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AEROSOL POLLUTION IN SOME CHINESE CITIES

(IUPAC Technical Report)

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Abstract: Emissions caused by the use of coal and by traffic have caused serious photochemical smog and aerosol pollution with unique characteristics in most Chinese cities. This report gives an overview of aerosol concentrations in China based on data obtained from both the literature and recent research by the authors. The results show that TSP (total suspended particulate) and PM-10 (particles with aerodynamic diameter 10 μm) concentrations frequently exceed the National Ambient Air Quality Standard and that ambient aerosol concentrations constitute a serious air pollution problem. PM-2.5 concentrations are also high and account for 60 % of the PM-10 mass. Organic carbon and sulfate are the most abundant components of PM-2.5, while crustal elements represent a minor portion. Nitrate concentrations are almost the same as sulfate in summertime, which implies that NOx control is very important in lowering fine particle concentrations and in improving air visibility. The chemical mass balance (CMB) method was applied in Beijing to identify the sources of PM-2.5. The main sources include fugitive dust, coal burning/industrial processes, traffic emissions, and secondary aerosol produced by atmospheric chemical conversion.

1. INTRODUCTION

The well-documented [1–3] adverse effects of fine particles on human health have led to increased research on the sources and atmospheric behavior of these particles. Penetration into human lungs and its potential effects depends on the size of the particles, which provides a reason for sampling of particles as a function of size. The classical total suspended particulate samplers collect aerosol with an aerodynamic diameter of typically 100 μm or less. PM-10 samplers will collect particles with a diameter of less than 10 μm, which can *enter* the human air tract. Particles smaller than 2.5 μm (PM-2.5) will penetrate into human lungs.

Ambient aerosol is partly caused by direct emissions of particles into the atmosphere (primary aerosol), as well as by the oxidation of gaseous compounds such as sulfur dioxide, nitrogen oxides, and volatile organic compounds to nonvolatile products such as sulfates, nitrate, and low-volatile oxidized organic compounds.

Air pollution in China during the past decades has been typically caused by the use of coal, which is, by far, the most important fuel. SO_2 and particulate matter are responsible for serious damage to the environment. The air pollution arising from the use of coal burning has not been effectively controlled, and other sources of emissions have also become important, owing to the recent rapid economic development and urbanization. Fast growth of the number of vehicles in China, especially in mega-cities and economically developed regions, such as Beijing, Shanghai, Guangzhou, the Pearl River delta, and the Yangtze delta, has caused a sharp increase in concentrations of NOx, volatile organic compounds, particulate matter, and ozone [4–6]. Emissions due to the use of coal and traffic together lead to serious pollution, characterized by enhanced photochemical smog, high concentration of fine particles, and poor visibility. These pollution problems not only cause local problems, but also lead to the degradation of regional air quality. As a result, the traditional Chinese control policy that focuses on single cities is no longer effective to abate urban air pollution, and efforts to reduce emissions in one mega-city only cannot improve regional air quality of the surrounding areas.

Since the 1990s, a number of projects have been set up to study air pollution, to develop emission inventories, and to develop monitoring strategies in Beijing, Guangzhou, Qingdao, the Pearl River delta, and the Yangtze delta. This paper gives an overview of the results obtained in those areas with focus on aerosol pollution.

2. STATUS OF AEROSOL POLLUTION

2.1 Mass concentration of aerosol

China, like many other developing countries, is exposed to serious pollution due to high aerosol concentrations. Total suspended particulate (TSP) has been measured in China for many years, and the data have revealed that serious pollution problems are encountered in most of the country. Figure 1 presents TSP pollution in China in 1997 on the basis of annual mean concentrations from 84 monitoring stations. The annual average concentrations of TSP within the state-controlled network ranged from 32 to 741 μg/m^3, with a national average of 291 μg/m^3 [7]. TSP concentrations in 67 cities (72.0 % of the monitored cities) exceeded the Grade II (200 μg/m^3) air quality standards for cities of the China National Ambient Air Quality Standard (NAAQS, GB3095-1996) [8]. Compared with southern China, TSP pollution in northern China is more serious, although a few cities in the south also suffer from heavy TSP pollution. TSP represents an air quality problem especially in Beijing, Tianjin, Gansu, Xinjiang, and Shanxi. The high TSP concentrations are generally attributed to coal combustion and fugitive soil dust.

