other substance, which may be designated by the Minister as those which are liable to affect adversely the human health or the living environment.

Air quality in Malaysia is reported based on the air pollution index (API) system which is obtained from the measurement of PM_{10} and several other gas pollutants that include sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ozone (O_3) and carbon monoxide (CO). Table 1.1 shows the API for Malaysia and compares them with the level of pollution and health measures. An API breakpoint at 100 corresponds to the respective recommended Malaysian Ambient Air Quality Guideline (MAAQG) concentration (Table 1.2), regarded as being safe levels for human health. In contrast, API values exceeding 100 are likely to cause negative health effects for the general population. In addition, Table 1.2 shows the ambient standards adopted by MAAQG and compares them with the National Ambient Air Quality Standards currently enforced in the United States, and WHO guidelines. The Malaysian guidelines are fairly consistent with the standards of the United States.

Based on API readings throughout the country, the air quality is considered as good to moderate, except for serious haze events that caused the API to reach hazardous levels, due to which in some areas, a haze emergency was declared. The main air pollution sources in Malaysia are from mobile sources, stationary sources that include industrial and power stations, open burning activities within the country that usually occur at solid waste dumping sites, plantation farms, accumulated small scale open burning by local people, and transboundary sources commonly from Indonesia.

Table 1.1 Comparison of API values with level of pollution and health measures

API scale	Status	Level of Pollution	Health Measures
0 - 50	Good	Pollution low and has no ill effects on health.	- No restriction of activities for all groups of people - To practice healthy lifestyle e.g. not to smoke, exercise regularly and to observe proper nutrition.
51 - 100	Moderate	Moderate pollution and has no ill effects on health.	- No restriction of activities for all groups of people - To practice healthy lifestyle e.g. not to smoke, exercise regularly and to observe proper nutrition.
101- 200	Unhealthy	Mild aggravation of symptoms among high risk persons e.g. those with heart or lung disease.	- Restriction of outdoor activities for high risk person. - General population should reduce vigorous outdoor activities.
201- 300	Very unhealthy	Significant aggravation of symptoms and decreased exercise tolerance in persons with heart or lung disease.	- Elderly and person with known heart or lung disease should stay indoors and reduce physical activities. - General population should reduce vigorous outdoor activities. - Those with any health problems should consult doctor.
301- 500	Hazardous	Severe aggravation of symptoms and endangers health.	- Elderly and persons with existing heart or lung disease should stay indoors and reduce physical activities. - General population should reduce vigorous outdoor activities.
Above 500	Emergency	Severe aggravation of symptoms and endangers health.	- General population advised to follow the orders of the National Security Council and always to follow the announcements through the mass media.

(Source: Department of Environment, 2009)

Table 1.2 Ambient Air Quality Standards – Malaysia, United States and WHO

Air pollutants	**Malaysia**[a] **(µg/m³)**	**USEPA**[b] **(µg/m³)**	**WHO**[c] **(µg/m³)**
Carbon monoxide (CO)			
8-h average	10 000	10 000	10 000
1-h average	35 000	40 000	30 000
Nitrogen dioxide (NO_2)			
Annual	-	100	40
1-h average	320	-	200
Ozone (O_3)			
8-h average	120	-	100
1-h average	200	240	N/S
Particulate matter (PM_{10})			
Annual	50	N/S	20
24-h average	150	150	50
Sulfur dioxide (SO_2)			
Annual	-	80	N/S
24-h average	105	365	20

N/S – Not Specified
[a] DoE (2007)
[b] USEPA (2009)
[c] WHO (2006) except for CO from WHO (2009)

Figure 1.1 shows the number of in-use vehicles in Malaysia that increased slightly from 2004 to 2007. The number of in-use or active vehicles was determined through annual road tax renewal. Vehicles that do not renew road tax for three consecutive years are considered as non active vehicles by the Road Traffic Department (RTD). The average percent increment of in-use vehicles every year from 2004 to 2007 is 10.8%. In 2005, the number of in-use vehicles increased 10.5% from 2004. In 2006, the percent increment dropped to 7% but increased about twice in 2007 to 15%.

