

Chapter 2

Research Background

2.1 Environmental Impact

2.1.1 Carbonaceous Particulate Matter

It has been widely realized that PM affects the local/regional air quality, human health and climate change significantly and is of worldwide interest. The exposure to ambient PM caused about 0.8 million death per year (WHO 2002a, b). In addition, the light extinction of PM could have obvious impacts on the ecosystem such as the yield of crop (Chameides et al. 2009; Russell et al. 1999). There are many ways to characterize the physic-chemical properties of PM, such as surface area, size, light extinction efficiency, chemical compositions of inorganic and organics, and toxic effects. One widely reported physical property is the PM aerodynamic size. Particles with different diameters have different transportation abilities and distinct health impacts (Ansmann and Müller 2005; Dockery et al. 1993; Englert 2004; Pope et al. 2009; Veselovskii et al. 2004). Fine PM may undergo longer air transport in environment and penetrate deeper into the end of bronchus and alveolus area, causing serious impacts on human health. Because of relatively large surface area of fine PM, there are many toxic organics bound in the fine PM, and hence increased the health risks of fine PM. In our daily lives, several commonly used terms include Total Suspended Particle (TSP, PM with diameter $<100\text{ }\mu\text{m}$), PM_{10} (PM with diameter $<10\text{ }\mu\text{m}$), $\text{PM}_{2.5}$ (PM with diameter $<2.5\text{ }\mu\text{m}$), $\text{PM}_{1.0}$ (PM with diameter $<1.0\text{ }\mu\text{m}$), and ultra fine $\text{PM}_{0.1}$ (PM with diameter $<0.1\text{ }\mu\text{m}$). Though a large number of researches had highlighted the high toxic potentials of fine PM, some studies also arose the attention on the toxic of coarse PM (Russell and Brunekreef 2009; Pope et al. 2009). The size distribution of PM can be separated into three modes, including Aitken nuclei mode ($D_p < 0.05\text{ }\mu\text{m}$), accumulated mode ($0.05\text{--}2\text{ }\mu\text{m}$) and coarse particle mode ($D_p > 2\text{ }\mu\text{m}$) (Tang et al. 2006; Whitby 2007). Aitken nuclei mode can be found in some new formed particles. PM in accumulated mode can be either coagulated from the Aitken nuclei mode, or directly produced from the condensation of vapors during the combustion process. PM in the emissions from the solid fuel

combustion is mainly in accumulated mode (Bond et al. 2004; Chen et al. 2005; Rogge et al. 1996; Reid et al. 2005). In general, secondarily formed aerosols are mainly in Aitken nuclei and accumulated modes. The size distribution of PM in the air is affected by a number of factors including ambient temperature and relatively humidity (RH) (McMurry and Stolzenburg 1989; Shi et al. 2007; Tan et al. 2009; Vasconcelos et al. 1994). In most cases, the PM size distribution in air follows the log transformed normal distribution (Tang et al. 2006; Hinds 1999).

In most developing countries, the air quality has been deteriorating and there are many programs studying the cause and potential impacts of severe air pollution (Balakrishnan et al. 2002, 2004; Cao et al. 2003a, b; Chan and Yao 2008; Kim Oanh and Zhang 2004; Kim Oanh et al. 2006; Pandey et al. 2006, 2008), however, current knowledge about the air pollution and its environmental impact is still lack, particularly when compared with the long history studies in developed countries (Ansmann and Müller 2005; Cercasov and Wulfmeyer 2008; Chow et al. 2008, 2011; Dockery et al. 1993; Englert 2004; Keeler 2004; Laden et al. 2000; Pope et al. 2009; Veselovskii et al. 2004). Because of the distinct sources and meteorological conditions, the emissions, ambient pollution levels as well as the potential impacts of PM have obviously temporal and spatial variations. In China, the fast economical development and large consumption of energy lead to severe air pollution during the last several decades (Cao et al. 2003a, b; Chan and Yao 2008; Davis and Guo 2000; Deng et al. 2008; Feng et al. 2006; Liu et al. 2008a, b; Lin et al. 2010; Remer et al. 2008; Sun et al. 2004; Wu et al. 2006; Zhao et al. 2009). The migration of population from the rural area to urban/sub-urban areas, particularly after the 1980s, leads to high population densities and serious environmental issues in the large cities. In China, a total number of city is about 660, among which about 170 ones are megacities with population over 1 million (Chan and Yao 2008). The urban population increased from 19.6 to 40.5 % during 1980–2005, and there would be about 350 million people living in the urban by 2025 (Zhu et al. 2011). Most of available studies are mainly in large cities, like Beijing, Shanghai and cities in the Pearl River Delta (Chan and Yao 2008; Hao et al. 2005; Zhang et al. 2009a). For example, it was reported that the main air pollutant is PM_{10} , and there were about 40 % days annually with 24 h average PM_{10} concentrations exceed the national standard even though the annual average concentration showed a decreasing trend from 1999 to 2005. The emission of PM in Beijing decreased from 2.70 million tones in 1994 to 58 thousand tones in 2005, but the industrial dust increased from 59 thousand tones to 100 thousand tones. Besides PM_{10} , $PM_{2.5}$ is of wide and increasing interest, especially in the last several years (van Donkelaar et al. 2010; WHO 2011; Yuan et al. 2012). According to the plan from the Ministry of Environmental Protection, some cities and pilot regions would start to monitor in daily air pollutant monitoring, and by 2015, all county cities should monitor and report $PM_{2.5}$ (Yuan et al. 2012). Since $PM_{2.5}$ is not only emitted from the primary sources but also formed during the secondary transformation, increased emissions of some precursors, for example NO_x and volatile organic compounds from vehicle emissions, would deteriorate the air quality and increase the technical and political difficulty in air pollution

control. It is expected that with the efforts of effective source control strategies and the implication of pollution control techniques, the emission and pollution level of PM would decrease in future. Even so, PM would still be the main air pollutant in China, and the severe air pollution levels should be arisen high attention by both scientists and policy makers (Chan and Yao 2008; Lei et al. 2011; Lin et al. 2010). One important and large source of PM in China is the consumption of traditional solid fuels including coals, wood and crop residues. In rural area, a large amount of solid fuels is used by residents for daily cooking and heating. The inefficient combustions of these fuels often cause serious air pollution in the region. It was reported that the PM₁₀ concentration in the kitchen during the cooking time might be 6 times of that during the non-cooking period, and the personal exposure level was three times for the cooker to that for the non-cooker (Jiang and Bell 2008). In addition to the use in residential household, biomass is also widely burned in open field, especially after the harvest. It was estimated that about 110–158 Tg biomass fuels were burned annually in open field (Streets et al. 2003; Cao et al. 2006).