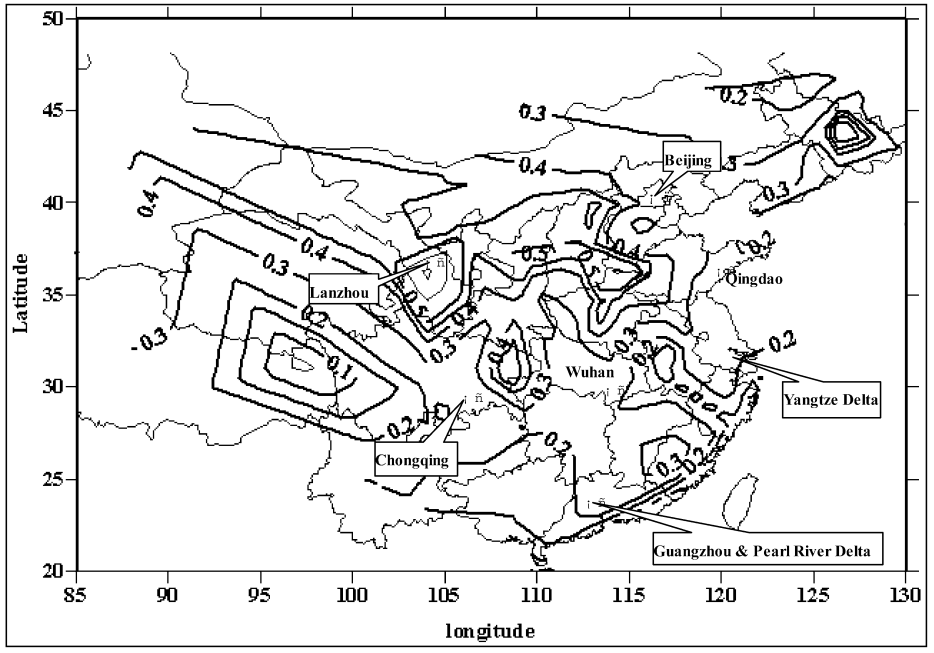


Fig. 1 Annual mean mass concentration of TSP in China in 1997 (μg/m^3).

The concentrations of PM-10 and PM-2.5 (particles with aerodynamic diameter less than 10 and 2.5 μm, respectively) are, however, seen as even more problematic because of their adverse effects on human health [1–3,9,10] and atmospheric visibility. Little research on PM-10 and PM-2.5 was conducted in China before the 1980s. Since the middle of the 1990s, fine particle pollution has been a con-

cern, and projects have been carried out to characterize the ambient PM-10 and PM-2.5 concentrations and their sources. The sampling sites were located mostly in urban areas of large cities, such as Beijing [11,12], Chongqing [13], Wuhan [13], Lanzhou [13], Guangzhou [13], Qingdao [14], etc. Annual mean concentrations in some cities and regions are shown in Fig. 2. The concentrations are averages of measurements over four seasons of a year (or years) with two-week sampling duration for each season. PM-10 annual mean mass concentrations in the cities range from about 70 to 220 $\mu g/m^3$, most of which cannot meet the Grade II (100 $\mu g/m^3$) of the NAAQS. The highest concentrations are observed in Guangzhou, Lanzhou, and Beijing. PM-2.5 annual mean concentrations vary from about 50 to 140 $\mu g/m^3$; the highest values are found in Guangzhou (in 1996) and lowest in Qingdao. All reported concentrations are all much higher than 40 $\mu g/m^3$ air quality standard for PM-10 in Europe and 15 $\mu g/m^3$, the recommended PM-2.5 standard in the United States. Regional measurements of PM-2.5 concentrations were conducted intensively during summer and winter campaigns in the Pearl River and Yangtze deltas. PM-2.5 concentrations can be as low as those in U.S. cities under conditions of good dispersion, as values measured in July of 2000 in the Pearl River delta show, but increased PM-2.5 concentrations are found under stagnant conditions (e.g., November 2000) owing to emissions from the large sources in the area. The concentrations of PM-10 and PM-2.5 shown in Fig. 2 are generally much higher than those measured in Hong Kong (55.3 $\mu g/m^3$ for PM-10) [15], Seoul (South Korea, 75 $\mu g/m^3$ for PM10) [16], Sapporo (Japan, 24.8 $\mu g/m^3$ for PM-10) [17], Birmingham (United Kingdom, 47 $\mu g/m^3$ for PM-10) [18], Coimbra (Portugal, 52.1 $\mu g/m^3$ for PM-10) [18], São Paulo (Brazil, 30.2 $\mu g/m^3$ for PM-2.5) [19], and Kangwha (background site for urban area in South Korea, 29.1 $\mu g/m^3$ for PM-2.5 observed from March 1996 to Dec. 1997) [20]. The aerosol concentrations in Wuhan and Chongqing are comparable to those in Kaohsiung (Taiwan, 111 $\mu g/m^3$ for PM-10 and 68 $\mu g/m^3$ for PM-2.5) [21].