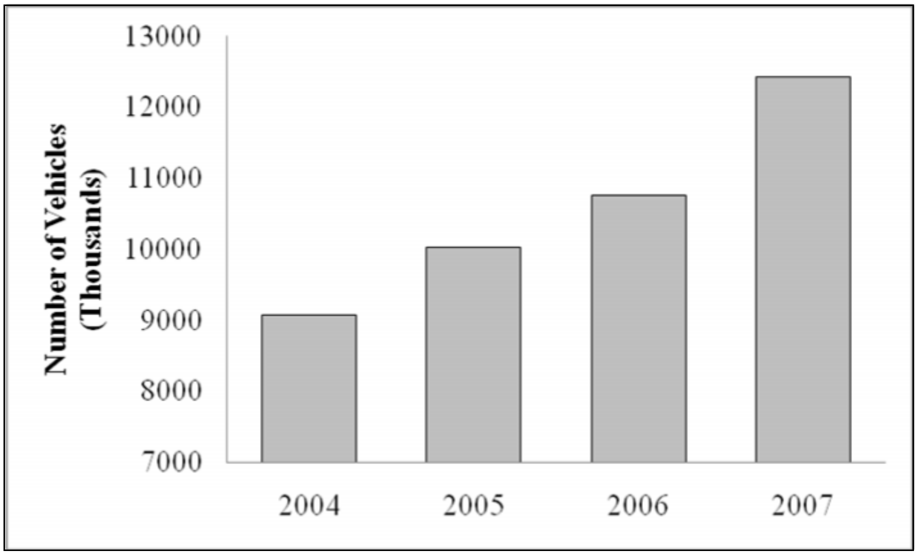

Figure 1.1 Number of in-use vehicles in Malaysia
(Determined through annual road tax renewal)

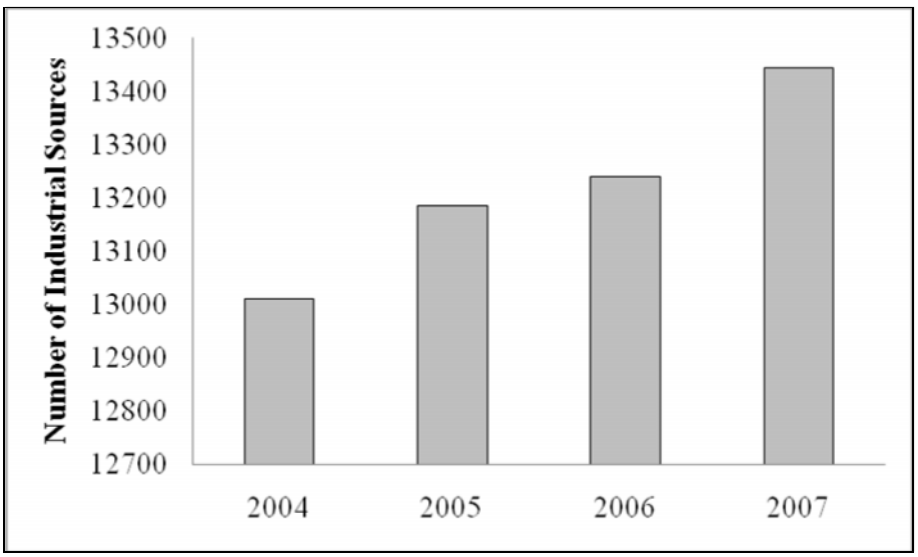

Figure 1.2 Number of industrial related sources contributing to air pollution in Malaysia

The number of industrial air pollution sources identified, which were subjected to Environmental Quality (Clean Air) Regulations 1978, increased from 2004 to 2007 (Figure 1.2). The percentage of increment in industrial sectors that contribute to air pollution problems is small. In 2005, the percentage of increment was 1.3%. For 2006, the percent increment was only 0.4% and in 2007, the percent increment increased to 1.5%. Even though industrial sectors only slightly increased from 2004 to 2007, when combined with power stations, they become stationary sources that mainly contribute to the PM_{10} emission load from 2004 to 2007. Figure 1.3 illustrates PM_{10} emission by sources in Malaysia in metric tonnes (mt) from 2004 to 2007. Industrial and power stations contribution to the PM_{10} emission load is 58% in 2004, 67% in 2005, 69% in 2006 and 54 % in 2007. Motor vehicles are the second highest contributor to the PM_{10} emission load in 2004 (31%), 2005 (20%) and 2007 (28%). In 2006, the second highest contributors are open burning and transbouandary pollution (others).

