The use of size distributions of spheroidal carbonaceous particles in swimming pool deposits for evaluating atmospheric particle behaviour

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ABSTRACT

To clarify the relationship between the transport distance of spheroidal carbonaceous particles (SCPs) and particle size, we investigated the spatial distribution of SCP sizes in swimming pool deposits in the central Osaka Plain, central Japan. Median particle size of SCPs generally decreases with distance (0 to ~20 km) downwind from the local coastal industrial area where SCP sources are distributed widely. This suggests that most SCPs found in the study area are derived from the industrial area. Samples with >40% of particles >20 μm were predominantly collected within 2 km of the industrial area, while samples with >40% of particles <10 μm were mostly collected over 10 km from the industrial area. Based on the results of our study and previous studies, we conclude that a higher proportion of particles of size >20 μm indicates that the origin of SCPs is within a few kilometres upwind of the sample site, whereas the presence of higher proportion of particles <10 μm indicates that their source is generally further than 10 km upwind. However, other factors may affect the size distribution of SCPs at a given location (e.g., fuel type, quality of the particle precipitator and topography of the terrain). Pool deposits provide more suitable samples than lake sediments for investigating atmospheric precipitation.

Keywords: Spheroidal carbonaceous particle (SCP) · Particle size · Swimming pool deposit · Osaka Plain · Particle behaviour

1. Introduction

Spheroidal carbonaceous particles (SCPs) are produced by high-temperature combustion of fossil fuels and emitted into the atmosphere. As SCPs are not produced from burning wood, biomass, or charcoal, they are unambiguous indicators of atmospheric deposition from industrial fossil fuel combustion. Under favourable conditions, SCPs can also be preserved in sediments. Therefore, SCPs in sediments have been used for monitoring atmospheric deposition of pollutants derived from fossil fuel combustion (e.g., Renberg and Wik, 1985; Kreiser et al., 1990; Flower et al., 1995; Wik and Renberg, 1996; Rose et al., 1998, 1999, 2004; Larsen, 2000; Nagafuchi et al., 2009; Hirakawa et al., 2011).

SCP concentrations in surface sediments have usually been examined to evaluate the regional spatial distribution of pollution (e.g., Wik and Renberg, 1991; Rose and Juggins, 1994; Alliksaar and Punning, 1998; Fott et al., 1998; Rose and Harlock, 1998; Rose et al., 1999). These studies showed that SCP distribution patterns are similar to those of other air pollutants. For example, Rose and Juggins (1994) showed that the pattern of SCP concentrations in the surface sediment of 146 lakes in Scotland is similar to that of modelled sulphur deposition. However, SCP concentrations do not reveal information about the emission source of the pollution; i.e., whether SCPs originate predominantly from local or distant sources (Vukic et al., 2006). Previous studies have investigated the relationship between the SCP size distribution and transportation distance from their source (Fott et al., 1998; Larsen, 2000, 2003; Vukic et al., 2006). Based on these studies, the deposition of SCPs seems to be primarily dependent on distance from the emission source and wind direction, although other factors (e.g., fuel type, boiler condition, and quality of particle retention equipment) may affect the SCP size distribution.

Previous studies used surficial lake sediments to investigate modern SCP size distributions. However, in most cases, the depositional age of these sediments is ambiguous, and the number of viable study sites in a given area is limited. To provide a detailed evaluation of the relationship between the SCP size distribution and distance from the source, SCPs deposited over a certain period should be examined at a number of sites close to the source.

We examined deposits mainly composed of atmospheric precipitation on the floor of 29 swimming pools located within several kilometres of each other to investigate the spatial distribution of SCP sizes on the central Osaka Plain, central Japan, where a large coastal industrial area is located. These pools act as reserves of atmospheric precipitation over the previous year, as they are cleaned annually. Swimming pools provide ideal samples for evaluating SCP size distribution, as they have no inflow of particles or clastics from external sources, with atmospheric precipitation being the only source of deposits. This differs from lake sediments, which are composed of atmospheric precipitation and clastics supplied from their watershed. Unlike lake sediments, the size distribution of SCPs in swimming pools therefore reflects the true size distribution at the sampling location. Furthermore, many swimming pools in schools are located near industrial areas in central Japan, providing many sampling sites from which the atmospheric precipitation distribution can be evaluated in detail.

