CHAPTER II
LITERATURES REVIEW

2.1 Particulate matter air pollution

2.1.1 Characteristics of PM-10 material

Particulate matter air pollution, by definition, is comprised of solid or liquid forms of various chemicals, varying in chemical and physical properties such as size, shape, density and composition, surrounded by air molecules (13). The size fraction of particulates usually is expressed in micrometer, μm, in terms of the aerodynamic diameter (d_{ae}), defined as the diameter of a sphere of a unit density ($1g/cm^3$) having the same settling velocity in air (14). PM-10 refers to Particulate Matter in which 50% of the particles have an aerodynamic diameter less than 10 μm.

The size distribution of ambient particle is commonly presented as consisting of two modes, a coarse mode with d_{ae} more than 1.3 μm and a fine mode with d_{ae} less than 3.5 μm. These two modes of particles have different origins and may overlap in the region between 1-3 μm (13). The coarse mode particles are generally derived from natural sources such as sea salt, and by industrial processes such as grinding and scraping. These particles in the large size fraction are mainly composed of oxides of crustal elements (e.g. silicon, manganese, aluminum etc.) from wind-blown dust, sea salt spray and plant materials (15-16). In addition, the coarse particles typically are typically not reactive and are stable. The fine mode particles usually originate from anthropogenic processes and can be divided into two sub modes. These are the nucleation mode (d_{ae} less than 0.1 μm), directly emitted from fuel combustion or condensation of gas after emission, and the accumulation mode (d_{ae} between 0.08-2 μm), particles of which are formed by either chemical reaction, gas to particle conversions or coagulation (17). The major components of fine mode particles are sulfate, nitrate, ammonium, elemental carbon, organic carbon, trace elements and water (18). The particles of the two modes differ considerably from each other in many aspects such as originating sources, size, chemical composition and the lifetime

suspended in the air. The coarse particles have a limited lifetime, and may settle down in minutes to a few hours whereas fine particles may stay suspended longer resulting in a longer travel distance (16).

2.1.2 Ambient air quality standards and guidelines

Generally, the most common referenced guidelines or standard for ambient air quality are those of the World Health Organization (WHO), the European Union (EU) and the United State Environmental Protection Agency (US.EPA). Most guidelines have been established based on health considerations with arbitrary margin of safety, technical feasibility and cost of attainment. Although, toxicokinetics and linkages to health outcomes of particulate matter are not clearly understood, WHO and some scientists suggest it should be no safe threshold level for particulate matter (19 - 21).

The 24-hour and annual average of PM-10 of the US.EPA primary and secondary standards are regulated with 150 and 50 $\mu g/m^3$ respectively while the WHO guidelines for Europe are 125 and 50 $\mu g/m^3$ respectively (22). However, the EU decided to limit PM-10 in 2005 with 50 $\mu g/m^3$ for the 24-hour standard and with 30 $\mu g/m^3$ for the annual mean. At present, the Thailand National Ambient Air Quality Standards (NAAQS) for 24-hour average PM-10 and annual average (arithmetic mean) PM-10 are 120 and 50 $\mu g/m^3$ respectively (23).

2.1.3 Ambient PM-10 concentrations

Air pollution by suspended particulate matter in Bangkok has been ranked in 1990 to be "the most serious threat to public health" (24). To monitor air quality in Bangkok, the Pollution Control Department (PCD) has set up 10 ambient air monitoring stations and 7 monitoring stations near the roadside with an additional 21 temporary roadside stations (23). Daily measurements of PM-10 have been performed by PCD since 1992. Even though the levels of PM-10 in the general areas of Bangkok have been declining significantly overall since 1997, the average PM-10 concentrations measured at temporary roadside stations in the city have not changed significantly.

Some measurements are still high, exceeding the 24 hrs limit value of 120 $\mu g/m^3$. The percentages of PM-10 readings that exceeded the 24 hour standard limit value in 1998, 1999 and 2000 are 6.38, 8.0 and 12.8 respectively. Moreover, for most of the roadside stations, the annual average of PM-10 concentrations exceeded the NAAQS (23).

