# Characteristic difference of spheroidal carbonaceous particles (SCPs) in surface sediments from Japanese and Chinese cities revealed by their chemical composition

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**Abstract** Spheroidal carbonaceous particles (SCPs) are produced by the high-temperature combustion of fossil fuels and emitted to the atmosphere. We examined the surface morphology and the chemical composition of SCPs in surface sediment samples from five industrial cities in Japan and China respectively, using SEM (scanning electron microscope) with EDS (energy-dispersive spectroscopy). Although there is a relationship between surface morphology and chemical composition of SCPs in China, chemical composition of SCPs in Japan does not depend on their surface morphology. The chemical compositions of SCPs are different between in Japan and China; SCPs in Japan are S-rich and those in China are Ti-rich. This suggests that we can find out the influence of China-derived pollutant particles, i.e., SCPs, in East Asian countries, such as Japan, South Korea and Taiwan, by EDS analysis on SCPs.

**Keywords:** chemical composition, China, East Asia, Japan, spheroidal carbonaceous particle, surface morphology, surface sediments

## Introduction

Spheroidal carbonaceous particles (SCPs), that are one of the pollutant particles, are produced by the high-temperature combustion of fossil fuels and emitted from thermal power station and other industries. Because under favorable conditions, these particles are deposited in sediments, particles from different sources can then be mapped, revealing the pattern of particle distribution in regions and entire countries (e.g., Griffin and Goldberg 1979, 1981; Rose et al. 1994, 1996, 1999; Rose 1995; Alliksaar et al. 1998; Hirakawa et al., 2011). Surface morphology and chemical compositions of SCPs are useful in terms of identification of the source fuel (e.g., coal, oil and oil shale) of SCPs (Griffin and Goldberg, 1979, 1981; Rose et al., 1994, 1996; Murakami-Kitase et al., 2010). Hirakawa et al. (2011) suggested that ~20μm SCPs in sediments from the industrial cities could represent the local combustion history in detail, as the influx of local SCPs is dominant at such sites. Larsen (2003) indicated larger particles (> 10μm) dominated in lakes closest to the city. The SCPs of 10-20 μm in size deposited in sediments from industrial cities could represent the local combustion history.

In East Asia, fossil fuel combustion has increased with industrial growth especially in China, which caused a problem of regional air pollution in the region. These pollutants are gasses (e.g., $SO_2$ and $NO_X$) and particles (e.g., smaller SCPs). In China, thermal stations and other industries use mainly coal, whereas in Japan, South Korea and Taiwan mainly oil is used in primary energy. In China, on a weight basis, coal consumption is three times larger than that of oil consumption, whereas in Japan coal consumption is approximately half of that of oil consumption (data is available at the BP web site, http://www.bp.com/). Taking into account of the proportion between oil and coal consumption in South Korea and Taiwan, the characteristic in SCPs produced in these countries are supposed to be similar to those in Japan rather than those in China. Therefore, it is possible to identify the Chinese SCPs from other industrial countries.

We have studied SCPs in sediment samples collected from many cities in Japan and China, to clarify the spatial and temporal distributions of SCPs in East Asia and identify their sources. Some results have been published (e.g., Murakami-Kitase et al., 2010; Hirakawa et al., 2011). Hirakawa et al. (2011) examined chemical composition and surface morphology of SCPs extracted from sediments core samples collected from three cities (Tokyo, Osaka and Nagasaki) in Japan and one city (Beijing) in China. They indicated that chemical compositions of the Chinese SCPs differ from those of the Japanese SCPs deposited during the 1990s and 2000s.

In this study, we examined chemical and morphological characteristics of SCPs in surface sediments deposited in five Japanese industrial cities (Chiba, Tokyo, Osaka, Yokkaichi and Nagasaki) and five Chinese cities (Beijing, Tianjin, Qingdao, Nanjing and Shanghai) in detail, to clarify difference in characteristics of the present-day SCPs between Japan and China. Data of Beijing are referred to Hirakawa et al. (2011).