2. Study Sites and Materials

Osaka Plain is located adjacent to Osaka Bay (Fig. 1), and is one of the largest plains in Japan. It is largely flat (<30 m elevation) and is surrounded by hills of approximately 200 m in elevation. In central Osaka Plain where we collected samples, precipitation in 2011 was 1614 mm at the Osaka meteorological station and 1444 mm at the Sakai meteorological station. Meteorological data show that the dominant wind directions are NEN to E and WSW to W. The monthly average wind speed in the central Osaka Plain was 2–3 m/s for the study period

(May 2011 to April 2012).

The coastal industrial area (Hanshin Coastal Industrial Belt) is mainly located on reclaimed land at the western end of the plain, and is one of the largest industrial areas in Japan. Heavy chemical industries have been located in the area since the beginning of the 20th century. Amounts of coal and oil consumption for industry in Osaka Prefecture at A.D. 2010 (most of that is consumed in the coastal industrial area) are 15,000 t and 2,950,000 kl respectively (see the Japanese Agency for Natural Resources and Energy Website, http://www.enecho.meti.go.jp), indicating oil is main fuel in this area. This area is likely to be the main source of SCPs that settle on the Osaka Plain, as the next closest large industrial area is at least 100 km away.

Potential sources of SCPs in this area are mainly oil refinery and iron-steel factory that combust fossil- fuel at high temperature (Fig.1). Larsen (2000) suggested oil refinery is possible to become major source of SCPs. These SCP sources are scattered in this area (Fig.1). Oil refinery probably emits oil-derived particles, whereas iron-steel factory probably emits particles derived from combustion of coal. Other potential main source is the incinerator plant that combusts trash with oil (Fig.1). Although two thermal stations are located along the coast, they use natural gas and never emit SCPs (Rose, 2001).

To evaluate the SCP distribution under certain wind conditions, we collected samples from deposits on the floor of water-filled swimming pools in 29 elementary and junior high schools located within several kilometres of each other (Fig. 1). These sites are located to the east of the coastal industrial area. Each pool is 25 m × 10 m in size, and water depth is generally 1.0–1.5 m. In most of the pools, the base gently slopes toward the centre of the pool. Samples of deposits, along with 4 l of water, were collected using a conventional pump from late April to May 2012. A sample was collected from each pool at the deepest point in the centre of the pool. Most samples consisted of deposited and floating mud and algae.

3. Methods

After mixing each 4 l sample well, we took a subsample of 200 ml for analysis. To remove algae, we added 200 ml of 30% H_2O_2 to each sample, which we then heated at 70–80 °C for 1 hour. We repeated this process twice and left the samples overnight. We then washed each sample through a 125 μm mesh and a 5 μm nitrocellulose filter to remove coarse and fine particles. We collected particles of sizes 5–125 μm with water, and mixed the samples well. A drop of the sample was placed on the 5 μm filter. We then dried the residue on the filter at 50 °C.

We observed particles on the filter at 200× and 500× magnification under incident light using an optical microscope. Referring to Rose (2008), we restricted SCP selection to black, rounded particles with many pores and a characteristic glossy appearance. We measured the diameter of 100–110 particles in each sample under 500× magnification using a CCD camera and NIH Image software for Macintosh (developed by the U.S. National Institutes of Health and available at http://rsb.info.nih.gov/nih-image/).

