Pergamon

*Atmospheric Environment* Vol. 30, No. 13, pp. 2355–2362, 1996

1352-2310/96 $15.00 + 0.00

1352-2310(95)00405-X

# AEROSOL SIZE DISTRIBUTION AND ELEMENTAL COMPOSITION IN URBAN AREAS OF NORTHERN CHINA

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(*First received* 3 *August* 1994 *and in final form* 16 *September* 1995)

**Abstract**—Based on monitoring data collected in the northern cities of China since 1980, the particle size spectra, distribution modes and the elements distribution features are discussed in this article. It is found that the major aerial contaminant in urban aerosol of northern China is fine dust. It makes up about 70% of the total suspended particulate matter by mass concentration. The spectra of aerosol particles in the range 0.5–15.0 $\mu$m are all unimodal structures. Regardless of narrow or broad profiles, their peak values fall between 0.5 and 1.0 $\mu$m, while their surface area spectra present a trimodal structure. The three peaks are in the range 0.5–1.8, 2–8 and 10–20 $\mu$m, respectively. Of these peaks, the surface area concentration of particulates had made a large contribution to the first and the second peaks. The shape of mass distribution also presents a trimodal structure, and the positions of the three peaks are the same as those found in the surface area pattern. Mineral elements mainly exist in large size particles. The anthropogenic pollution elements, however, are contained in corpuscles ($< 2.0$ $\mu$m). Compared to the situation in winter, some element concentrations of particulates in summer are found to have generally decreased. Copyright © 1996 Elsevier Science Ltd

*Key word index*: Aerosol size distribution, elemental composition, aerodynamic diameters, TSP in China.

## 1. INTRODUCTION

Aerosols in general are a type of complex material contained in various organic and inorganic compounds, and there are obvious differences in size, volume, shape, chemical constituent, etc. Among them, they can absorb and scatter the solar radiation in the long wave region, and they can alter the balance of the earth–atmosphere system. As a result, climate change in all scales may result. Moreover, aerosols play a carrier's role, i.e. they deliver chemical compounds from certain places to other places. Generally speaking, aerosols in the stratosphere may cause a cooling effect on the air near the ground, while in the troposphere they may lead to warming of the air layer where the atmospheric particulates exist. In addition, aerosols have a strong surface absorption which can expedite the absorption of $SO_2$ and can oxidize it to become $SO_4^{2-}$ with time. Through the action of photooxidation, eventually $SO_2$ in the atmosphere will probably be oxidized into $H_2SO_4$, and may form acid deposits.

Different sizes of particulate matters may cause different environmental effects. For example, acid rain formation is affected by the high content of chemical aerosols contained in man-made pollutants with a particle size in the diameter range 0.001–2 $\mu$m. As aerosol size decreases, the pH value and the content of $Ca^{2+}$ decrease, but the contents of $SO_4^{2-}$ and $NH_4^+$ increase. This is one of the important factors for acid rain formation (Zhao and Lin, 1987). A study shows that there is a serious amount of suspended particulate matter in widespread cities over northern China (Ning *et al.*, 1987), and that fine particles which are less than 2 $\mu$m in aerodynamic diameter are able to penetrate into the lung (Wang and Yang, 1986; Han, 1987), and consequently impair public health. In addition, yellow sand with a diameter between 0.5 and 3.0 $\mu$m originating from a loess plateau can be transported to Korea, Japan, and occasionally to distant Hawaii by the westerlies (Braaten and Cahill, 1991; Chung, 1992; Tanaka, 1989).

The purpose of the present study is to show the results of our analyses on total suspended particulates (TSP) and element particles measured in five cities in northern China. The characteristics of dust pollution observed in China are also compared with the result obtained in developed countries.