## Materials and Methods

The sediment core samples were collected from Chiba, Tokyo, Yokkaichi, Osaka and Nagasaki in Japan and Beijing, Tianjin, Qingdao, Nanjing and Shanghai in China (Fig.1). We used surface sediments (from surface to at most 6 cm depth) for SCPs analysis, many of which (Chiba, Tokyo, Osaka, Nagasaki, Beijing, Tianjin and Shanghai) were dated with $^{210}$Pb profiles; they deposited within the last ~10 yrs. Table 1 provides brief descriptions of the analyzed samples in this study. Hirakawa et al. (2011)

suggested that ~20 μm SCPs in sediments in industrial cities represent the local combustion history in the case that the city is located ~500 km away from other cities where large amounts of SCPs are emitted. In this study, the cities in China are located at least ~100 km away from each other; those in Japan are located at least ~50 km away from each other. In fact, there are differences in chemical composition of SCPs among cities in Japan. Therefore, most SCPs collected from Japanese cities and Chinese cities may be produced in and around each city.

Following the extraction method that modified from Murakami-Kitase et al. (2010), samples were subjected to chemical digestion to remove all material except for SCPs. Digestion involved the sequential application of 60% concentrated nitric acid ($HNO_3$), 10% Hydrogen peroxide ($H_2O_2$), 46% hydrofluoric acid (HF), and 6M hydrochloric acid (HCl). The extracted SCPs were mounted on glass slides and coated with a thin layer of carbon. Observations of the surface morphology and chemical analyses of SCPs were performed using a JEOL JSM-5500 scanning electron microscope (SEM) equipped with an EDAX-EDS system (CDU-LEAP detector + Genesis software), housed at the Department of Geosciences, Osaka City University, Japan. The SEM was operated at an accelerating voltage of 20 kV and beam current of 500 pA. The obtained X-ray spectra confirm that the SCPs consist mainly of carbon. We used EDS to quantitatively analyze the concentrations of Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, and Zn. The intensities of the characteristics X-rays were corrected using ZAF method. Using the above method, we analyzed at least 25 particles in each sample (Table 1), except for sample in Beijing referred to Hirakawa et al. (2011). Particles that we measured are 5–50μm in diameter and most of the particles are 10-20μm.

## Result and Discussion

Based on SEM observation on surface morphologies of SCPs, they are classified into two categories: non-convolute type and convolute-type, although some SCPs cannot be classified into two categories (Table 2). Surfaces of the SCPs of the non-convolute type are smooth, rough or irregular, whereas those of the convolute type are convoluted and layered. According to Griffin and Goldberg (1979, 1981), the non-convolute type SCPs are derived mainly from coal combustion and the convolute-type ones are from oil combustion. Of 204 SCPs found in Japan, non-convolute-type is 115 (56%); convolute-type is 57 (28%); and other or unclassified type is 32 (16%). Of 169 particles found in China, non-convolute-type is 132 (78%); convolute-type is 7 (4%); and other or unclassified type is 30 (18%). The ratio of non-convolute to convolute type in China (20:1) is much higher than that in Japan (2:1). These ratios may reflect the ratios of fuel types used in thermal stations and other industries between China and Japan. In China, thermal stations and other industries use mainly coal whereas in Japan mainly oil is used in primary energy; in China on a weight basis coal consumption is three times larger than that of oil consumption, whereas in Japan coal consumption is approximately half of that of oil consumption.

Chemical compositions of SCPs in Japanese and Chinese cities are shown in Table 3. Difference in chemical compositions among Japanese cities is small; average compositions of SCPs are Si (37wt%), S (24wt%), Na (10wt%) and Ca (5wt%). Difference in chemical composition among Chinese cities is also small and averaged compositions are Si (44wt%), Ti (12wt%), Na (11wt%) and Ca (6wt%), whereas chemical difference between Japanese cities and Chinese ones is large, especially for S and Ti. Figure 2 shows relationship between S/Si and Ti/Si for SCPs from Japan and China. The SCPs from different countries plotted in different fields, although some of them are overlapped. In each field, they contain both the non-convolute type SCPs and convolute type ones, suggesting that difference in chemical compositions of SCPs between Japan and China are much larger than those between surface morphologies in each country.

The previous studies (e.g., Murakami-Kitase et al., 2010) indicated the difference of the surface morphology of SCPs is useful in terms of identification of the source fuel type of SCPs, because there is clear relationship between surface morphologies and chemical compositions of SCPs. However, some researchers suggested that chemical compositions of SCPs are much useful rather than their surface morphology, because the non-convolute type SCPs can be emitted from oil combustion and the convolute type SCPs can be derived from coal combustion (e.g., Rose et al., 1994; 1996).