4. Results and discussion

4.1. SCP size distribution and particle behaviour in central Osaka Plain

Median particle diameter (size) of the SCPs in each sample is shown in Fig. 2. Figure 3 shows variations in median SCP size along a west-to-east transect. The median SCP size tends to become smaller from west to east with increasing distance from the coastal industrial area. The SCPs with the largest median size of 25.4 μm are found at the site on the reclaimed land, surrounded by the industrial area. The SCPs with the smallest median size of 9.0 μm are found 12 km away from the industrial area. Median SCP sizes of over 17 μm are limited to the area within 5 km of the industrial area. These results indicate that the size of SCPs that settle primarily depends on distance from the emission source. To clarify the relationship between particle size and distance from the industrial area, the study area was divided into zones of 0–2, 2–5, 5–10, and >10 km from the industrial area (Fig.1). Because main potential SCP sources are scattered in the industrial area, distance from the industrial area is almost corresponding to distance from the main potential sources. Figure 4 shows the 10th, 25th, 50th, 75th and 90th percentiles of particle size in each zone. In each percentile examined, particle size decreases with increasing distance from the industrial area. For example, the 50th percentile (median size) of particle size is 17.4 μm at sites located 0–2 km from the industrial area, 14.7 μm at 2–5 km, 12.3 μm at 5–10 km, and 11.5 μm at >10 km. In the zones at 0–2, 2–5, and 5–10 km from the industrial area, at least 25% of particles are over 20 μm, whereas in the zone at >10 km, nearly 80% of particles are under 20 μm. Referring to Vukic et al. (2006), we made histograms of size distribution, divided into four size ranges of 5–10, 10–20, 20–40, and >40 μm (Fig. 5). The variation in the SCP size distribution with distance was most striking for two particle size categories: particles >20 μm dominate the 0–2 km zone (43%) and decrease to 22% of total particles at >10 km from the industrial area; and particles <10 μm contribute a low proportion (19%) of particles found in the 0–2 km zone, but increase to 43% at >10 km from the industrial area. These results indicate that the distribution patterns of particles of sizes >20 and <10 μm are significantly different from each other, probably reflecting a difference in atmospheric particle behaviour.

4.2. Comparison of SCP size distributions with previous studies

The size distribution of SCPs settling on a site depends on the original size of particles emitted and enviromental factors such as distance from the source, wind velocity, wind direction, and topography of the terrain. The original particle size depends on the type and quality of fuel burnt, combustion parameters, and the type (e.g., oil, coal, or shale oil) and quality of the particle precipitator (Vukic et al., 2006).

Larsen (2003) examined the size distributions and concentrations of SCPs in surface sediments from 20 lakes along a 62 km west–east transect in the Bergen area of Norway. The study covered an area with elevation between approximately 100 and 400 m, including lakes located up to 300 m higher than the city area. The main fuel burnt in Norway is oil, similar to that in Japan. Larsen (2003) showed that large particles (>10 μm) contribute more than 50% of the SCP size distribution in lakes downwind and close to the city (up to 20 km away). In contrast, further downwind (up to 40 km from the city), smaller particles (5–10 μm) constitute over 50% of SCPs. In our study sites, located within approximately 20 km of the SCP source, large particles (>10 μm) contribute >50% in 27 of the 29 samples. In this aspect, our results are consistent with those presented by Larsen (2003). However, Larsen (2003) observed a smaller SCP size close to the source than we measured in our study. This is possibly due to the location of lakes at a higher elevation than the SCP source in Bergen, as large particles are difficult to suspend for the long periods required for transport to higher elevations (e.g., Pye, 1987), and are therefore less likely to be present in these lake sediments.

Vukic et al. (2006) analysed the size distribution of SCPs in the surface sediments of six water bodies under various conditions in the Czech Republic. In this case, SCPs were derived from burning brown coal, and the particle retention equipment was in poor condition, possibly producing larger particle sizes at the source. The authors suggested that the presence of more than 20% of particles with a diameter exceeding 40 μm in the sediments indicated a local origin of SCPs. Our study recorded only one sample with more than 20% of particles over 40 μm, found within a few kilometres of the industrial area. Other samples from our study may not have reached this criterion despite close proximity to the SCP source, as the original particle size was smaller than that reported by Vukic et al. (2006). Vukic et al. (2006) also found that the SCP size distribution correlates well with distance from the particle source. In the local area, average SCP diameter was over 30 μm (corresponding to a median SCP diameter of over 22 μm), decreasing to 17–23 μm (median diameter of 13–19 μm) in polluted areas not directly affected by local sources, and less than 15 μm (median diameter of less than 12 μm) in a relatively clean region away from the source. Our study is generally consistent with this result, though the overall particle sizes are smaller, with median SCP diameters >17 μm predominantly found close to the source and median diameters of <11 μm predominantly located over 10 km from the source. Those smaller particles in Osaka Plain are probably due to the fuel type (oil being the main fossil fuel burnt in Japan) and Japanese particle retention equipment removing more large particles than in Czech one doing.