The PM-2.5/PM-10 ratios (ratio of PM-2.5 mass concentration to PM-10 mass concentration) in China, ranging between 0.5–0.7 with an average 0.6 as shown by the line in Fig. 2, are about the same as those observed in Europe or the United States or slightly lower. It can be inferred that Chinese cities might suffer from serious PM-2.5 pollution, even though only limited data are available, when the high PM-10 concentrations which have been measured and the PM-2.5/PM-10 ratios in Fig. 2 are considered.

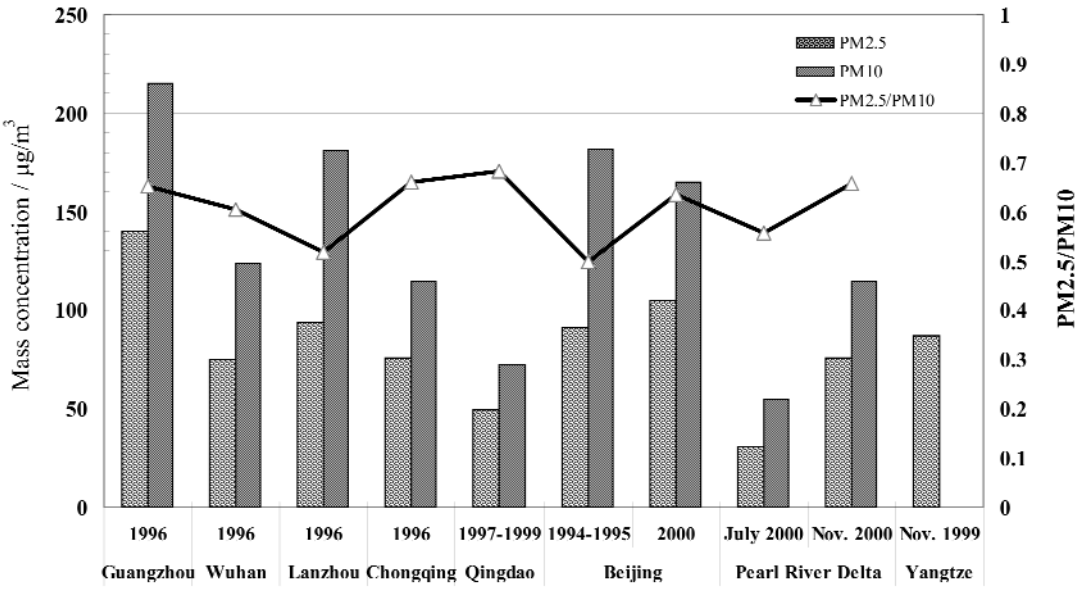


Fig. 2 PM-10 and PM-2.5 average mass concentrations in several cities of China.

2.2 Effect on atmospheric visibility

Probably the most noticeable effect of air pollution is reduction in atmospheric visibility. Pollutants (particles and some gases) scatter and absorb light in the air and cause poor visibility. Those particles, referred to as fine particles, are in general smaller than 2.5 μm in aerodynamic diameter.

The relation between PM-2.5 and visibility in China was studied in Beijing in June 1999 and January 2000 by TEOM (tapered element oscillating microbalance) and Nephelometer, respectively [22]. Figure 3 shows the expected anticorrelation between PM-2.5 and visibility based on hourly measurements. The visibility decreases rapidly with the increase in PM-2.5 concentration from 12 to 260 μg/m^3. The same PM-2.5 mass concentration leads to different visibility in summer and winter, indicating that visibility depends on other factors besides PM-2.5 [23,24]. Aerosol particles can grow by uptake of water vapor at higher relative humidity, which leads to decrease of visibility. This growth is a function, among other parameters, of the chemical composition of the particles.