Murakami-Kitase et al. (2010) applied linear discriminant analysis to chemical compositions of particles with distinction of convolute type and non-convolute type particles deposited from the 1930s to 1990s in Osaka; they showed that the overall success rate of >90% of reference of morphologically classified material correctly assigned to two groups that are chemically different each other. However, we reexamined data of Murakami-Kitase et al. (2010) and found that most of particles from sediments of the 1990s are not properly assigned to two groups different chemically each other. Based on the data of Murakami-Kitase et al. (2010) and Hirakawa et al. (2011), we examined the data for the sediments deposited in Osaka, Tokyo and Nagasaki until the 1980s using linear discriminant analysis. The overall success rate of morphologically classified material correctly assigned to the two particle types is 88.8%, indicating relationship between surface morphology and chemical composition. Using the same discriminant function coefficients obtained from a linear discriminant analysis of convolute-type and non-convolute-type particles from the sediments in Osaka, Tokyo and Nagasaki until the 1980s, we calculate discriminant score of SCPs that are morphologically classified in samples analyzed in the present study (Table 2).The present-day SCPs in China have high success rates of >85% whereas those in Japan have lower success rates (55–80%), suggesting that a distinct relationship between surface morphology and composition in Chinese SCPs but not in Japanese ones. We do not explain why the relationship between surface morphology and chemical composition changes at the 1990s in Japan. In modern days, combustion technology development (e.g., use of multiple fuels) makes more difficult to identify their source fuel type, although in parts of the world (e.g., China) single fuels are used extensively (Rose, 2001). At least in Japan, for SCPs emitted to the atmosphere after the 1990s, their surface morphology is not used as a proxy of past industrial activities, and that is probably connected to the combustion technology development.

The SCPs from different countries plotted in different fields in the S/Si vs. Ti/Si diagram (Fig.2); Japanese particles are characterized by more S and less Ti, whereas Chinese ones are characterized by more Ti and less S. Ti is one of the major elements of coal and has been used as an indicative of coal particles (e.g., Ganor et al., 1988), and some of Ti is included in Chinese coal and their fly ashes (e.g., Cao et al., 2008; Ma et al., 2010). In the thermal station Ti is used as $TiO_2$ in many instruments (e.g., pipe, chimney pipe and steam condenser) and also catalyst for $NO_X$ removal (e.g., Kusamichi et al., 1996). Miller and Linak (2002) imply that the chemical composition of fly ashes is dependent on the mineral composition of coal and boiler operation conditions. Therefore, high percentage of Ti in particles in China may be derived from concentration of the element in coal or addition of the one during the process of the particles production. On the contrary, in Japan many of particles with more S were found. Rose et al. (1996, 1999) and Alliksaar et al. (1998) showed that the proportion of S in fly ash samples collected directly from oil-fired power plants are higher than that from coal-fired power plants. Most particles with high S found in Japan are possible to be derived from oil. As a result, difference in chemical composition between SCPs in Japan and China may be derived mainly from the difference in the source fuel of SCPs in Japan and China.

In order to examine whether chemical composition of SCPs in Japan and China have meaningful statistical difference, linear discriminant analysis was used based on the 18 chemical compositions (Fig.3). The linear discriminant function calculated in this study provided an effective separation of the two groups, with only 17 particles (8%) of derived from Japan having discriminant scores greater than the discriminant index and 15 particles (9%) of derived from China having scores less than the index. The overall success rates of 91.4% for the correct assignment of chemically classified material to the two groups indicates that the chemical composition of particles in Japan and China have meaningful statistical difference.

## Conclusion

We examined the surface morphology and the chemical composition of SCPs in surface sediment samples collected from industrial cities in Japan and China using SEM-EDS. Regardless their surface morphologies, SCPs can be classified into two types of SCPs emitted from Japan and China, using S/Si vs. Ti/Si diagram. The difference in chemical composition between SCPs in Japan and China may be derived mainly from the difference in the source fuel of SCPs in Japan and China; oil in Japan and coal in China. Since main fossil fuel in industrial countries in East Asia, such as South Korea and Taiwan, is oil, and then oil-derived high-S/low-Ti SCPs would be emitted from the power stations and other industries in these countries. Therefore, EDS analysis of SCPs would be effective method to identify their sources in East Asia, in particular China-derived particles.