Hirakawa et al. (2011) and Momose et al. (2012) evaluated the surface morphology and chemical composition of SCPs (mainly ~20 μm in size) in surface sediments and sediment core samples recovered from industrial cities in Japan and China. The cities are located at least 50 km from each other. The morphological and chemical characteristics of SCPs differ significantly between the cities, even within the same country. This suggests that most SCPs of ~20 μm in sediments from industrial cities are derived from local combustion, provided that the city is located at least 50 km from other cities. Furthermore, this indicates that SCPs of this size are unable to be transported long distances, and are generally deposited within several tens of kilometres from their source. These results are consistent with the locations at which we found SCPs of this size.

Pye (1987) suggested that during aeolian transport, very small particles (<20 μm) are entrained by wind, then transported in suspension and kept aloft by turbulent eddies; in contrast, larger particles (20–70 μm) are transported via short-term suspension. This model is widely accepted (e.g., Sharifi et al., 1997; Fearnehough et al., 1998; Yang and Ding, 2008; Lancaster, 2009). Our results indicate that the behaviour of SCPs in the atmosphere is similar to that of other particles, where the mode of transport is changed at the particle size of 10-20 μm.

Based on results of our study and previous studies, we can conclude that where SCPs of >20 μm constitute more than 40% of total SCPs, the source is generally located within a few kilometres in the upwind direction. Furthermore, the presence of a high proportion (>40%) of SCPs <10 μm indicates that their source is at least 10 km upwind, regardless of the size of particles emitted. Although these findings cannot be applied to all cases, SCP size distributions may provide useful information for evaluating pollution sources.

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Figure captions

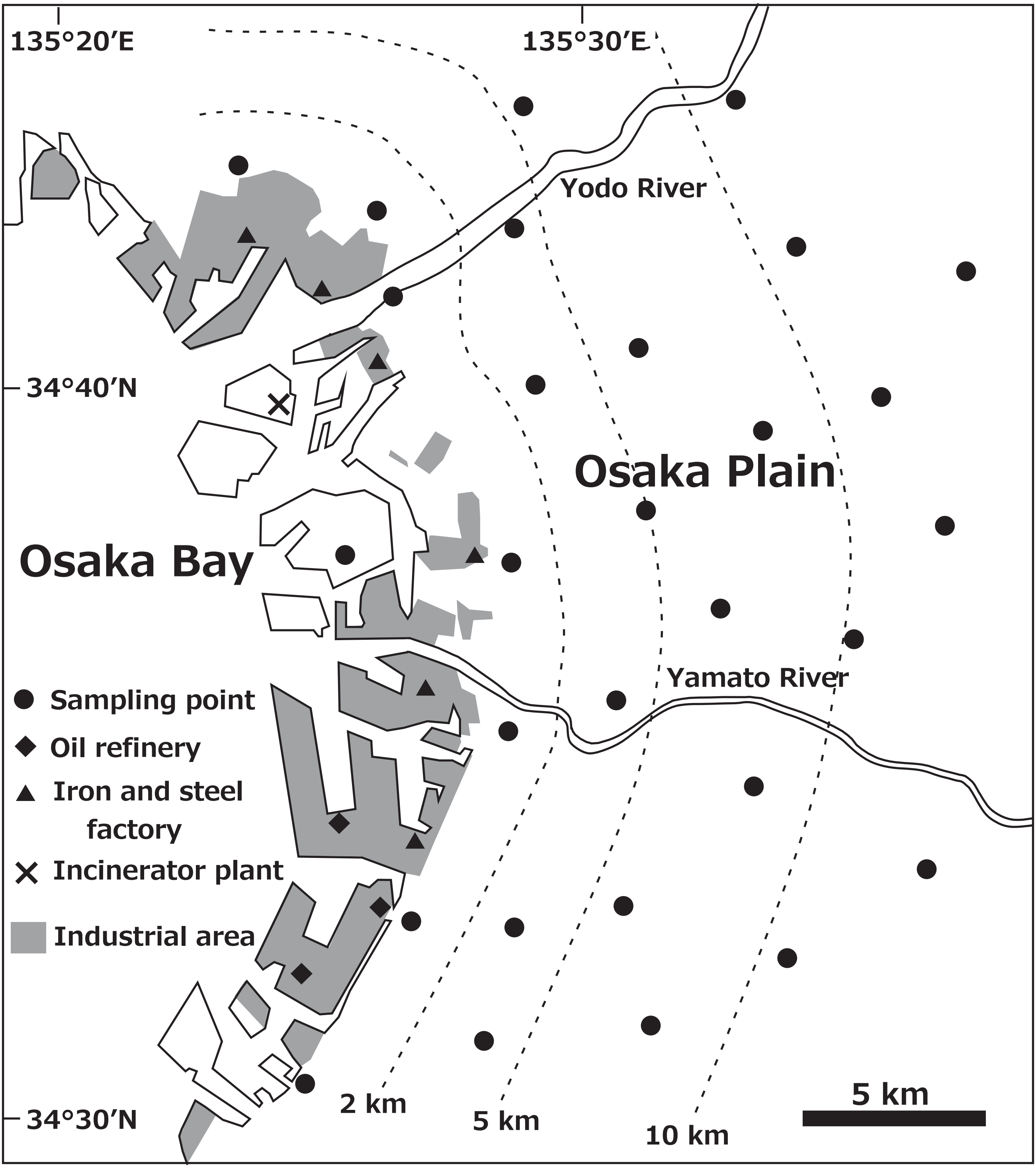
Fig. 1. Map showing sampling points, main potential SCP sources and coastal industrial area. Dotted lines indicate distance from coastal industrial area.