Carbon fraction, mainly including organic carbon (OC) and elemental carbon (EC), is an important chemical composition of PM. Sometimes, the term “black carbon (BC)” is used. In general, the climate modelers prefer to use BC which often refers to the carbon or substance having light absorbing ability, whereas most people in the field of aerosol chemistry use the term EC. The analytical methods of EC and BC are usually different. For the former, thermal or thermal-optical methods are used in which the carbon was oxidized to CO₂ and quantified. The most widely used instruments to measure EC are EC/OC analyzer from Sunset, or DRI. BC is measured using the optical method at a specific spectrum, like 840 nm. Because the EC and BC data often correlated with one another and in many cases are comparable, they are sometimes considered as the same mass when the other measurement was unavailable (Bond et al. 2004; Chen et al. 2005; Chow et al. 2011). The sampling and analytical methods have significant influences on the obtained OC and EC results. However, current work on how to eliminate the influence of sampling and analytical methods on the analyzed carbon results is limited. It is expected that there would be more studies on the issues like how to estimate and control the absorption of gaseous organics, the vaporization of volatile organics in PM, and how to correct the difference or bias in different analytical temperature protocols and charring correction methods (Cheng et al. 2009a, b; Duan et al. 2007; Mader et al. 2001).

OC can be emitted from the primary combustion, and/or formed from the atmospheric reactions secondarily. Different from OC, EC is produced from the primary combustion sources such as the industrial processes, vehicle emission and coal combustions. Because of the enhanced secondary formation of OC, the ratio of OC to EC is usually higher in summer in comparison with that in winter (Chen et al. 2002; Yu et al. 2004). The ratio of OC and EC can be used to estimate the relatively contribution of secondary formation of OC in atmosphere (Tang et al. 2006; Tan et al. 2009; Turpin and Huntzicker 1995; Na et al. 2004). OC and EC, particularly the later, usually prefer to be present in fine PM (Chen et al. 1997; Tang et al. 2006; Tan et al. 2009; Venkataraman and Friedlander 1994;

Venkataraman et al. 1994), and absorb (for OC) or adsorb (for EC) organics causing the toxic of PM (Crutzen and Andreae 1990; Seinfeld and Pandis 2006; Allen et al. 1996; Venkataraman et al. 2002). OC and EC also influence the local/regional climate change through the light scattering and absorbing, respectively, leading to negative and positive radiative forcing, respectively (Yu et al. 2004; Chow et al. 2011). The net climate effect of carbon fraction in PM is often controlled by the ratio of OC to EC (Zhi et al. 2009). The positive radiative forcing of EC highly depends on the mixing status (inter- or external mixture). It was reported that the forcing of EC may range from 0.27 to 0.78 W/m² (Jacobson 2001). During the last three decades, the average temperature in the Arctic increased by about 1.5 °C, of which about 1.1 °C might be contributed by the warm effect of aerosol because of the light absorption of BC (IPCC 2001; Shindell and Faluvegi 2009). The emissions from open biomass burning may increase the ambient concentration in the Arctic by about 2 times (Koch and Hansen 2005; Stohl et al. 2006; Warneke et al. 2009, 2010). Since the PM in background air in the Arctic mainly consist sulfate and much small percents of nitrate, organics and BC (Law and Stohl 2007; Quinn et al. 2006), while PM from the biomass burning generally has relatively higher fractions of OC and EC but less sulfate (Warneke et al. 2009), the transport of PM from biomass burning to the Arctic may have much significantly impacts on the ambient concentrations of OC and EC in the Arctic (Warneke et al. 2010). The mass percentages of total carbon (the sum of OC and EC) vary in different sources (Chow et al. 2011; Duan et al. 2007; Seinfeld and Pandis 2006). For example, the mass percents of OC and EC in PM from residential wood combustion were reported to be around 50 and 12 %, and in the emission from residential coal combustion at 70 and 26 %, but in the industrial coal combustion, they might vary from 2–34 to 1–8 % (Chow 1995; Chow et al. 2011; Duan et al. 2007).

In addition to the impacts on air quality and climate change, OC and EC have significant impacts on human health since they often absorb and/or adsorb many toxics and can penetrate deeper into the lung area and bronchus (Bond et al. 2004; EPA 2012; McCracken et al. 2010; Zhi et al. 2009). The monitoring of particle carbon fraction has been included into some research programs and observation campaign from the 1980s (Qin et al. 2001). These programs provided important data on the concentration levels and dynamic change patterns of BC. In China, the air pollution of BC is of growing interest, especially after the large increase of vehicles and consumption of coals both of which are large emitters of BC. During the 1980s, there are some, though very limited observation of BC, and after the 1990s, more and more field BC observation studies come out. In general, we have very limited studies on the source, ambient level and impact of BC in China, and there are often large biases and uncertainties in the inventories and environmental impacts of BC (Gu et al. 2006; Zhang et al. 2009b; Zhi et al. 2009). For example, it was once reported that the flood in the south China and drought in the north China may be strongly related to the emissions of BC from residential biomass and coal combustion (Menon et al. 2002), but in the another study, the results were found to

be hard to be predicted when including the emissions of BC from residential solid fuel combustion into the atmospheric model (Gu et al. 2006; Zhang and Tao 2009).