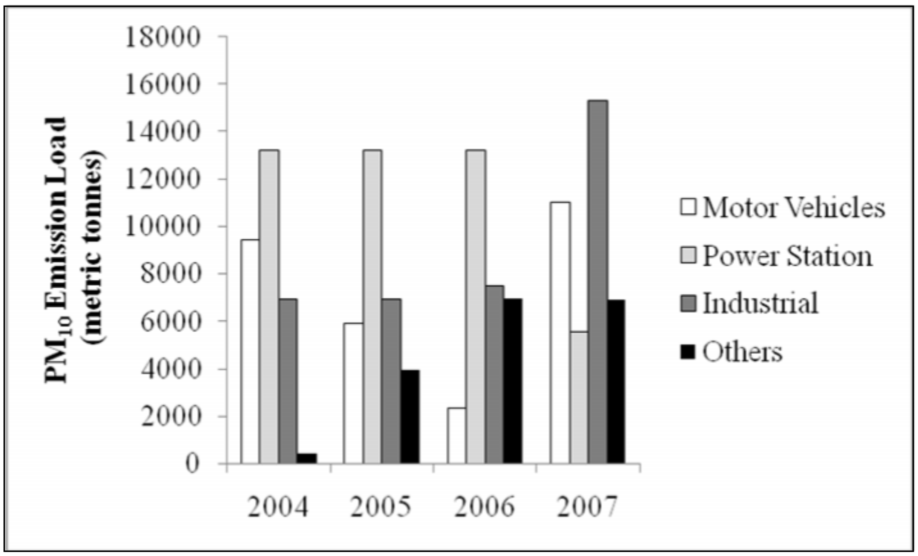

Figure 1.3 PM_{10} emissions by sources in Malaysia from 2004 to 2007

Aware of the great impacts caused by the air pollution phenomena, Malaysia is also involved in the work to control and to overcome the situation. The Department of Environment (DoE), under The Ministry of Natural Resources and Environment Malaysia, is the responsible authority in monitoring, controlling and enforcing the rules related to the environment.

In 1997, 16 new Continuous Air Quality Monitoring (CAQM) stations were set up by the DoE Malaysia, in addition to the existing 13 stations (DoE, 1997). Parallel with the rapid development in Malaysia, the increasing numbers of industrial activities associated with the increasing number of vehicles result in the need for monitoring work. Therefore, up to 2006, the total monitoring stations in the country are 51 stations that are strategically located in both residential and industrial areas. These include one reference station located in Jerantut, Pahang. The parameters monitored include Total Suspended Particulates, Particulate Matter (PM_{10}), Sulfur Dioxide (SO_2), Nitrogen Dioxide (NO_2), Ozone (O_3), Carbon Monoxide (CO), and lead (DoE, 2006).

1.2 PROBLEM STATEMENT

High particulate event in Malaysia is believed to be caused by the occurrences of haze events either originated from the neighbouring country or from local sources. Haze is defined as visibility impairment caused by the emission of air pollutants from numerous sources (ADEQ, 2009). Since 1980s, haze event have been recorded in Malaysia (Radojevic, 2001). Following that, there were several major fire outbreaks in South East Asia with small fires occurring almost annually (Qadri, 2001) which had caused high particulate event in Malaysia. The worst haze event occurred in Malaysia is in 1997 and 1998 caused by forest fires in Kalimantan, Indonesia (Nichol, 1998) destroyed an estimated 9 million hectares (ha) of land and forest area in Indonesia alone (Qadri, 2001). The fire emergency situation due to haze was declared in the state of Sarawak for 10 days from 19 September 1997 when API reached hazardous level and visibility was greatly reduce. The most recent serious smoke haze was observed in 2005 where haze emergency was declared in Pelabuhan Klang and Kuala Selangor when the API in both areas exceeded 500 on 11th August 2005. In consequences, PM_{10} is increasingly perceived to be a serious threat to human health during the haze event. Other effects include reduced visibility, transport disruption and economic losses (Qadri, 2001). Reported haze events in Malaysia since 1997 are listed in Table 1.3.

During forest fires, many combustion products were released into the atmosphere such as particulate matter, polycyclic aromatic hydrocarbons (PAHs), carbon monoxide, aldehydes, organic acids, semivolatile and volatile organic compounds, free radicals, ozone, inorganic fraction of particles, trace gases and other releases, and radionuclide (Malilay, 1998). Among these products, particulate matter was found as the main pollutant during haze episodes by many researchers (Muraleedharan et al., 2000, Abas, et al., 2004). Department of Environment Malaysia has also confirmed that the dominant pollutant during haze episode was PM_{10}. Furthermore, a continuous increase in the number size distribution of particles with diameters larger than 300nm is associated with visibility decrease (Bäumer et al., 2008). Therefore, high particulate matter concentration in the atmosphere is highly responsible for the occurrences of haze event. Kim et al., (2006) reported that visibility was well correlated with increases in mass concentration of sulphate, nitrate and elemental carbon (EC) particles. The average light extinction coefficient was high due to increase loading of fine particles in the atmosphere. Moreover, the present of black carbon (BC) with significant contributors from brown carbon and mineral dust have dominated to light absorption in the atmosphere. Source of this absorbing aerosols include biomass burning, combustion processes and dust entrainment (Moosmuller et al., 2009). Study by Noh et al., (2009) had also found that the greatest differences in optical and microphysical parameters of aerosols observed under severe haze condition in Gwangju, Korea were found in light absorbing properties.