Table 1. Mass concentration distribution of atmospheric particulates (mg $m^{-3}$)

| Diameter ($\mu$m) | Beijing | | Shengyang | | Taiyuan | |
|---|---|---|---|---|---|---|
| | Summer | Winter | Summer | Winter | Summer | Winter |
| > 7.0 | 0.15 | 0.26 | 0.04 | 0.05 | 0.20 | 0.46 |
| 3.3–7.0 | 0.05 | 0.08 | 0.02 | 0.04 | 0.06 | 0.11 |
| 2.0–3.3 | 0.02 | 0.06 | 0.01 | 0.02 | 0.04 | 0.06 |
| 1.1–2.0 | 0.02 | 0.05 | 0.02 | 0.03 | 0.03 | 0.07 |
| < 1.1 | 0.08 | 0.23 | 0.04 | 0.09 | 0.12 | 0.26 |
| Total | 0.32 | 0.68 | 0.12 | 0.23 | 0.45 | 0.96 |

## 2. MASS CONCENTRATION AND PARTICLE SIZE DISTRIBUTION OF TSP

The aerosol particle size discussed in this article refers in particular to the aerodynamic size. In 1985 and 1986, Cao *et al.* (1991) measured that the percentages of fine dust ($d < 10$ $\mu$m) to TSP in mass concentration in five cities were all about 70% particles, which suggests that the major portion of aerosols is fine dust.

Table 1 shows that over regions studied, the total mass concentrations of particles observed in the winters of 1986 and 1987 are about twice as high as they are in summer. In addition, both in summer and in winter, the highest mass concentrations appear in the particles size group between $d > 7.0$ and $d < 1.1$ $\mu$m. It can be seen from Table 2 that coarse particles rise higher than fine ones except over Shengyang in summer.

Over the central quarter in Dalian, the size distribution of TSP is presented in Table 3. During the heating season, the mass concentration of the particle with $d < 5$ $\mu$m make up 65% of TSP: $d = 5$–$10$ $\mu$m, 22%; $d > 10$ $\mu$m, 13%. In the non-heating season, the mass concentration of particles with $d < 5$ $\mu$m make up 61% of TSP: $d = 5$–$10$ $\mu$m, 20%; $d > 10$ $\mu$m, 18%. Whether during the heating season or during the non-heating season, the percentages of inhalable particles with $d < 5$ $\mu$m are quite high as a result of less dust-laden wind and secondary dust raising in the coastal city which is different from the other inland northern cities.

Table 2. The ratio of coarse to fine particles in aerosol particulates

| Place | Season | $d > 2.0$ $\mu$m | $d < 2.0$ $\mu$m | Coarse vs fine |
|---|---|---|---|---|
| Beijing | Summer | 67.7 | 32.3 | 2.10 |
| | Winter | 59.8 | 40.2 | 1.49 |
| Shengyang | Summer | 47.1 | 52.9 | 0.89 |
| | Winter | 62.0 | 38.0 | 1.63 |
| Taiyuan | Summer | 64.6 | 35.4 | 1.82 |
| | Winter | 66.8 | 33.2 | 2.01 |

Table 3. Mass concentration of each size classification in downtown of Dialian (mg $m^{-3}$)

| Diameter ($\mu$m) | Heating season | Non-heating season |
|---|---|---|
| < 5.0 | 0.110 | 0.063 |
| 5–10 | 0.037 | 0.020 |
| > 10 | 0.021 | 0.019 |
| Total | 0.168 | 0.103 |

## 3. FEATURES OF AERODYNAMIC SIZE SPECTRUM PATTERN

### 3.1. *Particle spectrum*

Urban particle spectra measured by laser Aerodynamics Spectrometer are illustrated in Fig. 1. The figures show particle concentrations vs spectrum features. The profile in general shows that no matter what type of city one is in, the spectra of aerosol aerodynamic size in the range 0.5–15 $\mu$m are unimodal in structure, and their peaks lie between 0.5 and 1.0 $\mu$m. There are different features, however, in various areas of different cities (Han, 1987; Zhao *et al.*, 1987, 1988), which can be classified into two types: narrow and broad profile types. The former includes downtown areas of Beijing and Shengyang. They indicate that the aerodynamic sizes of urban aerosol centralize in a small range and that their peaks fall in the range of about 0.8 m in diameter. Broad profile type, which is in the industrial areas of Lanzhou (Xigu) and Taiyuan, is unlike the narrow type. The profiles in highly industrialized areas have peak values which shift towards smaller sizes and fall in the range 0.6–0.7 $\mu$m.