2.2 Particulate matter and health effects

Particulate air pollution has been receiving much more attention since the finding was brought forth that health problems are significantly increased where ambient particulate concentrations are high. Several published epidemiologic studies of air pollution have evaluated the association between particulate matter and health outcomes for both acute (mortality, hospital admission, cardiovascular disease, respiratory symptoms and lung function) and chronic exposure studies (mortality, lung cancer, respiratory disease and lung function) (2, 18, 25, 26). The major route for particle to enter human body is the respiratory system. The factors affecting on particle deposition in the respiratory tract are size, shape, density and also the breathing pattern of each person. Particulates with aerodynamic size less than 10 μm can penetrate deeply into the respiratory system. In addition, health effects depend not only on the amount of particles inhaled but also their chemical properties and the sensitivity of the individual (22).

A number of epidemiological studies of particulate exposure have concluded that there are relationships between daily mortality or daily morbidity and ambient particulate air pollution (25 - 28). An association between short term changes in daily PM-10 concentration and daily mortality has been found in the areas with a daily average of PM-10 that is less than air quality standard in the USA, 150 $\mu g/m^3$ (25, 29). An increase in emergency room visits by of people aged fewer than 65 was also found where the daily PM-10 concentrations were less than 70% of the U.S. standard (3). In addition, results of analyses of the relationship between PM-10 long term exposure and health effects in a number of studies have yielded an association between long term exposure of particulates at low concentration (PM concentration = 30-35 $\mu g/m^3$) and respiratory symptoms (19) as well as an excess mortality rate at annual PM-10

concentrations of approximately 18 μg/ m^3 (4). Even though the relationships between the physical/chemical properties and toxicity of the particles and the mechanisms of how they are related to health effects are not yet clearly understood, it seems to reasonable to conclude that increased PM-10 levels may be one of the risk factors of premature death, cardiovascular disease and respiratory symptoms (5). Recently, summarizing the studies of PM-10 and health effects, Ostro (1994) has proposed that a 10 $\mu g/m^3$ increase in average annual PM-10 concentrations would result in an increase in the daily mortality rate of approximately 0.5 to 1.5% (20).

2.3 Exposure measurement

The term "exposure" is defined as an event that occurs when a person, with an emphasis on the human being who is the most important receptor in the given situation, comes into contact with a pollutant (6, 30). This definition makes exposure totally different from concentration and dose. Concentrations of pollutants may present without exposure unless pollutants have a way to come into contact with the human being. A dose, on the other hand, refers to the amount of pollutant that has crossed a physical boundary such as an epithelial cell in nose, lung or alveolar (6). In fact, for total exposure assessment, all of the exposure routes and all of the pollutants from all sources that have come into contact with a human body, have to be measured; e.g. soil, water food and air (3).

People normally move from place to place in a day. Therefore, they would be exposed to particulates at different levels because air pollution levels vary spatially and temporally (32). Thus, the integrated personal exposure should be determined by integrating the concentration over time when both the concentration and the duration of exposure are taken into account (7). The integrated personal exposure can be determined by either direct or indirect approaches. The direct approaches are measurements of the biological dose or personal sampling whereas the indirect approach can make use of ambient stationary or microenvironment measurements or models (6, 7).

2.3.1 Direct approach

2.3.1.1 Personal sampling

The main route of PM exposure is by inhalation. Therefore, in order to assess total PM-10 exposure, the concentrations of PM-10 in air entering the nose are measured by personal exposure monitors (PEMs). PEMs are worn by study participants and collect sampling air near the breathing area where there is no effect from exhaled air. Generally, PEMs for PM-10 are measurement devices that use identical techniques as those used for ambient PM measurement. Most of the PEMs are filter-based with different cut-offs and size fractions, for example PM-2.5, PM-3.5, PM-10 or multistage samplers. The other measurement technique is based on the principle of light scattering which is sensitive for PM with a d_{ae} >300 nm (8). For example, a personal nephelometer measures light scatter to obtain real time measurements of PM (33, 34). Even though human exposure data collected from direct measurements are more precise than the data from either ambient or microenvironment measurements used as an exposure surrogate, the second approach is often used because the personal exposure measurement data method is more expensive and requires more time and effort (7).