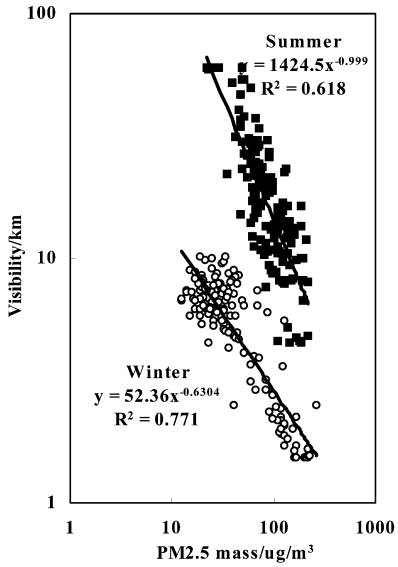
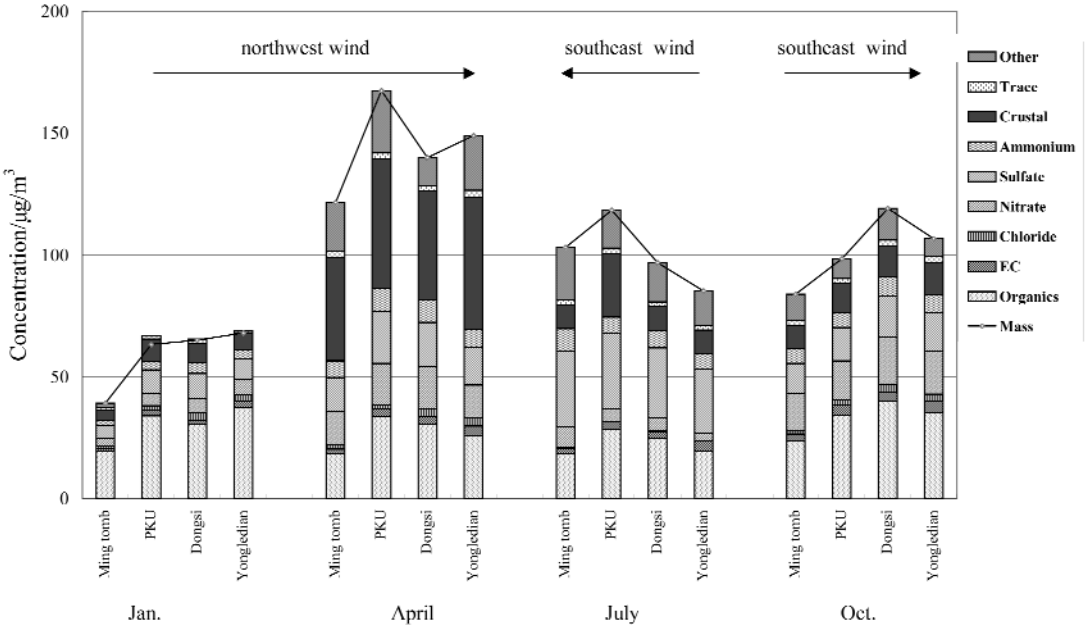


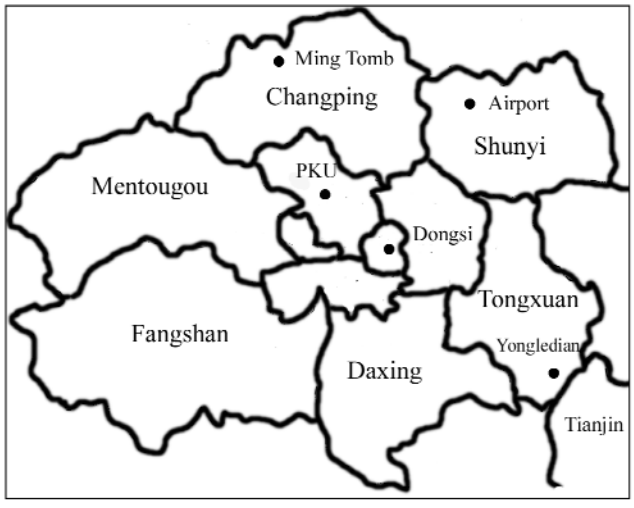
Fig. 3 Correlation between PM-2.5 and visibility (hourly mean).

2.3 Regional air pollution

Because of the fast economic and urban development, anthropogenic pollution has extended from the urban to the regional scale in some city-clusters of China. For example, the whole Beijing-Tianjin area and the Pearl River delta are sometimes covered with large polluted air masses leading to strongly reduced visibility and adverse effects on ecosystems and human health. Figure 4 presents the PM-2.5 concentration variation with wind direction in Beijing area and the Pearl River delta [25,26]. In the Beijing area, four ground-based aerosol sampling sites, i.e., Ming Tomb, Peking University (PKU), Dongsi, and Yongledian are positioned along the main wind directions, representing different areas in Beijing. PKU and Dongsi can be characterized as urban aerosol sampling sites. The Ming Tomb sampling site, about 40 km away from Beijing city in the northwest, is located at a historical scenic area with fewer anthropogenic emissions. Yongledian, approximately 60 km from the city in the southeast, is chosen to represent the rural area around Beijing. PM-2.5 concentration is lower in the upwind area of Beijing; the concentration increases significantly in the Beijing urban area (represented by the sampling sites of PKU and Dongsi) owing to large anthropogenic emissions; the PM-2.5 concentration is at about the same level or slightly lower in the air mass moving down wind of Beijing. A significant increase in

PM-2.5 concentration in downwind areas is observed, compared with upwind areas. The spatial variation of PM-2.5 concentrations at different sites scattered along different wind directions would give evidence that the effect of aerosol pollution in Beijing would not be limited to the city area but would extend to a larger area. Air quality in rural areas can occasionally become as affected as that in urban areas. The same is true in the Pearl River delta, according to the measurements. The concentrations of PM-10 and PM-2.5 gradually increased along the wind direction, which illustrates regional transport of aerosol (in Fig. 4, lower picture).