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## References

Alliksaar T, Hörstedt P, & Renberg I (1998) Characteristic fly-ash particles from oil-shale combustion found in lake sediments. Water, Air, & Soil Pollution **104**, 149–160.

Ganor E, Altshuller S, Foner HA, Brenner S, & Gabbay J (1988) Vanadium and nickel in dustfall as indicators of power plant pollution. Water, Air, & Soil Pollution **43**, 241-252.

Cao Dz, Selic E, & Herbell JD (2008) Utilization of fly ash from coal-fired power plants in China. Journal of Zhejiang University SCIENCE A, 2008 **9**, 681-687.

Griffin JJ, & Goldberg ED (1979) Morphologies and origin of elemental carbon in the environment. Science **206**, 563-565.

Griffin JJ, & Goldberg ED (1981) Sphericity as a characteristic of solids from fossil fuel burning in a Lake Michigan sediment. Geochimica et Cosmochimica Acta **45**, 763-769.

Hirakawa E, Murakami-Kitase A, Okudaira T, Inoue J, Yamazaki H, & Yoshikawa S (2011) The spatial and temporal distributions of spheroidal carbonaceous particles from sediment core samples from industrial cities in Japan and China. Environmental Earth Science. doi: 10.1007/s12665-001-0907-1

Kusamichi H, Iseki J, Moriya A, Kanai A, Nishimura T, Kanayama H, & Kusamichi T (1996)Titanium industry in Japan and the new technology. Agune Gijyutu Center, Tokyo, 196pp.

Larsen J (2003) Size distribution and concentration of spheroidal carbonaceous fly-ash particles (SCPs) in lake sediments as an aid to detecting locally deposited atmospheric pollution. Water, Air, & Soil Pollution **149**, 163-175.

MaCj, Kim Jh, Kim Kh, Tohno S, & Kasahara M (2010) Specification of Chemical Properites of Feed Coal and Bottom Ash Collected at a Coal-fired Power Plant. Asian Journal of Atmospheric Environment Vol.4-2, 80-88.

Miller CA, & Linak WP (2002) Primary particles generated by the combustion of heavy fuel oil and coal. National Risk Management Research Laboratory, Research Triangle Park, 50pp.

Murakami-Kitase A, Okudaira T, & Inoue J (2010) Relationship between surface morphology and chemical composition of spheroidal carbonaceous particles within sediment core samples recovered from Osaka Bay Japan. Environmental Earth Science **59**, 1723-1729.

Rose NL (1995) The carbonaceous particle record in lake sediments from the Arctic and other remote areas of the northern hemisohere. Science of The Total Environment **160/161**, 487-496.

Rose NL (2001) Fly-ash particles, Tracking Environmental Change Using Lake Sediments, Physical and Geochemical Methods. Kluwer Academic Publishers, 319-349.

Rose NL, Juggins S, Battarbee RW (1994) Fuel type characterization of spheroidal carbonaceous particles using surface chemistry. Ambio **23**, 296-299.

Rose NL, Juggins S, Watt J (1996) Fuel-type characterization of carbonaceous fly-ash particles using EDS-derived surface chemistries and its application to particles extracted from lake sediments. Proceedings of the Royal Society of London A **452**, 881-907.

Rose NL, Juggins S, Watt J (1999) The characterisation of carbonaceous fly-ash particles from major European fossil-fuel types and applications to environmental samples. Atmospheric Environment **33**, 2699–2713.

## Figure Captions

Figure 1 Locations of the sediment core samples recovered from industrial cities in Japan (Chiba, Tokyo, Osaka, Yokkaichi and Nagasaki indicated by black circles) and China (Beijing, Tianjin, Qingdao, Nanjing, and Shanghai indicated by gray circles).

Figure 2 S/Si-Ti/Si of SCPs extracted from sediments in Japanese cities (black circles) and Chinese cities (gray triangles) plotted on logarithmic scales.