1.3 OBJECTIVES

This research was carried out with four main objectives, that is:

i. To determine the characteristics of PM_{10} concentration and to investigate when the high PM_{10} concentration occurs using time series plot.
ii. To compare the fitted distributions of Weibull, gamma and lognormal distributions on predictions of PM_{10} concentration during years with haze events and years without haze events.
iii. To use Gumbel and Frechet distribution to predict high PM_{10} concentrations.
iv. To monitor primary PM_{10} concentration using simple instruments and to compare with standard instruments used by the Department of Environment, Malaysia, in order to introduce a cheap and simple method of monitoring PM_{10} concentration.

Table 1.3 Reported haze event in Malaysia since 1997

Year	Reported haze event			Reference
	Time	Areas most affected	Cause	
1997	Mid July to November	Sarawak and Klang Valley	Primarily due to forest and land fires in Kalimantan and Sumatra, Indonesia aggravated by unusual prolonged severe drought cause by El-Nino which started in March 1997. Reached a climax when a haze emergency had to be declared in the state of Sarawak for 10 days starting 19 Sept. when API reached 500 (hazardous level) and visibility was greatly reduce.	DoE, 1997
1998	March	Miri and Kota Kinabalu	Due to high concentration of PM_{10} from the forest fires around Miri and coupled with the dry weather conditions.	DoE, 1998
2002	July to September	Sarawak	Major fires in Kalimantan burning 1.2 to 1.5Mha during August to November 2002.	Heil, 2007; DoE, 2002
2004	June, August and September	West coast of Peninsular Malaysia and Southern part of Sarawak	Land and forest fires in several provinces in Sumatra coupled with the direct influence of south westerly winds.	DoE, 2004
2005	i)February and March ii) Mid May until mid October	i) Some areas in Klang Valley ii) Central, eastern and northern part of peninsular Malaysia experienced severe haze	i) Peat land fires in several areas in the state of Selangor. ii) Peat land fires in Selangor, land and forest fires in Sumatra and prolonged dry season in the region and south westerly wind. Haze emergency was declared in two areas in Klang Valley, namely Pelabuhan Klang and Kuala Selangor when the API in both areas exceeded 500 on 11th August 2005.	DoE, 2005
2006	July to October	Northern and Southern part of west cost of Peninsular Malaysia and Sarawak	Land and forest fires in several provinces in Sumatra and Kalimantan coupled with the direct influence of south westerly winds.	DoE, 2006

1.4 SCOPE OF RESEARCH

DoE recorded PM_{10} concentrations at 51 monitoring stations throughout the country. From these, ten years of PM_{10} monitoring records (1997 to 2006) that include years with and without haze event from five stations were selected to be analysed and modeled using probability distributions. The stations are Seberang Perai in Penang, Nilai in Negeri Sembilan, Bachang in Melaka, Kuching in Sarawak, and Jerantut in Pahang.

There are many types of theoretical distributions to fit the air pollutant concentration data (Lu, 2003). The most common distributions used in interpreting air pollutant data are the Weibull distribution, Gamma distribution and lognormal distribution. All these distributions were used in this research.

Every set of air pollutant data has different characteristics. Therefore, in this research, all types of distributions mentioned will be used to find the best distribution that will fit the data. In addition to that, extreme value distributions such as the Gumbel distribution and Frechet distribution will be developed to model extreme cases of PM_{10} concentration. Goodness of fit using performance indicators will be utilized to find the best model.

The best models are selected based on goodness-of-fit results to represent the PM_{10} monitoring records. It is then used to predict future PM_{10} concentration. The outcome will allow government and any other related bodies to prepare and to take action on mitigating the air pollution problem, especially the haze phenomena.

1.5 SIGNIFICANCE OF RESEARCH

Research on PM_{10} is significant due to its severe effects on human health and environment. Despite numbers of studies on PM_{10} carried out by researchers from Malaysia, statistical analysis is still lack in this country. There is abundance of pollutants data monitored by DoE Malaysia that need to be analyzed comprehensively and is best assist with statistical analysis and modeling. This study offers the statistical modeling using probability distribution model as a tool that can be used to model PM_{10} monitoring records and to predict future PM_{10} concentration so that action can be planned and taken by related agencies and government to tackle high particulate event in future years. In addition to that, this research is the first work that deals with extreme concentration of PM_{10} in Malaysia and had given very promising results from the model developed. Moreover, results obtained were verified during the model verification on site using direct reading nephelometer. The result can be used by scientist and government to predict high PM_{10} occurrences in future,

1.6 STRUCTURE OF THESIS

Chapter 2 defines particulate matter and discusses the details of PM_{10} physical and chemical characteristics. In addition, weather influence, measurement principles and method, and also sources of PM_{10} are also discussed.