2.1.2 Polycyclic Aromatic Hydrocarbons and Derivatives

Polycyclic aromatic hydrocarbons (PAHs) are a group of hydrocarbon organic pollutants with two or more benzene rings (Kanaly and Harayama 2000; Mumtaz et al. 1996; Neff 1979). They have been ubiquitously found in air, soil, water and organism. Because of the carcinogenicity and mutagenic toxicities, PAHs have been widely studied in many areas around the world (Fernandes et al. 1997; Gaspari et al. 2003; IARC 1987; Perera et al. 2002; Pufulete et al. 2004; Xue and Warshawsky 2004; Zhang et al. 2009c). There are many PAH isomers in environment, and they are usually a mixture in environment. The physical and chemical properties of PAH isomers differ obviously. PAHs with the same rings and molecular weight often have the comparable physic-chemical properties. In general, with the increase of aromatic rings and molecular weight, the solubility, vapor pressure and volatility decrease, but the hydrophobic effect increases (Smith and Harrison 1996). PAHs are included in the priority list in many countries and organizations. In the middle 1970s, 129 compounds are listed by U.S. EPA as priority control pollutants, among which there are 16 PAHs. PAHs are also the Regionally-based Assessment of Persistent Toxic Substances in the United Nations Environment Program, 2002. Because of the ability of volatility and long range transport (Becker et al. 2006; Halsall et al. 2001; Kaneyasu and Takada 2004; Lang et al. 2007; MacDonald et al. 2000; Mackay 2001; Primbs et al. 2007; Prevedouros et al. 2004; Sehili and Lammel 2007; Tao et al. 2006; Van Jaarsveld et al. 1997), PAHs are also included into the Convention on Long-range Transboundary Air Pollution by the United Nations Economic Commission for Europe, 1998. In China, some PAHs including naphthalene, fluoranthene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, benzo[ghi]pyrene, and benzo[ghi]perylene, are included into the priority control pollutant list (Zhou et al. 1990). Benzo[a]pyrene, as an example of toxic PAHs, is included in the environmental air quality standard (MEP 1996).

Pollution level and characterization of PAHs pollution in environment have obviously temporal and spatial variations due to distinct emission sources and the influence of meteorological conditions (Garban et al. 2002; Hafner et al. 2005; Liu et al. 2007; Liu 2008; Wang 2010; Zhang 2010). Once emitted into the air, PAHs often redistribute between the gaseous and particulate phases (Bidleman 1988; Goss and Schwarzenbach 1998; Lohmann and Lammel 2004; Pankow 1987), and undergo the complicated chemical reactions and physical processes like dry and wet deposition (Behymer and Hites 1988; Reisen and Arey 2002, 2005; Garban et al. 2002; Wang et al. 2010, 2011a, b). Due to the long range transport ability, PAHs can redistribute in the areas like some background sites and the Arctic area (Daly and Wania 2005; Ding et al. 2007; Garban et al. 2002; MacDonald et al.

2000; Van Jaarsveld et al. 1997; Wania and Mackay 1993; Wang et al. 2010). PAHs in air can enter into the soil and water through the deposition and washout process, and PAHs in soil could also be reemitted into the air or water (Cousins et al. 1999; Meijer et al. 2003; Wang et al. 2011a; Wild and Jones 1995). As a result, the PAH content in soil varies in site and time, which is heavily associated with the emission densities, physico-chemical properties of PAHs and structure and properties of soil (Heywood et al. 2006; Wilcke and Amelung 2000).

PAHs exposure would cause serious adverse human health outcomes through direct (inhalation or dietary exposure) or indirect exposure (the absorption of animals and plants) pathways (An et al. 2005; Duan and Wei 2002; Duan et al. 2008; Li et al. 2005, 2009a, b; Menzie et al. 1992; McClean et al. 2004, 2007; Tao et al. 2006; Wu et al. 2010; Xia et al. 2010). The relative contributions of inhalation and dietary exposures may vary in site and time. In the regions with serious ambient PAHs pollution, the inhalation exposure would contribute largely to the total exposure (Duan et al. 2008; Li et al. 2009a, b; Tao et al. 2006; Xia et al. 2010). For example, it was reported that for the non-smokers, the inhalation exposure made up to 90 % of total PAHs exposure (Suzuki and Yoshinaga 2007; Vyskocil et al. 2000, 2004). Meanwhile, in some regions where the air levels of PAHs are very low, personal exposed PAHs may be mainly from the ingestion of high molecular weight PAHs in food (Van Rooij et al. 1994; Vyskocil et al. 1997). PAHs would form carcinogenic PAHs-DNA adducts in vivo after exposure (Boysen and Hecht 2003; Cheng 1998; Jacob and Seidel 2002; Denissenko et al. 1996; Ramesh et al. 2004). The formation of DNA adduct is an important process of PAHs-induced toxicity. The risk of many diseases, like lung cancer after PAHs exposure has been reported in many epidemiologic studies (Karlehagen et al. 1992; Romundstad et al. 2000; Simioli et al. 2004; Vyskocil et al. 2004). It was reported that after the adjusting of gene susceptibility and spatial distributions of ambient PAHs and population, about 1.6 % of lung cancer cases (0.65×10^{-5}) could be attributed to the exposure to ambient PAHs in China (Zhang et al. 2009c).