Figure 3 Distribution of SCPs extracted from sediments in Japanese cities (upper) and Chinese cities (lower) on a standardized linear discriminant function. The discriminant index largely separated the two groups of SCPs in Japan (score<0) and China (score>0).

Table 1 Summary of description of the surface sediment samples from Japanese and Chinese cities with number of SCPs observed and measured using SEM (scanning electron microscope) with EDS (energy-dispersive spectroscopy).

Table 2 Number and percentage of each morphological type of SCPs in Japanese and Chinese cities. Success rate means the percentage of morphologically classified material correctly assigned to convolute-type and non-convolute-type, calculated from chemical compositions, using the discriminant function coefficients obtained from a linear discriminant analysis of convolute-type and non-convolute-type particles from the sediments in Osaka, Tokyo and Nagasaki until the 1980s (Murakami-Kitase et al., 2010; Hirakawa et al., 2011).

Table 3 Average chemical compositions with standard deviations (parenthetic values) of SCPs from Japanese and Chinese cities.

Figure 1 (Momose et al)

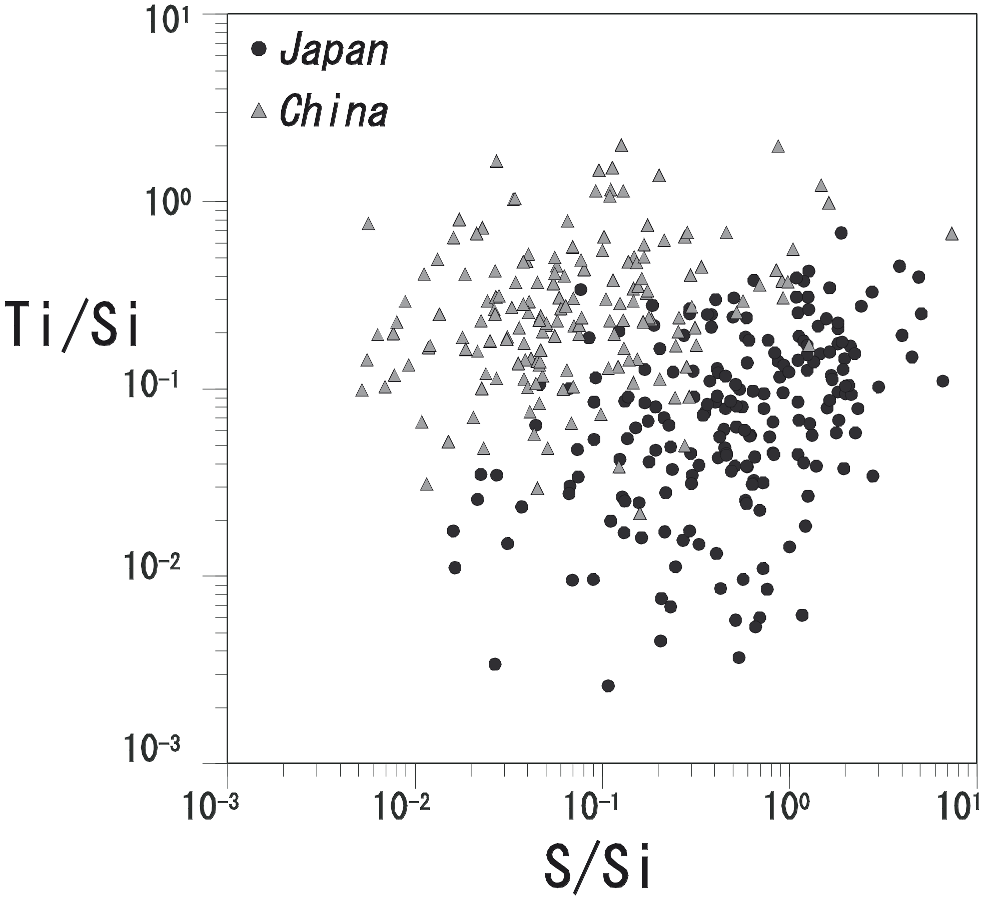

Figure 2 (Momose et al)