### 3.2. *Surface area spectrum*

Figure 2 reveals that the patterns of surface area distribution of urban aerosols vs aerodynamic size between 0.5 and 15.0 $\mu$m basically present a trimodal structure. Their peak values fall into groups of about 0.5–1.8, 2.0–3.0 and 10–20 $\mu$m, respectively. In current coal-burning polluted northern cities, particle surface areas of the first and second peaks make an extremely large contribution. In the case of Beijing and Shengyang more specifically, the first peak makes the largest contribution to total surface area concentration, while the third peak contributes the least. The average result

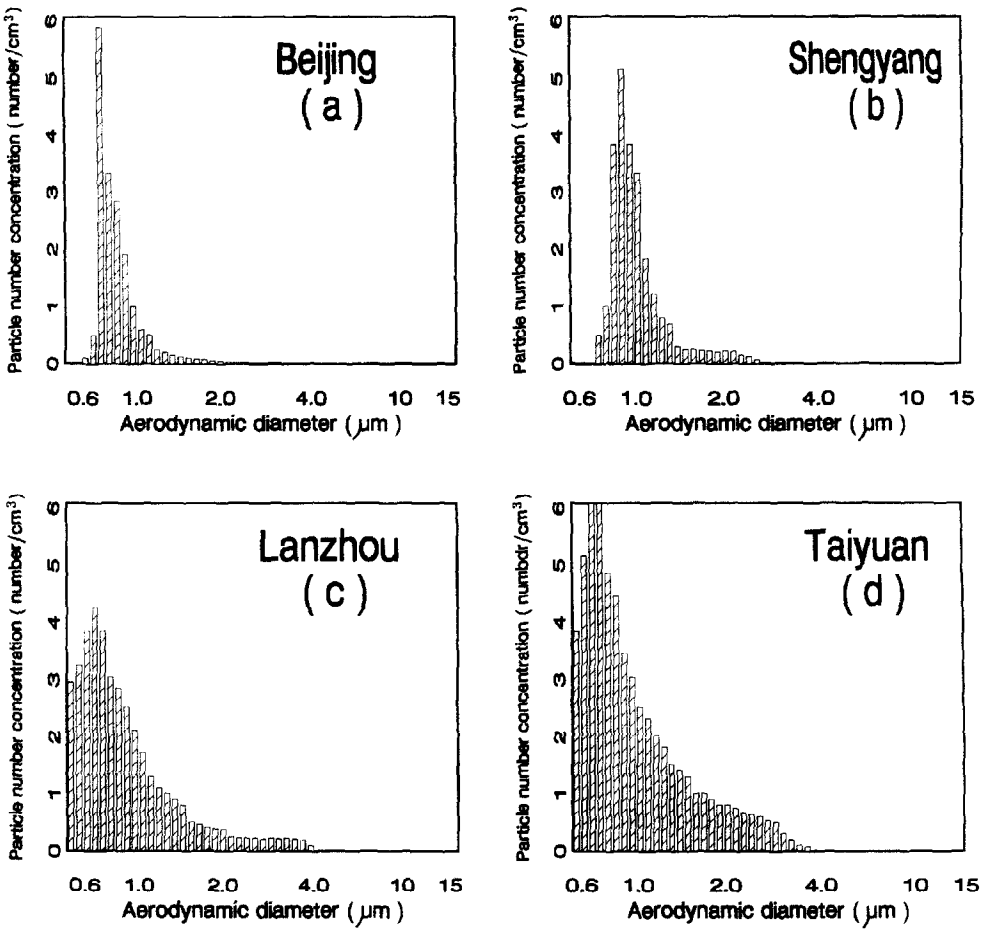


Fig. 1. The pattern of particle number distribution vs aerodynamic size of particles.

monitored in downtown Beijing during winter shows that the particle's surface area of the first peak makes up 76.6% of whole surface area of aerosol measured, the second peak makes up 17.9% and the third one, 5.5%. The pattern of average data in downtown Shengyang during winter is generally similar to the pattern of Beijing. The contribution of the second crest, however, has obviously increased, and the proportion of the first peak has comparatively reduced. In Lanzhou and Taiyuan, however, the contribution of surface area concentration from the first and second peaks in the range of large size has prominently increased. It will be seen that the strength of traffic emission as a source of pollution in Beijing and Shengyang is strong, and the relative content of aerosol particles in accumulation mode is higher than that in highly industrialized regions such as illustrated in Figs 2c and d.