Based on the information of source, pollution level and dynamic changes of PAHs, pollution control strategy on PAHs could be conducted effectively (Halsall et al. 1994; Buehler and Hites 2002; Wild and Jones 1995; Venier and Hites 2010). The pollution of PAHs may be expected to decrease after the effective implication of the regulations and laws, which actually occurs in many developed countries (Coleman et al. 1997; Jacob et al. 1997; Katsoyiannis et al. 2011; Menichini et al. 1999). Due to the emission sources of industrial and vehicle emissions, the PAHs pollution levels in urban areas in these developed countries are generally higher than those in the rural areas (Allen et al. 1996; Cotham and Bidleman 1995; Gigliotti et al. 2005; Motelay-Massei et al. 2005; Smith and Harrison 1996). In China, most studies on PAHs so far are in urban area (Duan et al. 2006; Guo et al. 2003a, b; Wan et al. 2006; Wang and Kawamura 2005; Wang et al. 2006a, b, 2007a, b; Wu et al. 2005, 2006; Xie et al. 2008), and much higher pollution levels and adverse health outcomes in these densely population regions have been identified in comparison with those in developed countries (Wang et al. 2006a; Wu et al. 2006; Zhang and Tao 2009; Zhou et al. 2005). Because of large consumption

of solid fuels under ineffective residential combustions in rural area, and the deployment of central heating system (Guo et al. 2004; Liu et al. 2007; Liu 2008; Mumford et al. 1987, 1993; Wang et al. 2009a; Wu et al. 2006; Wang 2010; Wang et al. 2011a, b; Zhang et al. 2007a, 2008a, b; Zhang and Tao 2009), the pollution of PAHs in the urban area is more or less controlled but the pollution in rural area is still very serious leading to comparable, even slight higher pollution levels of PAHs in the rural China. There are some research programs, although limited due to technical difficulty and expensively financial cost, in China focusing on the pollution characterization of PAHs in the last several years. In an extensive field study in the Beijing-Tianjin region, it has been found that the ambient PAHs pollution in the area was much higher than that in the developed countries (Okuda et al. 2006; Wu et al. 2005, 2006; Zhou et al. 2005), and there was no significant difference between the rural and urban areas (Liu et al. 2007). Significant seasonal distribution was revealed with high contamination level in the winter, followed by autumn and relatively low levels in the summer and spring. The distribution of PAHs was strongly correlated with the factors like emission density, population and GDP (Liu et al. 2007; Liu et al. 2008a). The source apportionment result showed that the main sources of PAHs in the Beijing-Tianjin region were industrial process, crop residue burning, coal combustion and coke production.

Besides parent PAHs, PAHs derivatives, such as nitrated, oxygenated, hydroxyl-PAHs, are of growing concern recently. In comparison with parent PAHs, PAHs derivatives often have more direct and strong toxic (Allen et al. 1996, 1997; Miller 1978; Bolton et al. 2000; Cheeseman et al. 1985; Palmer and Paulson 1997; Walgraeve et al. 2010). Some derivatives, like oxygenated PAHs (oPAHs) including ketones and quinones, are important intermediary metabolites of many carcinogens (Bolton et al. 2000). The mechanism(s) of these derivatives is not very clear, but it is widely accepted that this was related to the formation of free radicals, and the produce of reactive oxygen species (ROS). PAHs derivatives can be emitted from the primary sources including the incomplete combustions of biomass and coal, and formed secondarily through the reaction of parent PAHs and free radicals (Nielson 1984; Arey et al. 1989; Atkinson et al. 1990; Albinet et al. 2007, 2008a, b; Walgraeve et al. 2010). Nitrated and oxygenated PAHs had been measured in the smoke from the emissions of vehicle, residential coal and biomass burning, and affected by the factors like combustion temperature, air supply condition and relatively humidity (Gullett et al. 2003; Sidhu et al. 2005; Fitzpatrick et al. 2007). The secondary formed PAHs highly vary in sampling site and period (Eiguren-Fernandez et al. 2008; Kojima et al. 2010; Walgraeve et al. 2010). Because of limited studies available, it is not very clear the relative contributions of primary and secondary sources nowadays. .

Ambient measurements on PAHs derivatives are limited, and mainly in developed countries and regions (Allen et al. 1997; Lundstedt et al. 2007; Reisen and Arey 2002, 2005; Tang et al. 2005; Wilson et al. 1995). Some limited studies in China included the emission characterization of nitrated PAHs (nPAHs) from the vehicle emission (Xu 1984) and in some environmental samples (Hattori et al. 2007; Tang et al. 2005). Based on the literature reported datas, oPAHs

concentrations are comparable with the parent PAHs within the same order of magnitude, while nitrated PAHs are generally 2–3 orders of magnitude lower than the level of parent PAHs. It was reported that some quinones are chemically steady and can stay longer in atmosphere after emission (Church and Pryor 1985; McFerrin et al. 2008; Pryor et al. 1983). Similar to the parent PAHs, PAHs derivatives can be either in gaseous or particulate phases, but because of relatively low vapor pressure, these derivatives have mode tendencies to be present in particulate phase, especially fine PM (Angelo et al. 1999; Youhei et al. 2004; Walgraeve et al. 2010).

2.2 Emission Inventory

2.2.1 *Inventory of CPM*

PM can be emitted from both natural sources like soil dust and volcano eruption, and anthropogenic sources. Fine PM can be also formed from the secondary chemical reactions of some precursors (Odum et al. 1996; Andreae and Rosenfeld 2008). It was estimated that globally the anthropogenic emission of PM was about 210 Tg/year, and secondarily formed aerosol was about 139 Tg/year (Andreae and Rosenfeld 2008). In Europe, annual emission of PM was about 1.83 Tg, of which 0.52 and 0.23 Tg were from the indoor burning and agricultural activities (Amann et al. 2005). The annual emission of PM in Asia was about 30 Tg, of which 17 Tg was emitted from the open biomass burning (Hays et al. 2005; Streets et al. 2003). In regard of the carbon fraction, based on the fuel consumption data from the International Energy Agency (IEA) and reported EFs in the literature, it was estimated that total emission of BC in 1996 was about 7.95 Tg, among which open biomass burning, residential biomass burning and coal combustion contributed 42, 18 and 6 %, respectively (Bond et al. 2004, 2007). The historical emissions of BC and OC from 1850 to 2000 were found to increase from 1.0 to 4.4, and from 4.1 to 8.7 Tg, respectively (Bond et al. 2007). By updating the data for biomass burning, the estimated emission of BC in 1996 in the second estimation was lower than the previously estimated 7.95 Tg (Bond et al. 2007; Fernandes et al. 2007). In the latest report on BC, global total emission of BC in 2007 was about 7.6 Tg, among which open biomass burning, residential combustion, transportation and industrial sources contributed 35.5, 25.1, 19.0 and 19.3 %, respectively (EPA 2012). It was thought that because of increased use of fossil fuels, the emission of BC increased during the last several decades (Junker and Liousse 2008; Novakov et al. 2000), however, due to improved techniques in combustion and control technologies, the emission of BC has decreased after the 1980s (Junker and Liousse 2008). There are very large uncertainties in these inventories. Even though different methods reported similar temporal and spatial distributions, the estimated amounts varied dramatically because of the distinct data of EFs and fuel consumption.