(a)

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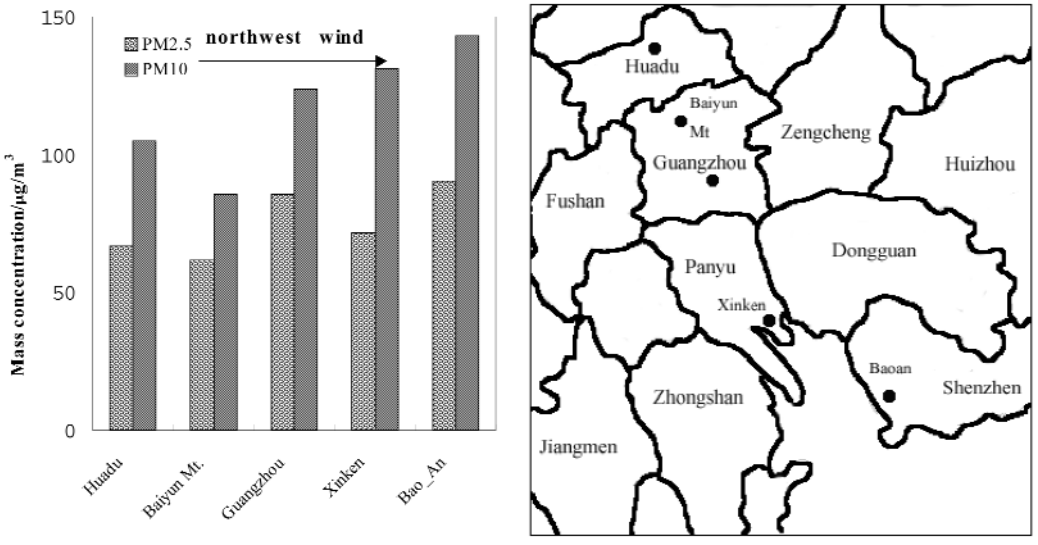

(b)

Fig. 4 PM-2.5 concentration variations along wind direction in Beijing (a) (2000) and in the Pearl River delta (b) (November 2000).

3. CHEMICAL COMPOSITION OF AEROSOL

3.1 Chemical composition of PM-2.5

As shown in Table 1, sulfate and organic carbon (OC) are the major components of PM-2.5. Sulfate concentrations range between 9.9 and 20.0 μg/m^3 with rather small spatial variations in most areas. Sulfate concentrations are relatively low in Beijing, 9.9 μg/m^3 in the year 2000, owing to large-scale measures for SO_2 emission reduction. The contribution of sulfate to PM-2.5 mass varies considerably in different cities and regions, ranging between 15–30 % of the PM-2.5 mass. In Qingdao, sulfate accounted for 32.3 % of PM-2.5 mass, which is almost twice that in other cities. This might be attributed to large emissions from both anthropogenic and natural (seawater) sources [27,28]. OC with concentrations in the range of 19–41 μg/m^3 accounts for 20–40 % of the PM-2.5 mass. The Yangtze delta has the highest OC concentrations and the highest fraction of OC in PM-2.5; see the data in Table 1.

Table 1 Main chemical compositions in several locations of China (μg/m^3).

Sites		Year	OC	EC	SO_4^{2-}	NO_3^-	NH_4^+	Mass
Beijing		1994–1995			15.2	5.2	6.5	91.0
Beijing [25]		2000	26.1	5.4	9.9	5.6	4.8	105.0
Pearl River delta [26]		Nov. 2000	19.2	1.7	14.4	2.3	2.4	84.3
Qingdao [14]		1997–1999			16.0	6.9	7.9	49.6
Yangtze delta [24]	Li-an	Nov. 1999	27.7	2.8	15.8	6.7	7.1	73.1
	Changsu	Nov. 1999	40.5	3.0	20.0	11.3	9.4	111.9
	Sheshan	Nov. 1999	33.2	2.0	16.2	9.3	6.6	83.8
Waliguan [29]		1994–1995		0.05–0.6				
Shangdianzi [30]		1999–2000		0.2–0.3				

Note: OC and EC are determined by thermal/optical carbon analysis (TOCA). Ions are determined by ion chromatography (IC).