### 3.3. *Mass spectrum*

It is evident from Fig. 3 that all aerodynamic diameters of measured particles fall in the range 0.5–15 $\mu$m and all the mass spectrum patterns of northern urban aerosol have typical trimodal shapes. Their three peaks emerge in the range 0.5–1.8, 2.0–8.0 and 10–20 $\mu$m, respectively, which is in line with the corresponding range of the three peaks in the surface area spectrum (Han, 1987; Zhao *et al.*, 1987, 1988). By comparing the three peaks of mass spectrum with each other, it is evident that the third peak makes a larger contribution in particle mass concentration to the whole mass concentration, which is quite unlike the result measured in western countries, where the mass spectrum is a bimodal shape with the peak values falling in the range 0.2–0.3 $\mu$m and about 10 $\mu$m, respectively (Whitby *et al.*, 1972, 1975). This diversity mainly results from different pollution sources. There are three major pollution sources for urban TSP in northern China: (1) *secondary pollutants* (fine particle) *source*: These mainly come from industries and the emission of cars, going through a series of physical and chemical transformations. They are a very important contributor to the first peak of mass concentration. (2) *Industrial processing and burning*:

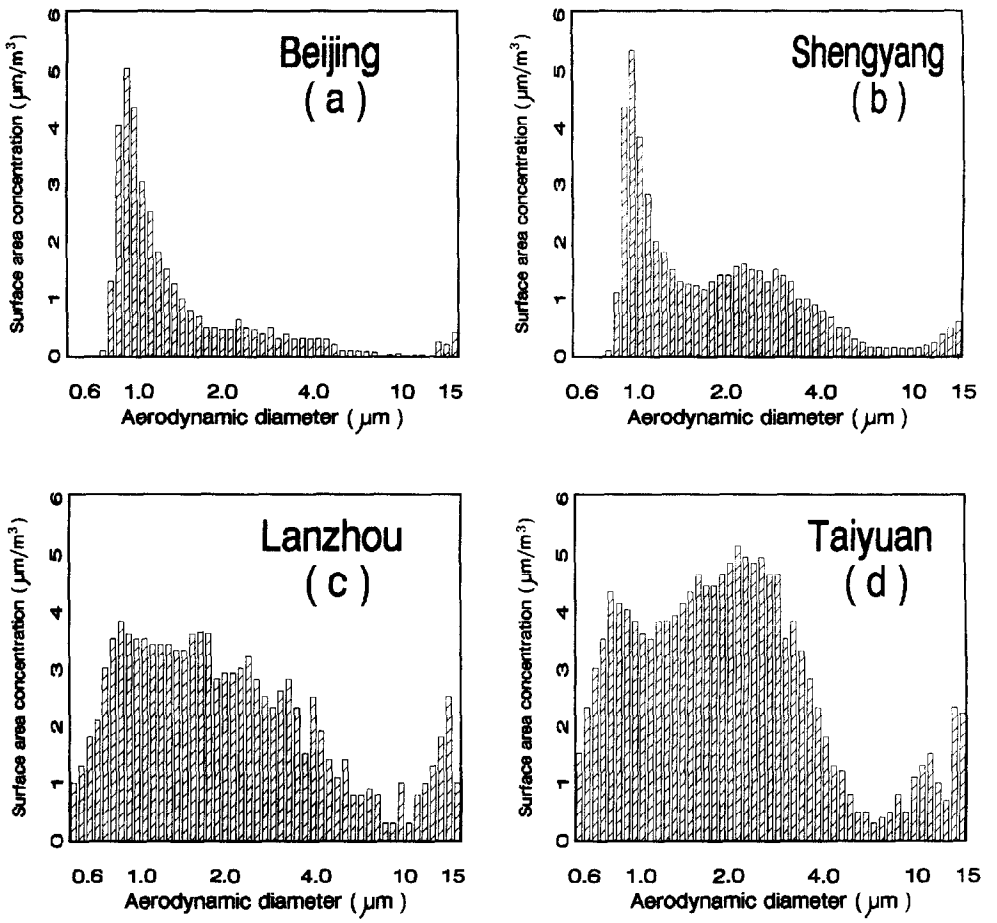


Fig. 2. The patterns of surface area distribution vs aerodynamic size of particles.