China is one large emitter of BC (Cooke et al. 1999; Bond et al. 2004). It was reported that anthropogenic emissions of BC and OC in Asia were 2.54 and 10.4 Tg, respectively, among which 1.05 and 3.4 Tg were from China (Streets et al. 2003). It is obvious that fuel type and burning condition in China differed dramatically from those in other countries and regions, and hence the lack of information of direct measurements on the emission characterization of the specific fuel-stove combinations in China would lead to considerable bias in the developed inventories. Based on the literature reported EFs of BC or PM, the emission of BC in China was estimated at 1.34 Tg, and about 605.4, 512, 82.5, and 36.9 Gg from the burning of residential coal, biomass, industrial coal and diesel, respectively (Streets et al. 2001). It was also predicted that with the development of approaches like new and clean techniques and the deployment of briquettes, the emission of BC would decrease to 1.22 Tg by 2020, and the residential biomass and coal combustions were still the two dominated sources contributing 535 and 387 Gg, respectively (Streets et al. 2001). Recently, Lei et al. (2011) estimated the emissions of BC and OC in China from 1995 to 2000. In the study, EFs of BC and OC were calculated from EF of PM and the mass fractions of EC and OC in PM, respectively. The national total emissions of PM_{2.5}, PM₁₀, TSP, BC and OC were 9.28, 13.5, 24.9, 1.13 and 2.87 Tg in 1990, and by 2005, they increased to 12.95, 18.83, 34.26, 1.51, and 3.19 Tg, respectively (Lei et al. 2011). The emission of PM increased from 1990 to 1996, and after 1996 there was a short decrease, and then increased again after 2000. The emissions of BC and OC increased after 2000. Residential solid fuel combustion is a dominated source of CPM in China. Based on the estimated consumption of biomass fuels and literature reported EFs, it was estimated that the total emissions of PM and BC increased from 2000 to 2007 at a rate of 2.6 %, and at 3.62 and 0.43 million tones in 2007. Due to limited domestic measurements, in most emission inventories EFs reported for other countries and cities are often adopted, which often bias the estimated amounts. In general, the emission of BC in China during 1995 to 2001 was 1.37–1.71 Tg (Streets et al. 2001; Bond et al. 2004; Cao et al. 2006; Streets, et al. 2003; Zhang and Smith 2007). Residential combustions of coal and biomass fuels are the dominated sources, and the open burning of biomass burning after the harvest also contributes largely to the total anthropogenic emission. It is realized that when there were still very large emissions from residential solid fuel combustion, with the increased emissions from the gasoline and diesel powered vehicles (Zhi et al. 2009; Wang et al. 2012), anthropogenic emission of BC would be an important air pollutant of concern and requires further studies.

2.2.2 Inventory of PAHs

PAHs are mainly produced from the incomplete combustion processes (Mastral and Calleán 2000; Tsubulsky et al. 2001), and are ubiquitous in environment after the emissions of both anthropogenic and natural sources. Generally, the sources of

PAHs include the industrial, transport, domestic burning, agricultural activity and natural emissions (Tsibulsky et al. 2001; Zhang 2010). Volcano eruption, forest fire, and the biological formation are all able to produce PAHs. Though it is different to estimate the activity level and the EFs of PAHs from the natural sources, it is widely accepted that anthropogenic emissions contribute largely to the total PAHs in environment. PAHs from the industrial process are highly related to the techniques and pollution control technologies, and PAHs from the transport sources are related to the type of vehicles, usage, loading and operation modes, as well as fuel types. Residential combustion sources of PAHs include the burnings of biomass fuels and coals. In most developing countries, these solid fuels are widely used for daily cooking and heating by rural resident. Because of relatively low burning efficiency, residential solid fuel combustions often yield high emissions of incomplete pollutants, and it is the main source of PAHs globally and particularly in developing countries.

There are some inventories on the emissions of PAHs at the country and regional levels (Berdowski et al. 1997; Galarneau et al. 2006; Pacyna et al. 2003; Tsibulsky et al. 2001; Wenborn et al. 1999; Zhang and Tao 2009). For instance, it had been estimated that global total emission of BaP from anthropogenic source during 1966–1969 was about 5 Gg/y (Suess 1976). Zhang and Tao (2009) developed a global emission inventory of PAHs in 2004, and the differences among countries and different sources were addressed. The results showed that global total emission of PAHs was 0.52 million tones in 2004, and there was 0.29 million tones from Asia. China, India and U.S. were three large emitters with the national total emissions of 114, 90 and 32 thousand tones, respectively. In terms of the source contribution, biomass burning contributed over 57 % of the total, and the emission from industrial source only made up to 10 % the global total. The source profiles varied among different countries. In general, vehicle emission is the main source in developed countries, and in most developing countries, residential solid fuel combustion was the largest emitter. In comparison with less work on global emission estimation, there are more inventories on the national or regional scales (Tsibulsky et al. 2001; Berdowski et al. 1997; Pacyna et al. 2003; Wenborn et al. 1999). For example, Tsibulsky et al. (2001) estimated the emissions of 6 PAHs in the Soviet Union during 1990 to 1997 based on the literature reported EFs. Wenborn et al. (1999) reported a total emission of 1.7 Gg/year from U.K. and most of PAHs were from the industrial activities while the residential combustion was a minor source. Berdowski et al. (1997) estimated the emissions of FLA, BaP, BbK, BkF, BghiP and IcdP from 23 European countries, and pointed out that the emissions from residential source would increase while that from the industrial source will decrease gradually. The historical emission of BaP from Europe was reported by Pacyna et al. (2003). According to the report by European Monitoring and Evaluation program, total emissions of BaP, BbF, BkF and IcdP in Europe were 2.4 and 1.3 Gg in 1990 and 2003, respectively. It was reported that the emissions of 16 PAHs and 7 carcinogenic PAHs from the U.S. in 1990 were 26.5 and 2.0 Gg, respectively, among which firewood combustion contributed about 33 % of the total (U.S. EPA 1998). Afterwards, the U.S. EPA