Fig. 2. Median SCP size in each sample. The size of circles indicates the actual SCP median diameter. The greyscale colour of each circle shows the median diameter in terms of four size ranges (<11, 11–14, 14–17, and >17 μm).

Fig. 3. Variations in median SCP size with distance from industrial area.

Fig. 4. The 10th, 25th, 50th, 75th and 90th percentile of particle size in each zone (0–2, 2–5, 5–10, and >10 km from the coastal industrial area).

Fig. 5. Histogram of SCP size distribution in each zone (0–2, 2–5, 5–10, and >10 km from the coastal industrial area) divided into four ranges (5–10, 10–20, 20–40, and >40 μm).


135°20′E
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Yodo River
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Osaka Plain
Osaka Bay
Yamato River
Sampling point
Oil refinery
Iron and steel factory
Incinerator plant
Industrial area
2 km
5 km
10 km
5 km
34°30′N


135°20′E
135°30′E
34°40′N
34°30′N
5 km
20μm
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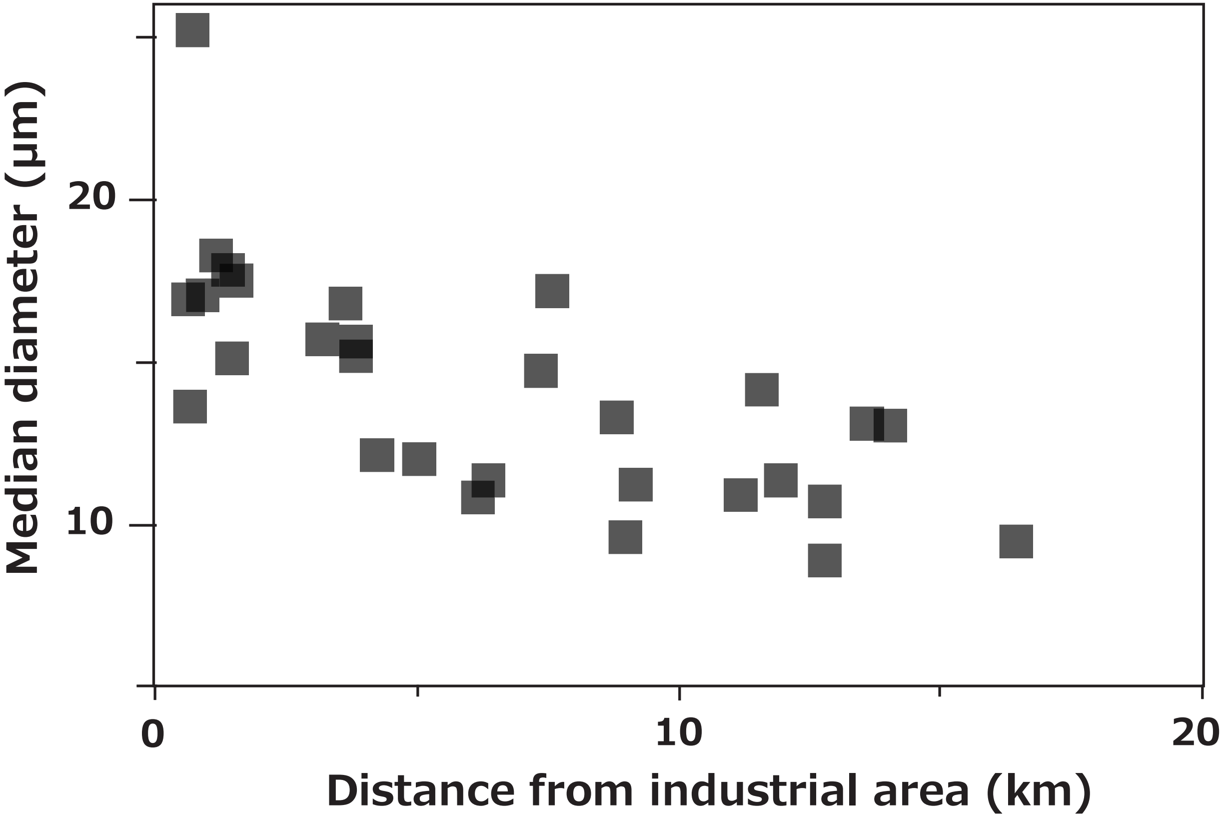
11 μm
14 μm
17 μm