These are major contributors to the second peak of distribution by mass. (3) *Dust-fall*: This originates mainly from traffic, construction sites, and mechanical pulverization. Unfortunately, there is little information about the third source in developed countries.

Although the appearance range of the three peaks is identical for the centre of a city and highly industrialized areas, the intensity of the second peak in Lanzhou (Xigu) and Taiyuan is much larger than that in Beijing and Shengyang. However, the secondary pollutant problem, in Beijing and Shengyang, is much more important than that of Lanzhou and Taiyuan. Beijing and Shengyang are larger cities than the other two.

The distribution shape difference is most likely a result of different types of particle pollution sources. Figures 4 and 3b reveal the variation from remote suburban to downtown Shengyang (Wang and Zhao, 1991). Under a background condition in the windward countryside (Fig. 4b), there is a little influence from traffic and industry. Therefore, the first and the second peaks almost disappear but the third one is on the right side of 10 $\mu$m. We conclude that the third peak is the result of natural soil dust. The most prominent feature in Fig. 4a is the strong, broad second peak, which indicates a contribution from the pollution of industrial production and burning processes. In Fig. 3b, pollution from tail gas emissions and secondary dust-raising produced from urban heavy traffic may play a decisive role in aerosol pollution. Thus, the corresponding peaks in the diameter range 0.6–1.8 $\mu$m and greater than 10 $\mu$m are very distinctive.

## 4. VARIATION OF ELEMENT CONTENT WITH PARTICLE SIZE

By monitoring the aerosol compositions in five cities, Beijing, Taiyan, Zibao (Shandong province) etc., Cao *et al.* (1991) found that about 80–90% of the element which enters the atmosphere due to anthropogenic pollution exist in corpuscles with diameters less than 10 $\mu$m. Also, Winchester and Bi (1984) analysed the elements of TSP in the downtown and countryside areas of Beijing, and found that the concentrations of Cr, Mn, Fe were quite high and of the fine particle type; possibly a result of industrial