updated the emission inventory of 7 carcinogenic PAHs between 1970 and 1995, and more important, a database was established so as to share the results for the scientists all over the world (U.S. EPA 2001). The emission of BaP in the Great Lake region and the Ontario was 26.8 Mg/year, and manufacturing industry, indoor firewood burning and open biomass burning were three main sources contributing 33, 28 and 13 % of the total emission (GLC 2007). It was pointed out that with the effective control on vehicle emission, the implement of regulations and control strategies, and improved combustion efficiencies of indoor cooking stoves, regional PAHs emission decreased obviously.

Emission inventories of PAHs in developing countries are rarely developed so far. To our knowledge, in addition to the global inventory by Zhang and Tao (2009) in which PAHs emissions were estimated at a national scale, one widely used inventory in developing country is that developed by Tao group (Xu et al. 2006; Zhang et al. 2007b, 2008a) for PAHs emission in China. The total emissions were 25.3 Gg/year, and residential wood, crop residue and coal combustion contributed 34, 26 and 20 %, respectively. The contribution of coking production was 16 %. In comparison with the composition profile in other developed countries or regions, the fractions of high molecular weight PAHs most of which are toxic ones were generally higher in the emissions from China. This might be due to relatively high contributions of emissions from low efficient residential biomass and coal combustions. Xu et al. (2006) analyzed the temporal-spatial and historical distribution of PAHs in China. After that, Zhang et al. (2007b) developed a high resolution emission inventory of PAHs at a county level. The estimated emission was 28.8 Gg, and the contributions of biomass burning, residential coal combustion and coking were 59, 23 and 15 %, respectively. The composition profiles varied significantly among different counties. The study on historical change suggested that with the development of social economic and energy consumption, the emissions of PAHs increased simultaneously. From 1950 to 1979, the total PAHs emission increased from 19.0 to 67.0 thousand tones, and between 1950 to 1961, because of the fast increase of coal consumption in the residential sector and widespread indigenous coking, PAHs emissions increased by about 1.4 times. After the middle of 1970s, because of the urbanization and family planning policy, the natural increase rate of rural population decreased and subsequent residential consumption of biomass stopped the increase. During the period, indigenous coking was the main cause of PAHs increase in China. In 2003, the coal law was issued and implemented, and most indigenous coking was banned. After that, the PAHs emission decreased accordingly.

2.2.3 Uncertainty in Inventory

In almost all inventories developed so far, there are considerable biases and uncertainties (Bond et al. 2004; Streets et al. 2001; Zhang et al. 2009c). Emission inventory is developed based on the activity level and corresponding emission

factor (EF). The information of activity level can be obtained from the field survey or statistical reports. EFs defined as the pollutant mass for per fuel mass or energy (unit: mg/kg or mg/kJ) is the basic input for the development of emission inventory. The lack of data on either activity level or EFs would lead to large biases in developed inventories. Nowadays, the information on the fuel consumption of major stationary sources is available and accessible for most studies. However, the information for the minor or scattered pointed sources is lack since they are usually difficult to count out. In terms of the EFs, in addition to the lack of sufficient data, variations in reported data due to influencing factors such as fuel properties, burning conditions and even experimental methods are more responsible for the uncertainties in the developed inventories. EFs reported in the literature can vary in orders of magnitude (Streets et al. 2001). The influences of many factors are completed and usually complicated. For example, the pollutant emission would increase under high excess air and subsequent low burning temperature and thermal efficiency (Fan et al. 2010; Johansson et al. 2004). The burning of high moisture fuel would require an amount of heat to vaporize water and hence lead to incomplete combustion of fuel and emissions of incomplete burning pollutant, but during the combustion of low moisture fuel, the fast burning would cause an oxygen deficient atmosphere in the stove chamber which may also lead to increased emissions of incomplete pollutants (Rogge et al. 1998; Simoneit 2002). Generally, the EFs are lower under high burning efficiencies, and the later is affected by a number of factors such as fuel moisture, fuel loading and air supply. It was reported that from 1990 to 2005, because of the implement of pollution control devices and technology improvement, the EFs of PM_{2.5} and TSP from the combustion sources in China decreased obviously, by about 7–69 and 18–80 %, respectively (Lei et al. 2011). EFs can be measured directly from the field or laboratory experiments, or calculated from the other related pollutants (Bond et al. 2004; Zhang et al. 2000). For example, EFs of particle-bound PAHs can be estimated from the EFs of PM and PAHs mass fraction in PM. In a study on global carbonaceous carbon inventory, Bond et al. (2004) quantitatively analyzed a number of factors influencing the emissions and identified the most significant factor. For instance, in the coking production, three most significant factors are technology, EF of PM, and the carbon fraction in PM. In the coal combustion, EF of PM was the main influencing factor, followed by the technology and carbon fraction. For most sources, EF is the most important factor or of relatively high importance in comparison with technology level and energy consumption data (Bond et al. 2004). It has been previously reported that relative variations in PAHs emissions from the aluminum electrolysis with pre-baked anode, non-transportation petroleum, industrial coal and gasoline and kerosene consumption in transportation were 314, 228, 161, 173 and 179 %, respectively. The calculated variations for the EFs (16.1 %) were significantly higher than that of 5.41 % for the activity level based on the Monte Carlo simulation, which indicated that EFs were the main source of uncertainty in developed inventory (Zhang and Tao 2009).