Chapter 3 discusses the importance of statistical analysis in environmental engineering. Besides that, central fitting distributions (Weibull, gamma and lognormal) and extreme value distributions (Gumbel and Frechet) used to fit PM_{10} monitoring records for this research, such as their application in environmental engineering and advantages of these distributions are also highlighted. The equation for the probability density function (pdf) and the cumulative distribution function (cdf) for all types of distributions used in this research are showed.

Chapter 4 describes the procedures employed in substitution of missing values, estimation of all distribution parameters, calculation method of the goodness of fits and estimation of the exceedences. The monitoring campaign for monitoring primary PM_{10} concentration using the E-sampler instrument is described. Calculation of the k-factor that relates PM_{10} concentration recorded by the E-sampler instrument and BAM1020 is also shown.

Chapter 5 shows the results of this research. The descriptive statistic and time series plot of PM_{10} monitoring records are discussed. Results obtained from each distribution, such as scale and shape parameters for all variables, are shown together with the cdf and pdf plots of every observed concentration in comparison with theoretical distribution. The best distributions that fits the monitoring records are selected based on performance indicator results and are used to estimate the predicted exceedences to compare with the actual exceedences. Analysis of the results of PM_{10} concentration recorded using E-sampler are also discussed.

Chapter 6 discusses the proposal for implementation of the Local Air Quality Management (LAQM) scheme in Malaysia, which includes the use of simple instruments (E-sampler) for monitoring PM_{10} concentration. The possible application of E-sampler in the Local Air Quality Management (LAQM) scheme is elaborated.

Chapter 7 concludes the research and listed the recommendations for further work.

CHAPTER 2

PARTICULATE MATTER

2.1 PARTICULATE MATTER

There are various definitions of the term particulate matter. Willeke and Baron (2001) define a particle as a single unit of matter, generally having a density approaching the intrinsic density of the bulk material. Otherwise, particulate matter, as defined by Godish (1997), is a collective term used to describe small, solid and liquid particles that are present in the atmosphere over relatively brief (minutes) to extended periods (days to weeks) of time. Therefore, particulate matter is important from its single unit to its behavior in the atmosphere. In addition, Van Der Wal & Janssen (2000) defined PM_{10} as inhalable particles with an aerodynamic diameter of approximately 10µm or less.

Particulate matter varies in size, shape and chemical characteristics. Besides, it can be classified into two types, primary and secondary particles, based on its origin and process of formation. Primary particles refer to particles that were directly emitted from a source into the atmosphere, from either natural or anthropogenic sources (QUARG, 1996). The major sources of primary particles are (i) petrol and diesel vehicles, the latter being the source of most black smoke, (ii) controlled emission from chimney stacks, and (iii) fugitive emissions. These are diverse and uncontrolled, and include the resuspension of soil by wind and mechanical turbulence, resuspension of surface dust from road and urban surfaces by wind, vehicle movements, and other local air disturbance, and emission from activities such as quarrying, road and building construction, loading and unloading of dusty material (Watt and Hamilton, 2003). On the other hand, secondary particles are formed in the atmosphere as a result of chemical processes involving gases, aerosol particles and moisture (Godish, 1997). The major sources are the atmospheric oxidation of sulphur dioxide droplets to sulphuric acid, and the oxidation of nitrogen dioxide vapour to nitric acid (Watt and Hamilton, 2003).

2.2 SOURCE OF PARTICULATE MATTER

The sources of particulate matter can be anthropogenic (man made) or of natural origin. Figure 2.1 shows the detailed break down of global PM_{10} sources (Colls, 1997). On a global scale, over 80 per cent of particle production comes from natural origin (Colls, 1997). 60 percent of natural sources are primary that include suspension of soil dust, salt particles from sea spray, forest fires and volcanic action. Otherwise, 40 percent is secondary natural particles (Colls, 1997) that include sulphates, nitrates and hydrocarbons, which are produced by direct, catalytic, and photochemical oxidation by sulphur, nitrogen and volatile hydrocarbons (Godish, 1997). Another 20 percent of global particle production comes from anthropogenic sources (primary and secondary), which have profound effects on the atmosphere in urban and heavily inhabited areas (Singh, 1997). Anthropogenic or manmade particles are released into the atmosphere from human activities such as industrial emissions, traffic emissions, power production and agriculture and household activities (Van der Wal & Janssen, 2000).