2.3.2 Indirect Approach

2.3.2.1 Microenvironment measurements

Microenvironment has been defined as "a chunk of air space with homogenous pollutant concentrations" (35). Microenvironment can be either indoor or outdoor locations or in vehicles, for example, a bedroom, a kitchen, a park or a car. Thus, under condition where the indoor PM-10 concentrations of all rooms in a building are homogenous, they could be described as one single microenvironment. Conversely, if PM-10 levels in the rooms are different or are not homogenous distributed, several microenvironment measurements are required to be more accurately descriptive. Microenvironment was also introduced as a volume in space, during a certain time interval, during which the variance of concentration in the volume is significantly less than the variance between that microenvironment and its surrounding microenvironments (32). In the microenvironmental model approach, integrated

exposure for an individual can be calculated as the time weighted average of the concentrations in microenvironments that the individual visited for a time interval (35, 36). Thus, the concentrations of PM-10 in each selected microenvironment including information about time fraction spent in each microenvironment are required in order to determine the personal exposure in this context. Time-activities diaries and questionnaires are important tools for quantification of the time fraction spent in each microenvironment (8). Although, the microenvironmental approach is much more demanding and expensive in order to take all microenvironments into account, especially in large-scale population studies, but it is more reliable than the use of ambient air measurements for estimation of exposure.

2.3.2.2 Ambient measurements

Recently, a number of air pollution epidemiological studies have reported using exposure data are obtained from ambient air measurements conducted on at a fixed site station. Of necessity, the population in the study area is assumed to have been exposed to the same pollution concentration. The exposure data of most of the studies exposure data are based on one central monitor. In cases where the pollutant is homogenously distributed, the monitoring site is uncritical. However, for most pollutants the concentration levels vary spatially. Thus, the average concentration from several monitoring sites or ambient monitoring networks has been determined in order to provide more representative exposure data. However, their validity and reliability remain to be determined by comparison with personal exposure measurement (8).

2.4 Personal, indoor and outdoor PM-10 measurement studies

2.4.1. Relationships between personal and indoor/outdoor PM-10 concentrations

In general, there are three types of study design that can be used to evaluate personal exposure to PM-10 as well as the relationship between personal PM concentration and ambient PM concentration: 1) longitudinal, in which each study participant is measured repeatedly for multiple days; 2) pooled (or lumping all participants), in which each participant is measured for one or two days with different

days for different subject; and 3) daily-average, in which many participants are measured on the same day (18). Longitudinal studies, treating each participant separately, show a higher correlation of personal exposure with simultaneously measured outdoor air concentration than do those studied by cross-sectional (lumping) analysis (37).

In the Particle Total Exposure Assessment Methodology (PTEAM study), PM-10 personal exposure was assessed for two consecutive 12-hour periods for 178 non-smoking participants living in Riverside, California, USA. The average indoor and outdoor concentration was 95 and 86 $\mu g/m^3$ in the daytime and nighttime respectively. In addition, the average personal PM-10 concentration during daytime (150 $\mu g/m^3$) was higher than that obtained during nighttime (77 $\mu g/m^3$). The cross-sectional correlation coefficient between personal and fixed site outdoor during daytime, 0.37, was lower than the analogous correlation coefficient during nighttime, 0.54 (9, 38).

From the Total Human Environmental Exposure Study (THEES) that conducted 24-hour averaged PM-10 personal exposure measurements for 14 consecutive days on 14 residents of Phillipsburg, New Jersey, USA, the average personal PM-10 exposure and indoor and outdoor concentrations were 86, 54 and 60 $\mu g/m^3$, respectively. The median of longitudinal correlation coefficients between personal and outdoor PM-10, was 0.68. The median R^2 of the 14 individual longitudinal regressions was 0.46 whereas that for cross-sectional (14 daily) regressions was 0.06 (39).

The investigation by Janssen et al., conducted 24-hour averaged PM-10 personal exposure for 4-8 days on 45 children (age 10 -12) in Amsterdam, the Netherlands (40). The mean personal exposures and ambient concentration were 105 and 38.5 $\mu g/m^3$ respectively. The median individual correlation coefficients between personal and outdoor concentration was 0.63. Janssen et al. also studied PM-10 exposure in 37 non-smoking adults (age 50 - 70) in Amsterdam. Personal, indoor and outdoor PM concentrations at central site were measured for 24 hr concurrently and repeatedly for 7 to 9 days. The mean 24 hour personal PM-10 concentration, 61.7 $\mu g/m^3$,

was higher than those for indoor, 35.0 $\mu g/m^3$ and for outdoor, 41.5 $\mu g/m^3$. the median of individual person-outdoor correlations was 0.50 (range from -0.41 – 0.92) while the median of cross-sectional correlations, simulated by randomly sampling one measurement per subject for use in calculation correlation and repeating the procedure 1000 times, was 0.34 (11).