EC concentrations are also high in urban areas if compared with values observed in more remote areas. The EC concentrations in PM-2.5 are in the range of 1.4–5.4 μg/m^3 with the highest value in Beijing. In Waliguan, a global background station in terms of air pollution in Qinghai, west China, EC concentration is only 0.05–0.6 μg/m^3 [29]. At Shangdianzi, a regional background station in Beijing, EC concentration is 0.2–0.3 μg/m^3 [30]. The spatial variation of EC concentrations reflects the geographical variation in emissions. The concentration of nitrate is relatively low with large variations. However, it must be pointed out that nitrate concentrations in Beijing, Qingdao and the Pearl River delta could be severely underestimated because of chemical interferences in filter measurements (see Slanina in this issue). Online measurements by the steam jet aerosol collector (SJAC) have shown that nitrate is present at almost the same level as sulfate in Beijing during the summer [31]. The filter pack method underestimates nitrate concentration by some 70 % compared with results obtained by SJAC; see Fig. 5.

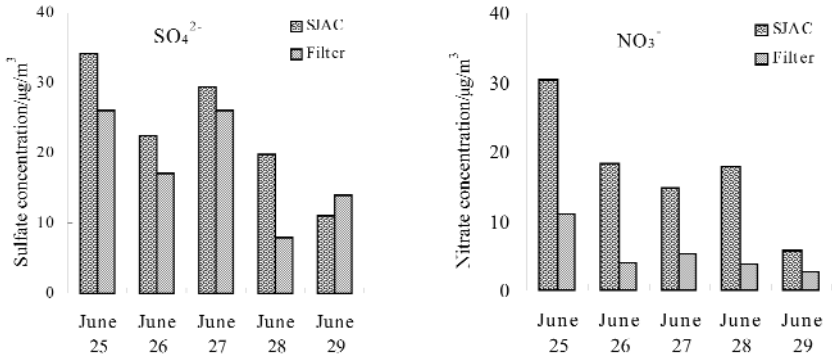

Fig. 5 Comparison of 24-h averages of SO_4^{2-} and NO_3^- by filter pack and SJAC methods in Beijing (2000).

The concentrations of OC, SO_4^{2-}, NO_3^-, and NH_4^+ in other countries are listed in Table 2. The concentrations of those aerosol components in Chinese cities (see Table 1) are quite high by comparison. In particular, the concentrations of SO_4^{2-} and NO_3^- are generally about 2–4 and 5 times higher, respectively. This is mainly due to strong local emissions of SO_2 and NOx and the high oxidation capacity of the atmosphere. However, the case was not same for EC. The concentrations of EC in China are lower than that in São Paulo and comparable to those in Kaohsiung, Sapporo, and Birmingham.

Table 2 Chemical compositions in other areas (μg/m^3).

City	Year	OC	EC	SO_4^{2-}	NO_3^-	NH_4^+
Kaohsiung (Taiwan) [21]	Nov. 1998–April 1999	10.4	4.0	ND[b]	ND	ND
Hong Kong[a] [15]	1990–1994	ND	ND	8.5	2.03	2.5
Sapporo (Japan)[a] [17]	June 1987–Dec. 1988	ND	3.4	3.9	0.9	0.85
Birmingham (UK) [18]	Feb. 1992, Aug. 1992	ND	1.2	4.1	1.0	1.6
Ankara (Turkey) [32]	Feb. 1993–June 1993	ND	ND	6.4	2.1	
Sequoia National Park (California, USA) [33]	May 1999–Nov. 1999	ND	ND	0.6–1.5	1.1–2.0	1.0–1.9
São Paulo (Brazil) [19]	July 1997–Sept. 1997	15.8	7.6	ND	ND	ND

[a]Chemical components of PM-10.
[b]Not detected.

The averaged chemical compositions of PM-2.5 in Beijing [25] and Guangzhou [26] cities are shown in Fig. 6. In Beijing, organic carbon (OC) is the largest fraction in PM-2.5, accounting for 36 %.

Sulfate, nitrate, ammonium, and EC represent respectively 9, 5, 5, and 5 %. The sum of these species represents around 65 % of the mass of PM-2.5 while the contribution of crustal components is only 12 %. This implies that secondary aerosol is the largest fraction of PM-2.5 owing to fossil fuel emissions, and that fugitive dust represents a rather small contribution. Chemical characteristics of PM-2.5 are quite similar in Beijing and Guangzhou.