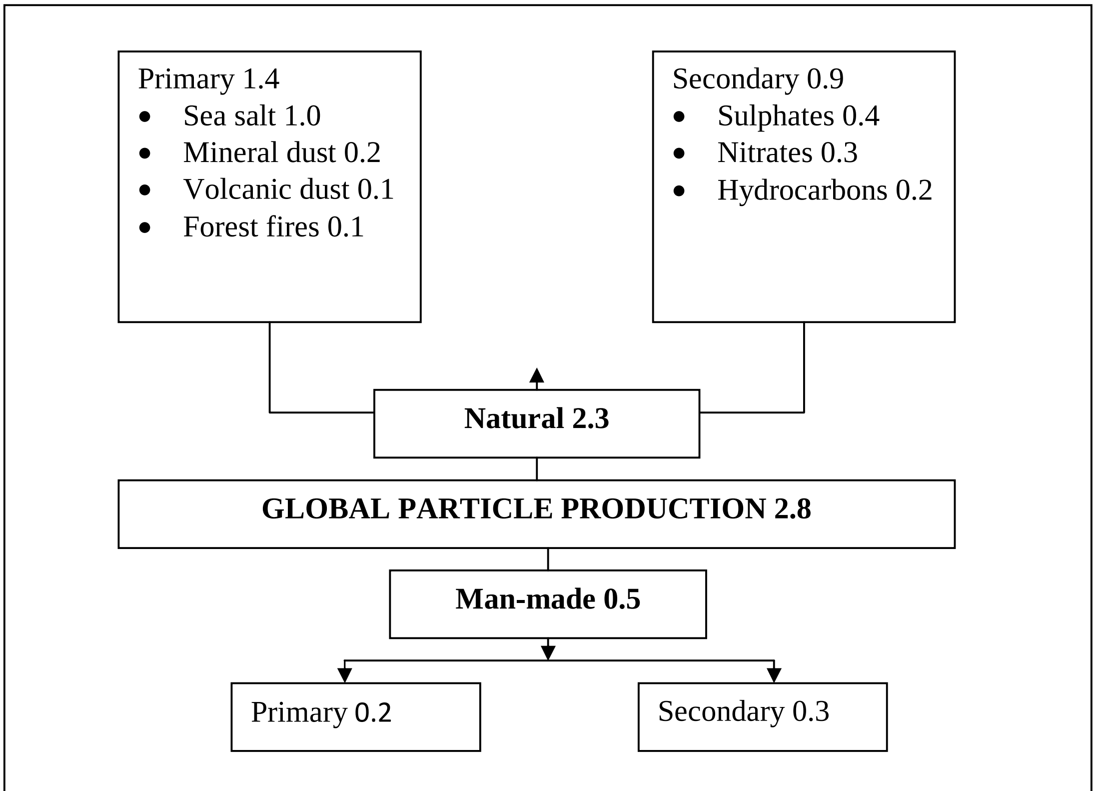

Figure 2.1 Global Particle Production (x 10^9 tonne per annum) (Colls, 1997)

2.3 PHYSICAL CHARACTERISTICS

The most important physical characteristics of particulate matter are size distribution, mass concentration and shape. Aerodynamic diameter is commonly used for particle sizing; it is the diameter of a standard density (1000kg/m^3 or 1g/cm^3) sphere having the same gravitational settling velocity as the particle being measured (Baron and Willeke, 2001). Otherwise, the particulate matter is measured in unit of mass concentration, which is a measure of mass particle in unit volume of gas and is expressed in g/m^3 or, since the amount of particle mass is generally very low, in mg/m^3 or µg/m^3. Particle size and concentration are intensely affected by many factors such as the source of the particle's formation and meteorological conditions (Baron and Willeke, 2001).

PM_{10} refers to particulate matter smaller than 10µm in aerodynamic diameter, and usually comprises the majority of particle mass. Within the PM_{10} size range, particles of less than 2.5 µm are described as fine, while the 2.5 to 10µm fraction is termed coarse (QUARG, 1996). Size distribution is important to human health as it determines the deposition of particles in different regions of the respiratory system. PM_{10} passes the thoracic fraction (respiratory system beyond the larynx) (Quarg, 1996) and penetrates the trachea and bronchial regions of the lung, distributing mainly at pulmonary bifurcations (Cormier *et al.,* 2006). The respirable fraction, $PM_{2.5}$ and ultrafine particles, $PM_{0.1}$ enters nonciliated alveolar regions and deposit deep within the lungs (Cormier *et al.,* 2006).