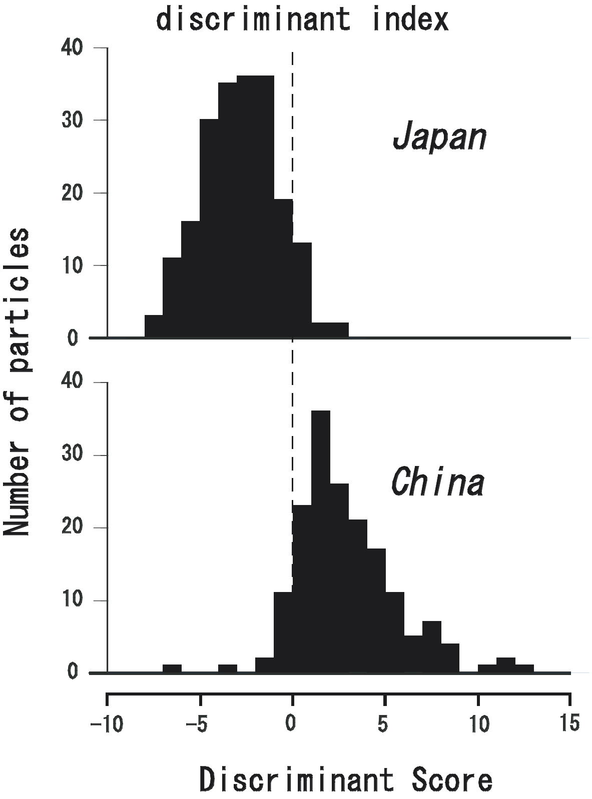

Figure 3 (Momose et al)

Table1 Summary of description of the sediment core samples from the different locations.

| | Area | Locality | Sampling date (D.C.) | Lithology | Number of measured particle |
|---|---|---|---|---|---|
| Chiba<br>(Lake Inba) | $1.2\times10\mathrm{km}^2$ | N35°48'09" E140°15'05" | 2007 | Clay | 44 |
| Tokyo<br>(Koike Pond) | $1.0\times10^{-3}\mathrm{km}^2$ | N35°35'55" E139°41'51" | 2007 | Sand with silt | 32 |
| Yokkaichi<br>(Pond) | $2.0\times10^{-3}\mathrm{km}^2$ | N35°03'56" E136°39'05" | 2008 | Silt | 45 |
| Osaka<br>(Osaka Bay) | $1.4\times10^{3}\ \mathrm{km}^2$ | N34°37'32" E135°21'23" | 2005 | Silty clay | 44 |
| Nagasaki<br>(Nagasaki Bay) | $1.1\times10\mathrm{km}^2$ | N32°44'34" E129°51'43" | 2007 | Sand with silt | 36 |
| Beijing<br>(Huairou Shuiku) | $6.3\mathrm{km}^2$ | N40°18'18" E116°36'39" | 2008 | Silty clay | 19<br>（reffered to Hirakawa et al., 2011) |
| Tianjin<br>(Qi Li Hai) | $1.9\mathrm{km}^2$ | N39°18'14" E117°33'30" | 2006 | Sand with silt | 30 |
| Tianjin<br>(Tuanbowa Shuiku) | $4.7\times10\mathrm{km}^2$ | N38°55'28" E117°08'30" | 2006 | Sand with silt | 34 |
| Qingdao<br>(Xiashan Shuiku) | $9.4\times10\mathrm{km}^2$ | N36°29'44" E119°24'38" | 2008 | Clay | 30 |
| Nanjing<br>(Gaoyou Hu) | $4.7\times10^{2}\mathrm{km}^2$ | N32°48'18" E119°24'04" | 2008 | Clay | 26 |
| Shanghai<br>(Chang Dang Hu) | $8.5\times10\mathrm{km}^2$ | N31°36'47" E119°31'01" | 2008 | Silty clay | 26 |

Table2 The ratio of surface morphology of SCPs in each city.