2.3 Residential Solid Fuel Combustion

2.3.1 Solid Fuels

Solid fuels mainly include natural combustible materials like crop straw, firewood and coal. Because of easy access, high abundance and relatively low cost, solid fuels are widely used in rural households for daily cooking and heating. There are over 3.0 billion people all over the world using solid fuels in daily lives, and most of them live in developing countries. Biomass fuels are one most widely used solid fuel in rural area. The consumption of biomass contributed about 14 % of global total energy consumption, and in developing countries, it could make up to about 40 % (IEA 2011a, b). During the burning of biomass fuel, combustible fractions such as lignin, cellulose and hemi-cellulose, were burned and emitted vaporized organics some of which would participate into the formation of PM and other incomplete pollutants. The process can be separated into two phases of flaming and smoldering with or without obvious fire. Low efficient biomass burning often produces large amounts of incomplete particles and gaseous pollutants (Andreae 1991; Andreae and Merlet 2001; Simoneit 2002). In China, biomass fuel contributed about 24 % of national total energy consumption. The burnings of crop residues and wood materials are the major sources of pollutants like PM, volatile organic compounds and SO₂ (Cao et al. 2005, 2007; Crutzen and Andreae 1990; Tian et al. 2011).

Coal is the most abundant and widely used fossil fuel. The consumption of coal in China is very large, and it can be expected that in the years to come, coal would still be the main energy source in the country. The coal consumption increased from 603 Mtce in 1980 to 1389 Mtce by 1996, and after that although there was a short decrease, it raised up to 2656 Mtce in 2007. Coal consumption comprises up to 70–76 % of national total energy consumption (Chen and Xu 2010). Coal combustion process covers the phases of pre-heating, burning of volatile fractions and combustion of the other solid materials. Pollutant emissions from the first two phases were about 50 times of that in the last burning period (Bond et al. 2002; Butcher and Ellenbecker 1982). But it is noted that these three phases are not absolutely independent (Bond et al. 2002, 2004). It is accepted that at the initial burning period, a large number of pollutant was released and increased obviously, and then in the smoldering phase without obvious fire, pollutant emissions usually decreased.

The combustions of biomass and coal under relatively low burning efficiencies often produce various pollutants, and subsequently affect the indoor and outdoor air quality and human health. It was reported that the indoor PM_{2.5} concentration in the kitchen using briquette coal was 13 and 30 times of those using natural gas and liquid petroleum gas, respectively (Peng et al. 2005). Exposure to these toxic pollutants is associated with increased risks of various diseases (Chapman et al. 2005; Hu and Guo 2007; Tian et al. 2009; He et al. 2005; Lin et al. 2005; Mumford et al. 1987; Smith 1993; Zhang and Smith 2007). The occurrence of COPD in

population using solid fuels for daily cooking was 16–19 %, significantly higher than that of 8.7 % in the population using liquid petroleum gas (Wen et al. 2006). In China, exposure to smoke from residential solid fuel combustion caused about 0.42 million premature death, which was about 40 % higher than that of 0.30 million caused by the outdoor air pollution (Zhang and Smith 2007).

Residential solid fuel combustion is a major source for many pollutants like PM and PAHs, especially in developing countries. It is necessary to develop reliable inventories of these pollutants from solid fuel combustion so as to estimate the impacts on air quality, human health and also regional climate change of these pollutants of widespread concern.

2.3.2 EF Measurements

EFs can be affected by a number of factors and usually vary in orders of magnitude (Streets et al. 2001; Xu et al. 2006). Beside the use in the development of emission inventory, EFs are also widely used in the development and implement of many environment pollution control strategies. For instance, the emission standard is one most important term used to regulate the pollutant emission behaviors of most industrial sources. Emission permits, standards and control strategies based on the inadequate or even wrong EFs might ignore some important sources of target pollutants, and even result in wrong emission reduction plans (U.S. EPA 2006; Pouliot et al. 2012).

When the mass or energy of combusted fuel and the total mass of emitted pollutants are known, the EF can be calculated directly. This is one most widely used approach to measure the EFs, especially in laboratory chamber study. The fuel burning process differs significantly in field in comparison with that in laboratory chamber. Field measurement might be closer to the real practice and more reliable as an input for the study on emission and transport behaviors of these pollutants, however, it is realized that it is difficulty to collect all emitted pollutants in field. Alternatively, the calculation of EFs based on the carbon mass balance method is developed and commonly adopted in the measurement of EFs for fuel combustion. The method follows the assumption that carbon in fuel would be burned and released into the atmosphere as the forms of gaseous CO, CO₂, total hydrocarbon carbon (THC) and particle-bound carbon. The results calculated based on the carbon mass balance method were found to be comparable to the direct measured ones within 20 % (Dhammapala et al. 2006).

In most EF measurement experiment, sampling dilution system or cooling system is adopted to avoid the potential impacts of high temperature and relatively humidity (RH) in the emission smoke on sampling and EF measurements. In a dilution system, high purity inert gas or clean air is used to dilute the emission exhaust and cool the smoke temperature. In the system, the dilution ratio, dilution rate and temperature of diluted gas affect the mass and size distribution of PM significantly (Purvis et al. 2000; Lipsky and Robinson 2006). For example, it was

reported that when the dilution ratio increased from 20:1 to 350:1, the EF of PM_{2.5} decreased by about 50 %. The dilution also changes the gas-particle partitioning of volatile organics (Lipsky and Robinson 2006). In practice, it is necessary to test and control the dilution ratio and rate, and monitor the change of gas temperature and RH. The dilution system is often used in laboratory study (Hildemann et al. 1989; Hays et al. 2005; Schimidt et al. 2011; Schauer et al. 2001; Rogge et al. 1993a, b, c, d, 1994). The cooling system with a cooling device after the exhaust exit to cool the smoke temperature is another widely used sampling system (Bond et al. 2004), for example in the emission source measurement study by U.S. EPA. In addition, in some studies, especially field measurements, the sampling was done directly at the chimney exit (Roden et al. 2006, 2009). In this case, the exhaust is diluted naturally by ambient air and assumed that smoke temperature decreased significantly after the mixing with cold ambient air once emitted.