The size distribution of atmospheric particles, together with chemical composition, provides important information on the sources and processes of particles for assessing their health and climatic impacts (Gao *et al.,* 2009). Studies by Gao *et al.,* (2009) on diurnal variations of particulate matter in the Yangtze River delta in China found different diurnal patterns for particles in different size ranges. Large size particles (50nm – 1000nm) show two obvious peaks; one in the morning and the other in the late afternoon. It shows good correlation with vehicle exhausts as an important source of particles in the large size range. Conversely, smaller particles (10nm – 50nm) show a distinct peak at noon, suggesting that the particles come from light vehicles and due to particle formation events (formation of nucleation mode particles) and subsequent growth.

A particle may be extremely complex in shape, such as agglomerates (Willeke and Baron, 2001). *Agglomerate* is the term used to describe *a group of particles held* *together by van der Waals forces or surface tension*. Particles may also exist as *aggregate* where *a heterogeneous particle in which the various components are not easily broken up.* The term heterogeneous indicates that the individual components may differ from each other in size, shape and chemical composition. Particles may also be *flocculates*, which describes *a group of particles very loosely held together, usually by electrostatic forces, and can easily be broken up by shear forces within the air*.

The physical form of PM_{10} may be assessed by using scanning electron microscopy (SEM). Figure 2.2 shows the SEM image of a fly ash particle that is covered with soot particles. A straight fibre approximately 30 µm long and 1 µm wide is also part of this composite (Jones *et al.,* 2006).

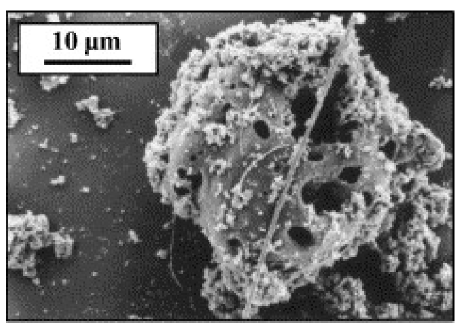

Figure 2.2 SEM image of fly ash particle (Jones *et al.,* 2006)

The effects of a particle's shape with its deposition in a neutrally stratified atmosphere were investigated by Vesovic *et al.*, (2001). Two simple mathematical models were used to show the influence of particle shape and deposition, the fugitive dust model (FDM) and particle trajectory model (PTM). Their results proved that the shape of the particle is a very important parameter in determining the deposition curves. However, as the particle size decreases, the differences in shape become less important in influencing the particle deposition.

In terms of size, smaller particles are more dangerous in relation to health effects due to their long residence time in the atmosphere (Marcazzan *et al.*, 2001). Particles of size less than 10 µm can easily penetrate through the nasal and head airways to reach the lungs. Otherwise, particles more than 10 µm can be removed by hairs at the front of the nose (Peavy, 1985). Therefore, particles that should be emphasized in any research are less than 10 µm in size because they can be inhaled by human beings and deposited in the human lungs. de Nevers (2000) stated that particles that cause significant air pollution problems are generally in the size range 0.01 to 10 µm, much smaller than the finest sand or the diameter of human hair.

2.4 CHEMICAL CHARACTERISTICS

There are various chemical compositions of particulate matter in the atmosphere. The composition of an individual particle depends on its source or origin and its subsequent atmospheric history. Because of the large variety of sources, atmospheric formation of secondary particles and atmospheric behavior, particles may contain hundreds of different chemical species (Godish, 1997). Therefore, by knowing the chemical composition of particulate matter, its sources and potential effects can be determined.

Airborne particles comprise three main categories of compounds that are insoluble minerals or crustal material, hygroscopic inorganic salts and carbonaceous material

(Watt & Hamilton, 2003). Background particulate concentrations are strongly dependent on the local geology and soil type, distance from the sea, amount and type of vegetation cover, season and weather conditions.

In urban areas particulate composition are more influenced by emissions from industrial processes and combustion. Carbonaceous compounds, nitrate (NO_3^-), ammonia (NH_4^+) and sulphate (SO_4^-) are the major components of PM_{10} in urban areas (Sun *et al.*, 2004; Putaud *et al.*, 2004). All the major components are hazardous to human health. In fact, black carbon (BC) contain in particulate matter can contribute to global warming by absorbing the solar radiation and re-radiating the sun's energy as infrared radiation that is trapped by the earth's atmosphere (Mishchenko *et al.*, 2004). Apart from that, geological species such as Si, Ca, Al, Fe, K and Mg were also found in urban samples of particulate matter believed to originate from dust uplift (Wu *et al.*, 2003).