In a Toronto, Canada study by Pellizzari et al. (1999), personal, indoor and outdoor and fixed site monitoring station measurement were conducted for 3 days. The mean 3-day concentrations were 67.9 $\mu g/m^3$ for personal, 29.8 $\mu g/m^3$ for indoor, and 24.3 $\mu g/m^3$ for outdoor PM-10 measurements. However, no correlations were reported (41).

Bahadori (1998) has investigated personal, indoor and outdoor PM-10 in 10 COPD (Chronic Obstructive Pulmonary Disease) patients whose activities are restricted (42). All personal, indoor and outdoor PM concentrations were monitored for two consecutive 12 hr periods concurrently and repeatedly for 6 days. The mean personal PM-10 concentration, 33 $\mu g/m^3$, was similar to that for outdoor concentration but higher than the mean indoor concentration, 22 $\mu g/m^3$, during the daytime. The results from the study showed that the personal cloud of these patients was less than that among other groups. In addition, there was considerably poorer correlation between personal and outdoor PM-10.

In another study done by Rojas-Bracho et al. (2000), 18 COPD patients were followed for up to 1-3 seasons (6 -18 days each). PM-10 personal, indoor and outdoor concentrations were measured for 12 hour periods during the daytime. Mean personal, indoor and outdoor PM-10 concentrations were 37.2, 31.9 and 22.2 $\mu g/m^3$ respectively. The median longitudinal correlation of person-outdoor concentrations was 0.35 (43).

From the study of indoor/outdoor PM-10 and PM-2.5 in Bangkok by Tsai et al. (2000), it was found that ambient air concentrations are generally correlated with indoor concentrations and potentially could be used to estimate personal exposure for residents living with no indoor sources of particulates. Indoor microenvironments in the

shop houses, including the shops, bedrooms and the living rooms have higher PM-10 concentrations than the ambient levels (44).

From a review by Wallace (37), a number of studies have shown a small cross-sectional correlation between PM-10 ambient concentrations and personal exposure with Pearson's R values ranging from –0.08 to 0.62, whereas, the individual correlations are larger with R ranging from 0.26 to 0.68. The cross-sectional relations have a disadvantage resulting from interpersonal variations. Therefore the correlations are always smaller than individual longitudinal correlation.

2.4.2 Factors affecting PM-10 exposure concentrations

Most urban areas encounter particulate air pollution. The major contributing source of particles in ambient urban air is road traffic (12). Some studies have found that PM-10 is higher at a main street or traffic influence site (45, 46). In addition, in several studies on the influence of outdoor air to indoor PM levels, the contribution of the outdoor source was estimated at about 30-70% depending on household characteristics and the existing indoor sources (38, 47). Smoking is found to be a significant factor affecting both personal exposure and indoor PM-10 concentrations (9, 11, 38). Cooking and cleaning also contribute to the difference between personal and outdoor concentrations (38, 43). In addition, burning joss sticks also significantly influences the to PM-10 exposure levels of the worshipper and indoor PM concentrations (48). These results indicate that exposure to PM-10 air pollution depends on geographic location, climate activity patterns and also building construction and individual human activities.

2.5 Source identification techniques

Different types of PM from various sources contribute to personal PM-10 exposure, which possibly results in different health outcomes. Of necessity, sources and component of personal PM should be estimated. A source apportionment technique or receptor modeling, in which pollutants measured at a receptor site are apportioned to their corresponding sources, has been used to identify and quantify the contributions of

various PM-10 sources to personal PM exposure. Several types of receptor models, for example chemical mass balance, multivariate and microscopic approaches have been applied in a number of PM source apportionment studies. Chemical mass balance (CMB), a widely used receptor model, requires either unique tracer or chemical composition profiles of all potential PM emission sources. Therefore, in case source profiles are not available or the number of sources is not known or identified, their source contributions are excluded. However, the approach can be conducted on individual samples.