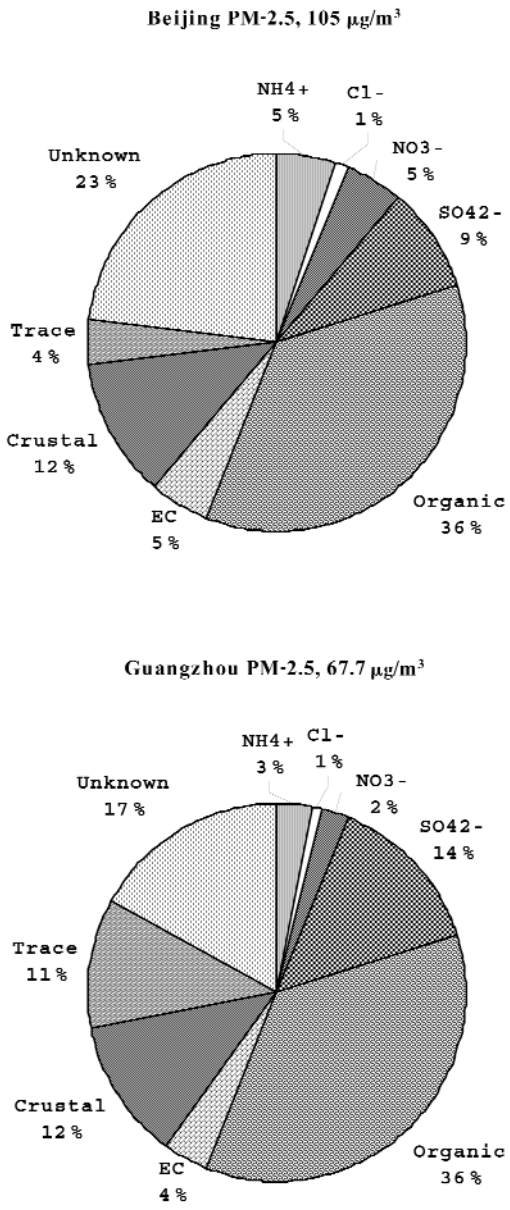

Fig. 6 Chemical composition of PM-2.5 in Beijing and Guangzhou urban area in 2000.

3.2 Diurnal variations of sulfate and nitrate concentrations

The diurnal variations of SO_4^{2-} and NO_3^- concentrations in fine particles in Beijing for different seasons of the year 2000 are given in Fig. 7 [25]. Sulfate concentrations in May, June, September, and

December vary typically over a wide range of 5–50 μg/m^3. The low concentrations are largely due to meteorological conditions, especially precipitation. In June (June 2000 in the figure), a sulfate peak is found during 10:00–12:00 a.m., 50 μg/m^3, which is caused by strong photochemical activity and fast conversion of sulfur dioxide to sulfate in summertime. In December, the peak SO_4^{2-} concentration appears 2 h earlier (0800–1000 h, Dec. 2000) and is lower than that in June. This peak was is probably the result of sulfur dioxide emissions and heterogeneous oxidation of SO_2 in wintertime.

Elevated PM-2.5 nitrate concentrations are found in May and June of 2000 in Beijing. Nitrate concentrations are typically 8–55 μg/m^3 in May and June, and higher than 10 μg/m^3 in September and December. In May and June, the diurnal variation shows a peak in nitrate concentrations (about 30 μg/m^3 in May and 56 μg/m^3 in June) during morning hours (1000–1200, May and June 2000 in the right-hand part of Fig. 7). Comparison of the seasonal and diurnal variations of sulfate and nitrate shows that these two ions have the same concentration level in June owing to the relatively low emission of SO_2 in summertime. In December, PM-2.5 sulfate concentrations exceed the nitrate concentration by a factor of 3 because of additional SO_2 emissions from residential heating. Another factor could be the slower conversion of nitrogen oxides to nitrate as photochemical activity in wintertime is at low level. The high nitrate concentrations have the implication that attention should also be given to NOx control in abatement measures of fine aerosol.

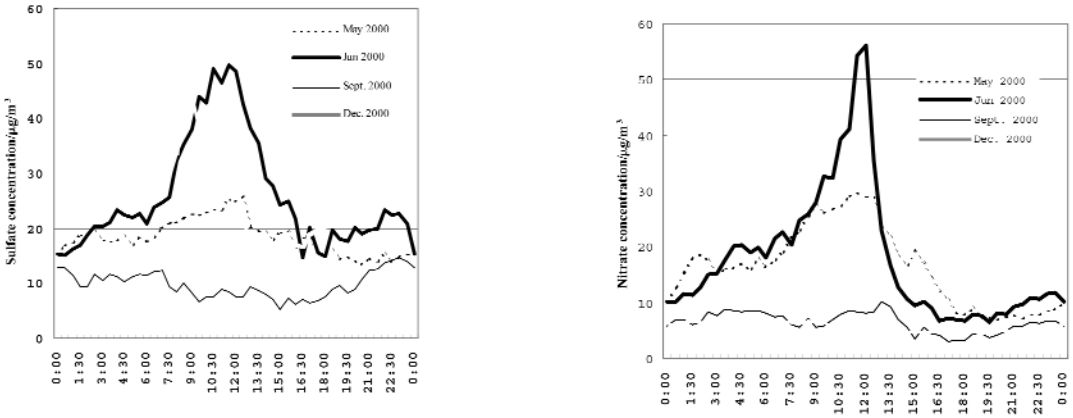

Fig. 7 Diurnal variations of SO_4^{2-} and NO_3^- concentrations in Beijing by SJAC online measurements (2000).