There has been a long research on the emission measurements of particle and gaseous pollutants in developed countries (Gupta et al. 1998; Jetter and Kariher 2009). For example, by using a dilution system (Hildemann et al. 1989), Schauer and Rogge and the colleagues investigated the EFs of PM and particle-bound pollutants for a variety of sources like residential wood combustion, vehicle emission and open biomass burning (Simoneit et al. 1993; Schauer et al. 1999a, b, 2001, 2002a, b; Rogge et al. 1991, 1993a, b, c, d, 1994, 1997a, b, 1998). They also analyzed the influence of fuel type and burning conditions on the EFs and composition profiles. Jenkins et al. (1993, 1996a, b) simulated open burning of biomass fuels in a laboratory tunnel, and measured EFs of PM and PAHs at 5.05–7.27 g/kg and 5.04–683 mg/kg, respectively, varying under different burning conditions. Dhammapala et al. (2006, 2007a, b) measured EFs of PM, OC, EC and organics from the burning of crop straw. They also compared the results from direct calculation and that using the carbon mass balance method, and found that the results were comparable (Dhammapala et al. 2006).

Although there are lots of tests on emissions from biomass burning in the literature for developing countries, the studies mainly focused on the open burning rather than the combustion in residential sector. In most developing countries and regions, like Asia and Africa, more attention should be paid to the residential solid fuel combustion since it produces largely to the total pollutant emission. Venkataraman and Kim Oanh and the colleagues measured EFs of PM, CO, VOCs and OC for a variety of solid fuels burned in different stoves that were often used in Asian countries like India, Thailand and Malaysia by using a hood and dilution system in laboratory (Bhattacharya et al. 2002; Kim Oanh et al. 1999, 2002; Venkataraman and Rao 2001; Venkataraman et al. 2002). Roden et al. (2009) conducted a series of measurements on PM emission from wood combustion in Honduras and found that the EF of PM in field measurement was 6.1 g/kg, about 4 times of that of 1.5 g/kg measured in laboratory study. For the same fuel and stove, the EF of PM was significantly different between the field (6.6 g/kg) and laboratory studies (1.8 g/kg).

In China, much less work has been done. Moreover, most studies so far are in the laboratory chamber. Zhang and colleagues measured EFs of CO, CO₂, NO₂,

SO₂, VOCs and PM for a variety of fuels like coal, crop residue, wood, kerosene and natural gas burned in different stove types (Edwards et al. 2004; Tsai et al. 2003; Zhang et al. 1999, 2000; Zhang and Smith 1999). The results for a large fuel/stove combination are one most widely used database for the development of emission inventory in China. The research group led by Dr. Hao designed a sampling system and measured the EFs of CPM and gases for residential biomass burning in rural China (Li et al. 2007a, b, 2009a, b; Wang et al. 2009b). Chen and colleagues reported the EFs of PM, OC and EC for a variety of coals with different maturity (Chen 2004; Chen et al. 2004, 2005, 2006, 2009; Zhi et al. 2008, 2009). In their study, a cooling system was developed to avoid the influence of high temperature on sampling. They found that PM emission varied significantly among different coal types. The emission for anthracite was about 6–15 times lower than that for the bituminous. PM emitted from residential coal combustion was fine with high abundance of sub-micron particle. Among these limited measurement studies, PAHs emission was much less (Dou et al. 2007; Li et al. 2008; Zhang et al. 2008c, 2011), and because of different methods used, variety in fuel and stove types, these reported results varied dramatically. Lu et al. (2009) compared EFs of PAHs from the rice straw burning under different fuel moistures, burning temperatures and air supply amounts. The results showed that the EFs of PAHs increased with the increase of combustion temperature ranging from 200 to 700 °C. With the increase of air supply, the EFs of PAHs increased and decreased with a maximum at air supply of 40 %. It is thought that adequate air supply is necessary to achieve low pollutant emission. For PAHs emissions from wood combustion, Zhang et al. (2003) reported PAHs EFs for four wood fuels burned in different conditions, but the study was done in Austria using local fuel and local purchased stove. PAHs EFs for coal reported in the literature ranged from 6.0 to 882 mg/kg, depending on coal property, form and burning conditions (Cui et al. 1993; Chen et al. 2004, 2005, 2006; Dou et al. 2007; Liu et al. 2009). Emissions from the burning of anthracite with low volatile matter content are generally lower than those for sub-bituminous and bituminous coals. Li et al. (2003) measured PAHs in the emissions from the burning of four different coal types in a laboratory tunnel, and found that PAHs in the exhaust were about 30–60 times of that in the coal indicating the formation and emission of PAHs under high temperature pyrolysis and pyrosynthesis. Total EFs of 16 PAHs ranged from 0.010 to 0.225 mg/kg, or 0.001–0.009 mg/KJ. Dou et al. (2007) reported PAHs EFs for bituminous coals from Shanxi and Beijing were 882 and 880 mg/kg, and for anthracite were 104 and 71 mg/kg, respectively. The emission was lower for the briquette compared with the raw chunk, and the emissions from the coal stove with one chimney were about 4–12 times of that burned in two-chimney stove (Liu et al. 2009). Chen et al. (2005) measured EFs of 14 priority PAHs for five different coals. The EF for anthracite was about 3 orders of magnitude lower than that for the bituminous.

The main objective of this thesis is to measure emissions of CPM (EF_{CPM}) and PAHs (EF_{PAHs}) from the residential combustions of different solid fuels. Among various sources, we focused on the residential solid fuel combustion since it is one

major source of these pollutants in China, particularly in rural area, and there are limited data available so far. It is hoped that the study can provide important first-hand data for the future development of CPM and PAHs inventories in China, and also as a part of input in environmental and ecological analysis modeling, the results can provide helpful information in the analysis of the impacts of these pollutants on human health and climate change.

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