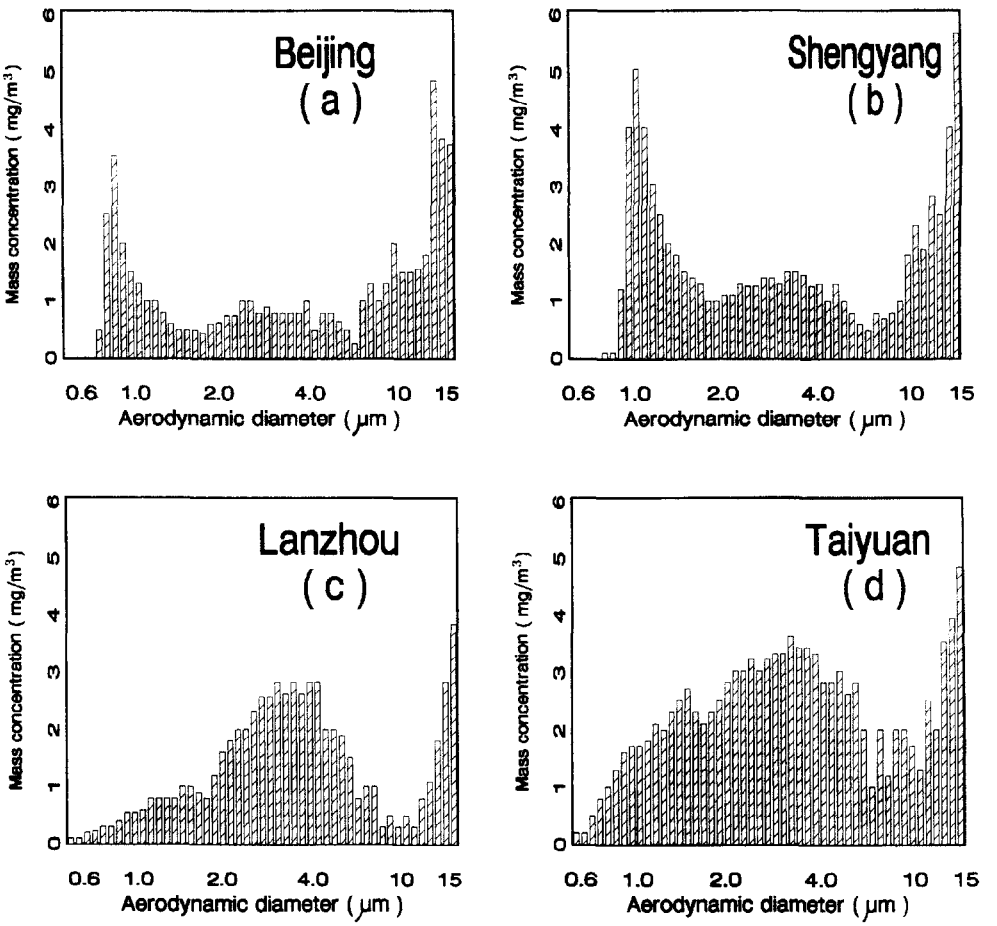


Fig. 3. Patterns of mass spectrum vs aerodynamic diameter of aerosol.

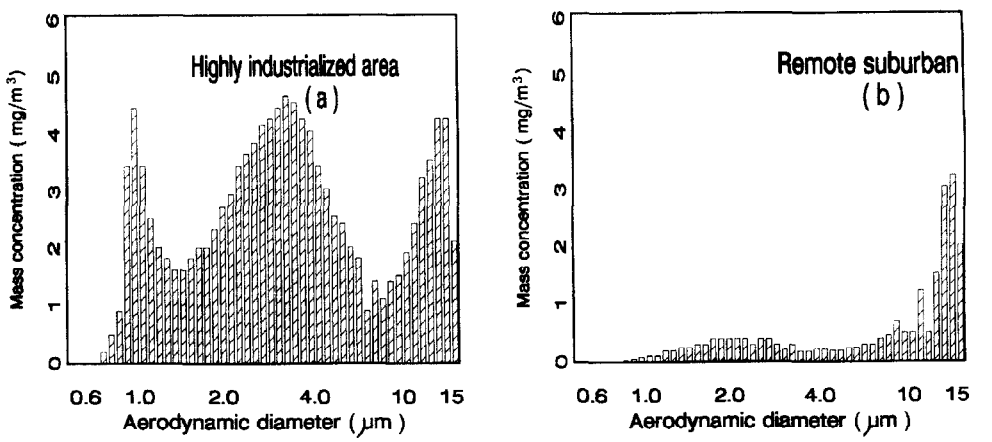


Fig. 4. Comparisons of mass distribution in different areas of Shengyang.

emissions. The silicon content in fine particles, which may come from coal-burning under high temperature, was also quite significant. The Pb, Zn and Cu contents in coarse particles ($2.0 < d < 15$ μm) in downtown were higher than those in distant villages. The element distribution feature in suspended particulate matters in Shengyang was similar in Beijing in many ways. But mineral elements, which mainly existed in coarse particles and a very substantial part derived from human activities, were found in inhalable particles (Ye *et al.*, 1987).

The monitoring on the campus of Beijing Normal University goes further into the question of the relationship between element content and particle size distribution. The 10 stages of cascade impactor are classified in Table. 4

In winter, the elements Sr, Ca, Fe, Ti, V, Co and Si present a unimodal shape and are mainly concentrated in the range of coarse particle mode (4–8 μm); on the contrary, the elements K, Mn, S, Zn, Pb, Ni, Cu, Br, Cl, As and Se present a bimodal structure and are mainly concentrated in the range of fine particle mode (0.5–1.0 μm) and coarse particle mode. The shape of a bimodal structure reveals that there are contributions from both natural and anthropogenic sources. There is a difference in distribution patterns between the element's mean contents and their particle sizes in summer and winter (Fig. 5). Whether for coarse particles or fine particles, the element's mean contents in summertime have comparatively decreased, especially in the range of coarse particle mode, such as in spectrum distribution of S, Pb, Zn. These suggest a large quantity of coarse particles emission during heating season. The coarse particulates may originate from the inadequate burning of smaller boilers and domestic stoves.

Table 4. The diameter of 10 stages of cascade impactor

| Stage no. | Diameter (μm) | Stage no. | Diameter (μm) |
|---|---|---|---|
| 0 | < 0.06 | 1 | 0.06–0.12 |
| 2 | 0.12–0.25 | 3 | 0.25–0.50 |
| 4 | 0.50–1.00 | 5 | 1.00–2.00 |
| 6 | 2.00–4.00 | 7 | 4.00–8.00 |
| 8 | 8.00–16.0 | 9 | > 16.0 |

## 5. SUMMARY AND CONCLUSIONS

Based on our measurements and analyses the following summary and conclusions can be given.