4. SOURCES OF AEROSOL

A chemical mass balance (CMB) receptor model has been developed, based on the presence of elements, ions, OC/EC, and polycyclic aromatic hydrocarbons (PAHs) in aerosol to assess the contributions of different sources of airborne fine particle mass. The model has been applied to the Beijing area using annual average chemical composition and source profiles obtained from measurements and the literature. Eight sources of particles are identified, including coal burning, soil dust, construction dust, road dust, vehicle exhaust, biomass burning, organics, and secondary sulfate and nitrate, as shown in Fig. 8 [25]. Approximately 53 % of PM-2.5 mass is assigned to primary sources and 30 % to secondary sources. The largest contribution to primary aerosol emissions in Beijing is found to be fugitive dust (10 % for road dust, 14 % for soil dust, and 3 % for construction activities) mainly because of the dry climate, low rate of forest coverage, and occasional dust storms. Coal burning constitutes an important source in Beijing with the contribution of 15 %, owing to widespread use of coal in industrial processes, electricity generation, and domestic heating. Particles in vehicle exhaust are responsible for only 6 % of PM-2.5 mass concentration. A contribution of biomass burning is detected, attributed to combustion

of corn straw in summer and autumn and biofuel in other seasons, and estimated to be 5 % on an annual basis. About 30 % of the mass of fine particles can be attributed to secondary formation of sulfate, nitrate, and organics. The large contribution of secondary aerosol would reflect fast conversion of gaseous primary emissions as sulfur dioxide, nitrogen oxides, and gaseous organic compounds to secondary products as sulfates, nitrates, and nonvolatile oxidized organics in Beijing.

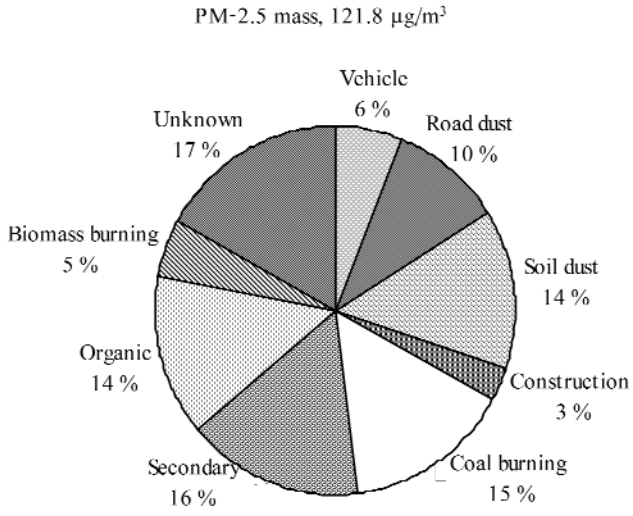

Fig. 8 Source apportionment of ambient PM-2.5 in Beijing (2000).

5. CONCLUSION

Some data sets for aerosol mass and composition, obtained both in our recent research and from the literature, are presented and evaluated. The results give an overview on status and characteristics of aerosol pollution in major cities in China.

- TSP and PM-10 mass concentrations are so high in many areas of China that the national standards are exceeded significantly and frequently. PM-2.5 concentration is also very high, and the annual mean concentration varies from about 50 to 140 μg/m^3 in different areas.
- According to some studies, PM-2.5 constitutes about 60 % of the PM-10 mass on average. This fraction of fine particles affects the visibility and causes air pollution on a regional scale.
- Organic carbon and sulfate are the most abundant components of PM-2.5, while crustal elements constitute a minor part of PM-2.5. In summertime, nitrate is present in almost same concentration levels as sulfate, which implies that NOx control should be emphasized to abate fine particle pollution and to improve visibility.
- Results of source apportionment by CMB show that fugitive dust, coal burning, industrial processes, and vehicle exhaust are the main sources of primary aerosol. Secondary aerosol as represented by nitrate and sulfate contributes 16 % to PM-2.5 mass concentration, and organic carbon aerosol 14 %. This indicates fast conversion of primary emissions in Beijing.

6. ACKNOWLEDGMENT

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