In rural or background sites, secondary aerosols that are sulphate is the major component in PM_{10} resulting from fossil fuel and biomass combustion (Putaud *et al.*, 2004). Carbonaceous compounds were found lower at background location (Wu *et al.*, 2003). Sulphate potentially increases the acidity in the atmosphere, then form acid rain (Fellenberg, 2000). Sulphate that are produced from SO_2 gas can also affect the climate by reflecting solar radiation out into space and typically cause cooling (Mishchenko *et al.*, 2004).

A study in Eastern Spain had also been conducted by Alastuey *et al.*, (2004) on monitoring of atmospheric particulate matter around sources of secondary inorganic aerosol. From their study, the highest proportion of mineral dust components during non-African dust events were mainly within the coarse fraction (2.5 – 10μm) of PM_{10} (~ 80% of Fe, 79% of Al_2O_3, 76% of Mg, 76% of Ti and Sr, 70% of Ca). Marine components are also mainly found in the coarse fraction of PM_{10} (~ 77% of Na and 80% of Cl) compared to $PM_{2.5}$. Besides that, during the summer period, nitrates occur mainly in the coarse fraction of PM_{10} (82% of NO_3^-).

In conclusion, chemical composition of particulate matter mainly depends on the emission source. Research by Ibrahim *et al.*, (2005) within the vicinity of a rice processing found that rice husk ash contributed to high Si content in particles being sampled. In addition, soil dust and windblown rock-derived material in the atmosphere was high in one of the sampling sites due to earth work at a construction site that increased the earth crust materials.

2.5 WEATHER INFLUENCE

Meteorology plays a very important role in the formation, presence, behavior and prevalence of PM_{10} (Varadarajan, 2004). In the initial dispersion process from point or area sources, pollutants are released into the ambient environment where their transport and subsequent dilution depend on local meteorological phenomena and the influence of topography (Godish, 1997). The local dispersion processes were influenced by meteorological factors such as wind direction, wind speed and temperature.

2.5.1 Wind Direction

Wind is the movement of air. The movement originates in unequal distribution of atmospheric temperature and pressure over the earth's surface and is significantly influenced by the earth rotation (Peavy, 1985). PM_{10} dispersion is significantly affected by variability in wind direction. If wind direction is relatively constant, the same area will be continuously exposed to high pollutant levels. If, on the other hand, the wind direction is constantly shifting, pollutants will be dispersed over a large area and concentrations over any given exposed area are lower (Godish, 1997). The effect of wind direction is to find out the direction of transport of released pollutants (Nejad and Ramli, 2005).

A country with a tropical climate (Malaysia) experienced uniform temperature and continuous high humidity. Seasons in this country are distinguished according to the changes of wind flow patterns and rainfall. As it is located near the equator, the wind over the country is generally light and variable. However, there are some uniform periodic changes in the wind flow patterns that describe the four seasons experienced by the country, namely, north east monsoon (November to March), transitional period (April to May), south west monsoon (June to September), and another transitional period (October to November) (Meteorological Department, 2008). PM_{10} are found to reach the maximum concentration during the dry season in Seberang Perai, Penang (Ramli *et al.,* 2008). The wind from the south west carried the pollutants emitted from Indonesia caused by biomass burning for agricultural purposes, thereby increase the PM_{10} concentration.

Sun *et al.,* (2004) found that wind direction is one of the factors that cause high PM_{10} concentration in one of their study area in Beijing, China. The north west wind in winter brings the industrial pollutants from steel industries at Capital Steel Company to a residential area (Yi Hai Garden), making it the most polluted site of the three sites in winter.

2.5.2 Wind Speed

Horizontal winds play a significant role in the transport and dilution of pollutants. As the wind speed increases, the volume of air moving by a source in a given period of time also increases (Godish, 1997).

If the emission rate of air pollutants is relatively constant, a doubling of the wind speed will halve the pollutant concentration, as the concentration are found to have inverse relationship to the wind speed (Lu and Fang, 2002; Nejad and Ramli, 2005). Study conducted by Nejad and Ramli (2005) on urban air quality monitoring using GIS as a management system found that the resulting pollutant concentration is inversely proportional to the wind speed. There is a simple relation between the frequency distribution of wind speed and frequency distribution of air pollutant concentration (Lu and Fang, 2002). The distribution of PM_{10} and $PM_{2.5}$ can be successfully estimated from the distribution of wind speed through simple relationship. The concentration of air pollutant, C, at cumulative probability, p, is inversely proportional to the wind speeds, u, at probability of (100-p) when the distributional types and shape factors of both data are the same. They have shown the relationship as $K = C_p u_{(100-p)}$, where k is constant.