Multivariate receptor models are based on analysis the correlation between concentrations of chemical constituents assuming that highly correlated components come from the same sources. Many multivariate methods are based on factor analysis and have been employed to identify and apportion source contributions of air pollutants in many studies (49 - 52). Principal components analysis (PCA) is one of the most common factor analysis methods which have as its purpose is to reduce the number of dimensions in a multivariate data set while preserving most of variance in the original data. The original variable set is transformed into a new set of variables, or factors. The variables can be grouped by their correlations. That is all variables within a particular group are highly correlated among themselves but show relatively small correlations with variables in a different group. Basically, the main assumptions of PCA are: 1) the chemical composition of the emission sources is constant, 2) the chemical constituents are stable and do not react with each other and their concentrations are linear addition, 3) the variation in concentrations of samples is dominated by the changes in sources contributions, 4) the effect of processes that affect all sources equally (e.g. atmospheric dispersion) is smaller than the effect of processes that influence individual sources (e.g. wind direction), 5) there are more samples than source types, 6) the measurement errors are random and uncorrelated, and 7) the eigenvector rotations are physically meaningful (53). The process of PCA begins from original data to create the correlation matrix. For instance, PM exposure data for n measurements of chemical component i species can be displayed as a data matrix and then is transformed to a correlation matrix for each pair of chemical concentrations. The

correlation matrix is decomposed into the metrics of eigenvalue and the corresponding eigenvectors to extract components or factors. A factor loading matrix is also then created from decomposition of the correlation matrix. Factor loading or correlation coefficient between variables and factors describes the relationship between each variable and each factor based on the same eigenvalue. The factors are simply formed by linear combinations of the corresponding original variables. Thus, the first principal component explains the highest percent of the original variance. The significant factors are those having an eigenvalue greater than 1 and they are orthogonal to each other. Therefore, each sample collected at the receptors represents a linear combination of these factors (50, 53). There are two forms of equation to express the relationship between factors and variables as show below.

$$Z_i = L_{i1}F_1 + L_{i2}F_2 + \ldots\ldots\ldots\ldots + L_{ip}F_p \qquad 2.1$$

$$F_p = W_{p1}Z_1 + W_{p2}Z_2 + \ldots\ldots\ldots + W_{pi}Z_i \qquad 2.2$$

Where

Z_i = Standardized variable: Z_1, ... Z_i

L_{ip} = Factor loading of factor p for standardized variable i

F_p = Common factors: F_1, ..., F_p

W_{pi} = Score coefficient of standardized variable i for factor p

Supposedly, each factor may be interpreted to be a single source of PM, thus PCA can be applied even though the source profiles are not available or the number of sources is not identified. However, PCA can determine the possible sources of PM but not the relative contributions of the sources. Consequently, factor analysis in combination with multiple linear regression analysis (FA/MR) is used to identify and quantify sources and their contributions. The technique using factor analysis to identify unique tracer variables (factors) and then these tracer variables are used as predictors in a multiple linear regression model having total PM concentrations as the response variable.

From the study by Thurston and Spengler in 1985, a principal component analysis with multiple linear regression analysis was employed to identify and quantify the major sources contributing to inhalable particles (IP) concentrations measured at a monitoring sire in metropolitan Boston, MA. The result indicated that automotive, oil and refuse incineration emission contributed roughly 25% of IP mass while soil and sea salt aerosol accounted for nearly 30%. The other 30% was ascribed to the coal combustion and the unexplained source was approximately 15%.

In another study done by Okamoto et al., 1990 reported that application of a varimax rotated factor analysis to a data set of total suspended particulate matter (TSP) measured in the Tokyo metropolitan area, Japan, five source types which were refuse incineration, soil and automobile, secondary particles, sea salt and steel mill, were identified. Then the contributions for each source type which were quantified by a multiple regression analysis were comparable with those of which were estimated by CMB model.

Lucarelli et al., 2004 reported the results of the analysis of PM-10 particulate samples, measured at three different site in Florence, Italy, Four main sources (traffic, oil combustion, soil-dust and wind transported sea salt) were identified by PCA. Traffic was determined as the major source for both the high traffic site and in the urban background site.