| | non−convolute | | convolute | | other | | success rate | |
|---|---|---|---|---|---|---|---|---|
| | % | number of particles | % | number of particles | % | number of particles | % | |
| Chiba | 56.8 | 25 | 22.7 | 10 | 20.5 | 9 | 65.7 | (23/35) |
| Tokyo | 51.4 | 19 | 29.7 | 11 | 18.9 | 7 | 76.7 | (23/30) |
| Yokkaichi | 46.5 | 20 | 32.6 | 14 | 20.9 | 9 | 55.9 | (19/34) |
| Osaka | 50.0 | 22 | 36.4 | 16 | 13.6 | 6 | 76.3 | (29/38) |
| Nagasaki | 80.6 | 29 | 16.7 | 6 | 2.8 | 1 | 71.4 | (25/35) |
| Beijing | 100.0 | 19 | 0.0 | 0 | 0.0 | 0 | 100.0 | (19/19) |
| Tianjin(Qi Li Hai) | 86.7 | 26 | 3.3 | 1 | 10.0 | 3 | 96.3 | (26/27) |
| Tianjin(Tuanbowa Shuiku) | 65.7 | 23 | 8.6 | 3 | 25.7 | 9 | 88.5 | (23/26) |
| Qingdao | 86.7 | 26 | 0.0 | 0 | 13.3 | 4 | 100.0 | (26/26) |
| Nanjing | 75.0 | 21 | 0.0 | 0 | 25.0 | 7 | 100.0 | (21/21) |
| Shanghai | 63.0 | 17 | 11.1 | 3 | 25.9 | 7 | 90.0 | (18/20) |
| **All of Japanese cities** | **56.7** | **115** | **28.1** | **57** | **15.3** | **31** | **69.2** | **(119/172)** |
| **All of Chinese cities** | **78.1** | **132** | **4.1** | **7** | **17.8** | **30** | **95.7** | **(133/139)** |