(1) The major ambient contaminant in the urban aerosol of northern China is fine dust. It makes up about 70% of total suspended particulate matter by mass concentration.

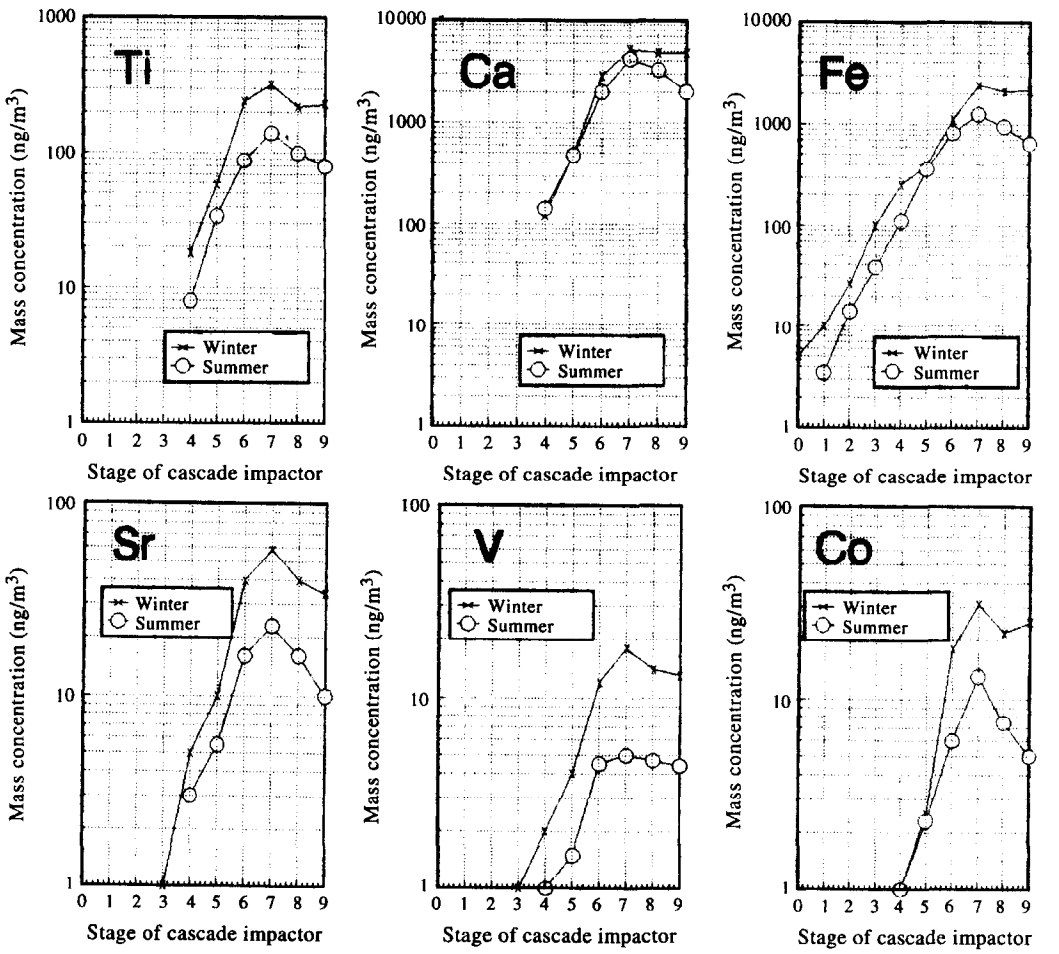


Fig. 5a. Distribution patterns of element contents–particle sizes during the heating and non-heating seasons.