Median diameter (μm)
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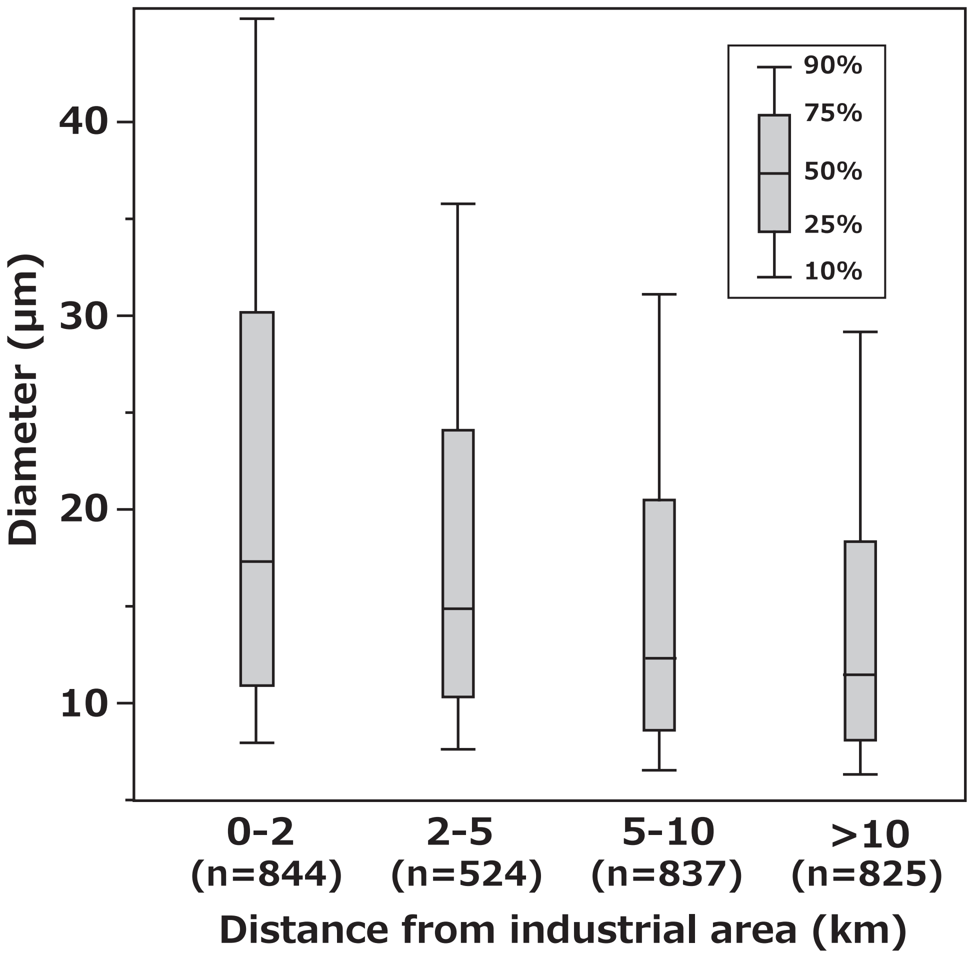
20
Distance from industrial area (km)