Table3 Average chemical compositions and standard deviations of SCPs

| | Na | Mg | Al | Si | P | S | Cl | K | Ca | Ti | V | Cr | Mn | Fe | Co | Ni | Cu | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Chiba | 9.52 (2.10) | 3.05 (0.73) | 2.73 (0.68) | 41.14 (12.74) | 1.34 (0.86) | 25.38 (14.68) | 1.75 (1.75) | 0.98 (0.44) | 5.50 (1.56) | 1.85 (0.98) | 0.71 (0.39) | 0.41 (0.30) | 0.40 (0.26) | 0.71 (0.42) | 0.66 (0.46) | 1.18 (0.66) | 1.20 (0.67) | 1.49 (0.72) |
| Tokyo | 13.27 (3.79) | 3.22 (0.92) | 2.93 (2.01) | 37.54 (15.09) | 0.54 (0.31) | 21.41 (14.19) | 7.47 (8.64) | 1.46 (0.70) | 5.52 (1.68) | 0.47 (0.28) | 0.62 (0.32) | 0.43 (0.29) | 0.46 (0.28) | 0.71 (0.34) | 0.65 (0.36) | 0.97 (0.40) | 0.97 (0.39) | 1.37 (0.40) |
| Yokkaichi | 10.64 (2.36) | 3.14 (0.87) | 2.68 (0.80) | 39.82 (12.14) | 1.13 (0.86) | 19.70 (12.78) | 3.52 (2.32) | 1.50 (0.89) | 5.85 (1.56) | 4.64 (2.68) | 0.76 (0.43) | 0.54 (0.50) | 0.49 (0.26) | 1.02 (0.55) | 0.79 (0.31) | 1.18 (0.45) | 1.11 (0.48) | 1.50 (0.58) |
| Osaka | 9.36 (3.18) | 2.39 (0.81) | 3.64 (3.26) | 29.92 (11.42) | 1.40 (0.84) | 30.30 (13.21) | 5.60 (6.97) | 1.64 (1.60) | 4.72 (1.45) | 4.26 (2.42) | 0.84 (0.44) | 0.43 (0.24) | 0.45 (0.21) | 0.84 (0.26) | 0.62 (0.29) | 1.10 (0.44) | 1.03 (0.35) | 1.46 (0.53) |
| Nagasaki | 9.02 (2.39) | 3.05 (0.85) | 2.71 (1.28) | 37.54 (13.06) | 1.09 (0.56) | 24.54 (17.87) | 4.30 (4.60) | 1.16 (0.59) | 5.22 (1.67) | 3.22 (2.88) | 0.86 (0.43) | 0.53 (0.31) | 0.58 (0.35) | 1.16 (0.68) | 0.82 (0.49) | 1.28 (0.52) | 1.29 (0.56) | 1.64 (0.64) |
| Beijing | 10.42 (1.65) | 8.59 (2.10) | 14.91 (4.72) | 38.93 (8.10) | 0.69 (1.14) | 5.02 (2.54) | 3.40 (1.03) | 2.59 (0.57) | 7.55 (1.66) | 3.16 (1.25) | 0.42 (0.27) | 0.40 (0.30) | 0.45 (0.30) | 0.94 (0.31) | 0.46 (0.32) | 0.65 (0.34) | 0.66 (0.29) | 0.77 (0.29) |
| Tianjin(Qi Li Hai) | 9.22 (2.06) | 2.95 (0.71) | 4.87 (1.23) | 40.82 (8.94) | 1.53 (1.17) | 6.28 (8.04) | 3.51 (2.44) | 1.25 (0.60) | 5.41 (1.04) | 17.41 (5.66) | 0.48 (0.37) | 0.36 (0.24) | 0.40 (0.31) | 1.06 (0.39) | 0.72 (0.33) | 1.06 (0.41) | 1.18 (0.41) | 1.51 (0.44) |
| Tianjin((Tuanbowa Shuiku)) | 10.52 (2.29) | 3.47 (0.55) | 5.23 (2.29) | 47.01 (8.30) | 1.47 (1.33) | 3.10 (2.35) | 4.07 (3.57) | 1.51 (0.50) | 6.53 (1.35) | 8.89 (4.76) | 0.60 (0.31) | 0.56 (0.37) | 0.57 (0.31) | 1.23 (0.53) | 0.80 (0.39) | 1.46 (0.60) | 1.31 (0.54) | 1.67 (0.75) |
| Qingdao | 11.90 (1.34) | 3.64 (0.83) | 3.57 (0.84) | 44.73 (10.53) | 1.06 (0.62) | 8.34 (9.72) | 3.42 (2.63) | 1.42 (0.33) | 6.42 (1.39) | 7.67 (2.44) | 0.60 (0.32) | 0.53 (0.31) | 0.58 (0.30) | 1.12 (0.50) | 0.87 (0.49) | 1.20 (0.63) | 1.22 (0.53) | 1.70 (0.61) |
| Nanjing | 9.88 (3.14) | 3.07 (0.95) | 4.51 (2.11) | 44.16 (12.07) | 1.34 (0.85) | 3.18 (4.98) | 1.52 (1.65) | 0.95 (0.46) | 5.36 (1.69) | 21.64 (12.53) | 0.21 (0.20) | 0.26 (0.27) | 0.25 (0.20) | 0.68 (0.46) | 0.44 (0.30) | 0.63 (0.35) | 0.79 (0.35) | 1.13 (0.44) |
| Changhai | 10.98 (3.34) | 3.33 (0.95) | 4.47 (1.75) | 40.65 (9.26) | 1.54 (0.84) | 9.79 (12.45) | 3.47 (3.58) | 1.39 (0.96) | 5.20 (1.18) | 12.04 (5.75) | 0.55 (0.45) | 0.37 (0.28) | 0.41 (0.30) | 1.04 (0.37) | 0.76 (0.38) | 1.05 (0.50) | 1.30 (0.52) | 1.65 (0.63) |
| **All of Japanese cities** | **10.31 (3.19)** | **2.96 (0.89)** | **2.95 (1.92)** | **37.15 (13.48)** | **1.12 (0.80)** | **24.38 (15.01)** | **4.44 (5.76)** | **1.35 (0.98)** | **5.36 (1.63)** | **2.95 (2.61)** | **0.76 (0.41)** | **0.46 (0.34)** | **0.47 (0.28)** | **0.88 (0.50)** | **0.70 (0.40)** | **1.14 (0.51)** | **1.12 (0.52)** | **1.49 (0.59)** |
| **All of Chinese cities** | **10.87 (2.90)** | **3.40 (0.85)** | **4.62 (2.06)** | **43.95 (10.29)** | **1.34 (1.05)** | **5.57 (7.94)** | **3.63 (3.57)** | **1.41 (0.69)** | **5.95 (1.46)** | **11.96 (8.71)** | **0.52 (0.38)** | **0.48 (0.40)** | **0.50 (0.35)** | **1.13 (0.70)** | **0.78 (0.44)** | **1.12 (0.59)** | **1.22 (0.57)** | **1.57 (0.67)** |