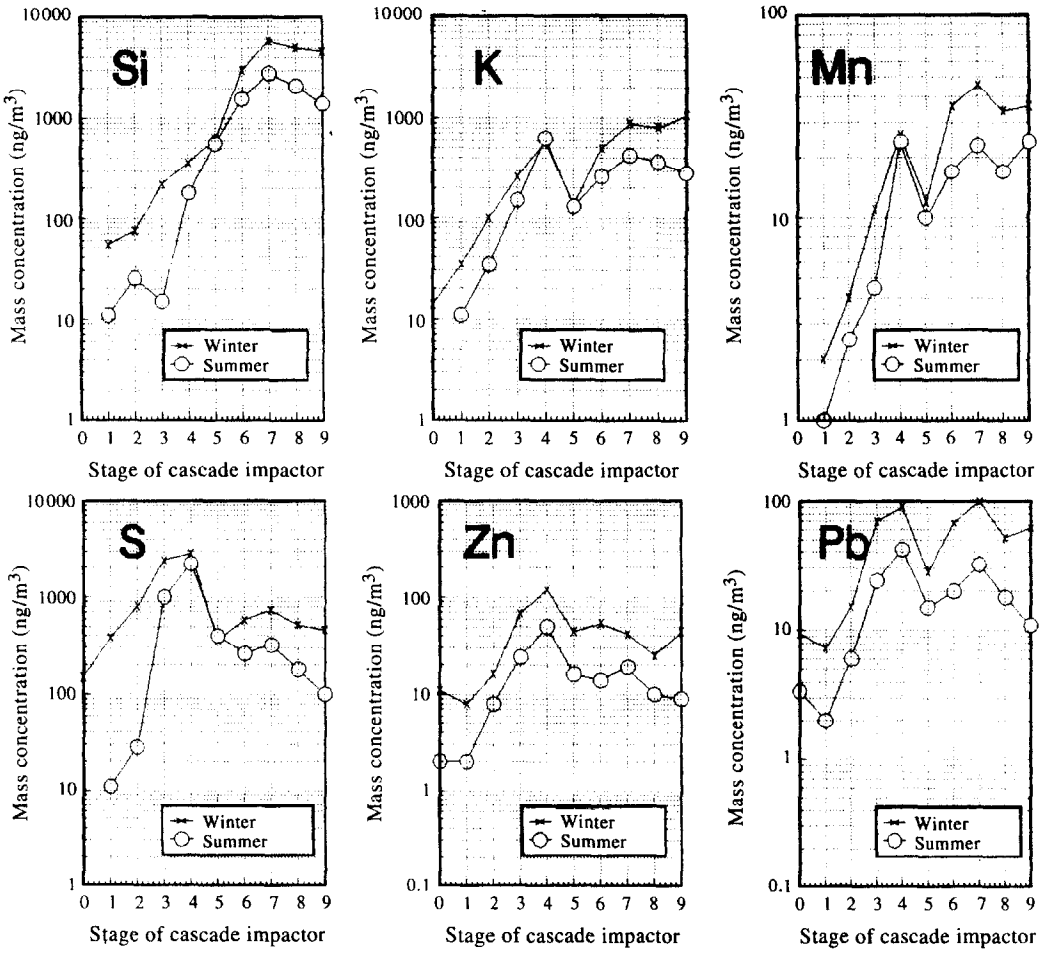


Fig. 5b. Distribution patterns of element contents–particle sizes during the heating and non-heating seasons.

(2) The spectra of aerosol particles in the range 0.5–15.0 μm are all unimodal structures. Regardless of narrow or broad profiles, their peak values fall in the 0.5–1.0 μm range, while their surface area spectra present trimodal structures. The three peaks lie within the range 0.5–1.8, 2–8, and 10–20 μm, respectively. Of these, the surface area concentrations of particles have made an enormous contribution to the first and the second peaks. The shape of mass distribution also presents a trimodal structure. The mass spectrum pattern in developed countries, however, presents a bimodal structure because the dust is basically eliminated at the source.

(3) Crystal elements mainly exist as large size particles. The anthropogenic pollution elements, however, are contained in corpuscles ($d < 2.0$ μm).

(4) In the northwest part of Beijing, the pattern of element concentration vs size of aerosol particles can be classified into two shapes: (1) unimodal structure; its peak value mainly falls into coarse particles with diameters between 4 and 8 μm; (2) bimodal structure; its peak value is mainly concentrated in the range 0.5–1.0 and 4–8 μm. Compared to the situation in winter, the concentration of some elements, such as Fe, Ti, Zn, Pb generally decrease during summer.

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