90%
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50%
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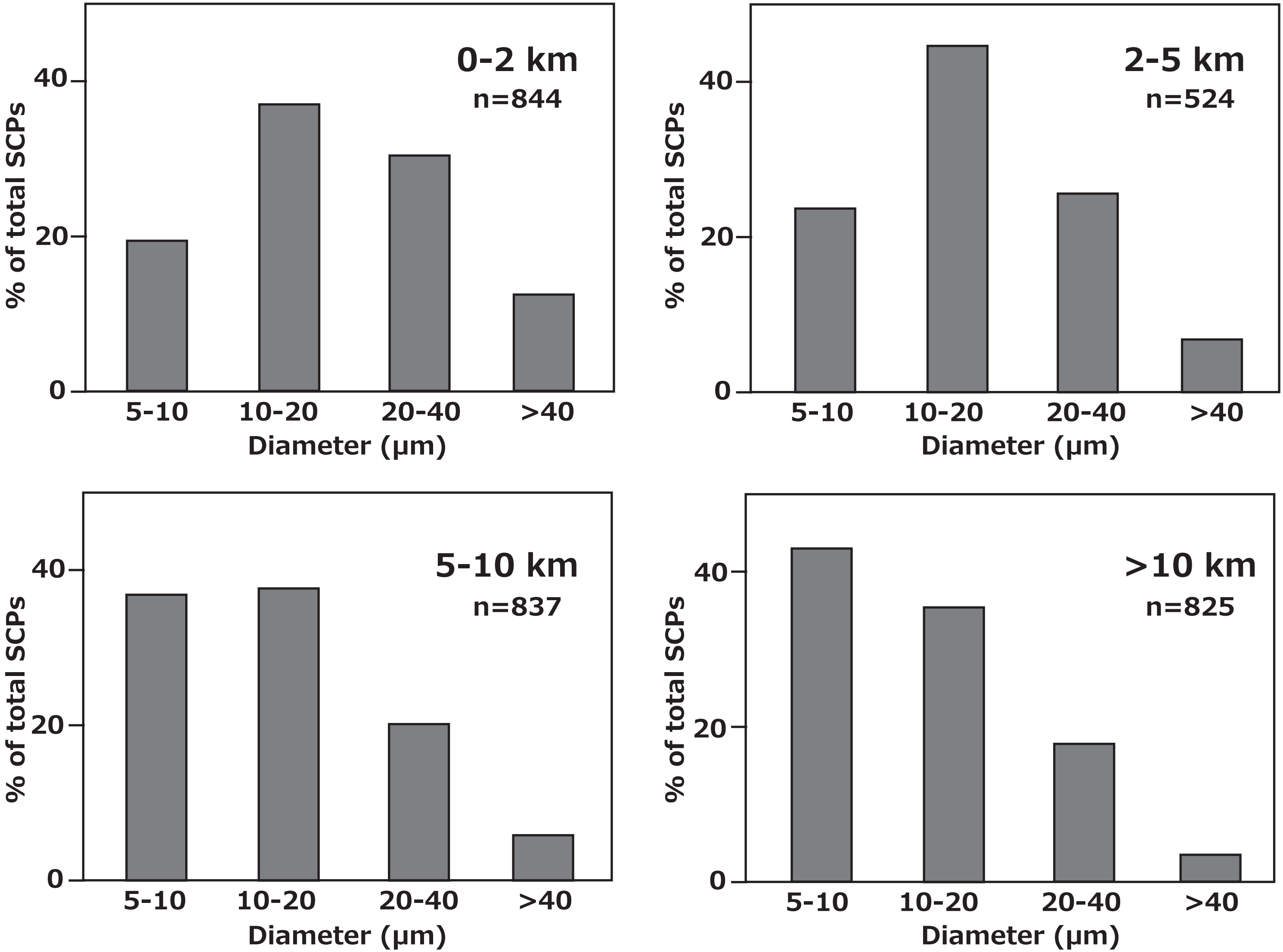
20
10
Diameter (μm)
0-2
(n=844)
2-5
(n=524)
5-10
(n=837)
>10
(n=825)
Distance from industrial area (km)


0-2 km
n=844
% of total SCPs
Diameter (μm)
5-10
10-20
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2-5 km
n=524
% of total SCPs
Diameter (μm)
5-10
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20-40
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5-10 km
n=837
% of total SCPs
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% of total SCPs
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