Aerosol and Air Quality Research, 15: 319–328, 2015

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2013.12.0360

Characterizing Particulate Pollutants in an Enclosed Museum in Shanghai, China

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ABSTRACT

The present study reported simultaneous measurements of particle number concentration (PNC), particle mass concentration, water soluble organic carbon (WSOC), organic carbon (OC), elemental carbon (EC), particulate morphology and main elemental properties. The measurements were conducted in a museum located in the downtown area of Shanghai. The impacts of storey, particle size and decorative flooring on particle characteristics inside museum were investigated. Size fractionated PNC in display halls belonging to the same storey exhibited similar daily variations. Particles within the range of 0.3–0.5 μm were identified as the most significant contributors to the overall size distribution measured, as ambient fine particles penetrated through the mechanical ventilation system. Multi-lognormal fitting OC concentrations can be assigned to the presence of tourists in the museum or originate from secondary OC formation. The latter can be estimated by comparing the relationship between OC and EC, as well as OC and WSOC.

Keywords: Element carbon; Organic carbon; Particulate matter; WSOC.

INTRODUCTION

Airborne particulate matter has become one of the most urgent issues in China. It not only has negative impacts to human health and the environment, but also influences the quality and durability of various materials. Under such circumstances, museum protection confronts the challenge of the sustainable management and inheritance. Under the circumstance of cultural relic being exposed to a low concentration of particulate matter or experienced only a short period of exposure, the damage on the cultural relic cannot be observed, but obvious effects such as surface alteration, color change or even weakening of the material may occur after a relative longer of time period (Vallero, 2007). A number of studies have concluded that PM can lead to soiling due to large particles (diameter is larger than 2.5 μm) that are abrasive. Large particles may also cause surface scratching (Brimblecombe, 1990; Phenix and Burnstock, 1990; Nazaroff, 1993; De Bock *et al.*, 1996). Particles within that size range are heavy enough to settle in a still air. On the other hand, small particles remain suspended in the air until being trapped or held down on certain surfaces by means of electrostatic attraction as well as other mechanisms, including turbulent diffusion, thermophoresis and gravitational settling caused by growing particles. Particles in the small size (diameter is less than 2.5 μm) span can enter display cases, deposit on objects, and thus soil the surface. In addition, more serious damage can occur if particles are further transformed by chemical reactions. These processes involve gases and lead to the formation of acidic components that are being deposited on particles, which will ultimately affect all acid-sensitive materials (Worobiec *et al.*, 2006). Normally such particles are hygroscopic enough to attract water and cause corrosion of metals. They may also contain traces of metals such as iron to speed up the deterioration of organic materials. High levels of salt crystals in the air affect coastal areas as well. Here salt absorbs water from the air, creating droplets of high salt concentrations (Cardell *et al.*, 2003). These droplets are then causing corrosion of most unprotected metals. In addition, new concrete emits alkaline particles as well (Kenjo, 1986). They darken oil paint films and decolour some dyes and pigments. The carbon rich component, is regarded as one of the most harmful and dangerous types for the preservation of valuable historical objects, which is due to their ability to stick easily to the surfaces as these component have strong adhesive properties (Cruz *et al.*, 2000). Therefore it is essential to document the spatial and temporal variation of particles and analyse its components to provide fundamental data for further investigations.

A limited number of studies on particulate pollutants

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inside museums have been conducted in several countries. A daytime and nighttime monitoring of total suspended PMs were conducted in Emperor Qin's Terra-Cotta Museum, China (Hu and Lee, 2009). They concluded that both size distributions and particle types were influenced by visitors significantly. The monitoring conducted in Plantin-Moretus Museum, Belgium was found that sulfur rich particles were frequently observed indoors during summer while calcium rich and calcium and-silicon rich particles were dominating during winter months (Gysels and Deutsch, 2002). In the museum in Goudi, urban area of Athens, it was also observed that resuspension led to an increase of fine and coarse particles by a factor of 30 and 80 times, respectively (Saraga *et al.*, 2011). Moreover, the measurement conducted in archaeological museum of Thessaloniki, N. Greece found that increasing $PM_{2.5}$ acidity existed indoors due to the presence of partially neutralized ammonium sulfate (Mouratidou and Samara, 2004). The study on single particle analysis showed that the relative abundance of carbon-rich particles inside the Royal Museum of Fine Arts, Belgium was greater than outside (Krupińska and Worobiec, 2012). In the Correr Museum, Italy, the majority of samples from two campaigns appeared to be composed of six to eight different particle types, from which calcium rich particles and the alumino silicates as well as organic material were the most important contributors (Bock *et al.*, 1996). The sampling in Royal Museum of Wawel Castle, Poland (Worobiec *et al.*, 2008) was concluded as well that the number of museum visitors was related to the increase of the particle concentration as well as the particle component.

In this study, measurements were conducted to characterize indoor PMs in the museum located in Shanghai Central Business District (CBD). Monitored parameters included particle number size distribution, daily variation of size-fractionated particle number, particle mass and morphological as well as elemental properties of PMs. In addition, the correlation between OC and WSOC was discussed aiming to quantify the source of the particulate matter. The aim of this study was to investigate the characteristics of particulate matter inside museum so as to identify the emission sources.

METHODOLOGY

Study Location and Particulate Matter Collection

A museum in Shanghai was chosen as the sampling location during the time period of September 15, 2009 to October 31, 2009. The museum was selected due to its location, which was positioned in Shanghai CBD, in close proximity of the intersection of two elevated main roads. It represented the location with a variety of pollutant sources originated from different types of vehicles. There were dozens of bus stations surrounding the museum (Appendix 1). The open time of the museum was from 9:00 to 16:50. The museum covered 39,200 m^2 in total, out of which, the display area accounted for 12,000 m^2. It should be pointed out that the temperature and humidity were kept constant in the museum throughout the year via central air-conditioning system, which was T = 20 ± 0.5°C, RH = 45 ± 2%. The air-conditioning system was working for 24 hours every day. The air change rate of the museum 6–10 times for opening period and high efficiency particulate air (HEPA) filter was installed to prevent particulate pollutant entering indoors. A total number of 9 display halls and 1 lobby were investigated in this study, including Ancient Chinese Bronze hall (BR), Ancient Chinese Sculpture hall (SC) and the lobby on ground floor, Ancient Chinese Ceramics hall (CE) situated on level 2, Chinese Calligraphy Archive (CA), Ancient Chinese Painting Gallery (PA) and Ancient Chinese Imperial Seal hall (SE) located on level 3, Ancient Chinese Jade hall (JA), Ancient Chinese Coin hall (CO), and Chinese Ming and Qing Furniture hall (FU) belonging to level 4. The particle number concentration was continuously sampled from the open time to midnight in each display hall. The sampling conducted in October included national day holiday, more visitors would visit museum during this time period. The flow rate of visitors during the National Day was 1.6 times greater compared to the working day.

Instruments and Pollutants

Measurement of Particle Mass Concentration

The NanoMoudi (Model 125A) (Offenberg *et al.*, 2004; Sardar *et al.*, 2005) was used to monitor the mass concentration of particles in different size intervals. Sampling flow rate was 10 L/min and cut sizes covered the range from 10 nm to 10 μm. Quartz filters (Offenberg *et al.*, 2004) (φ 47 mm) and PTFE filters (d = 0.45 μm, φ 47 mm) produced by Whatman Corp. were used. The filters were preserved in a chamber with constant temperature and humidity (T = 25°C and RH = 40 ± 2%) over a period of 24 hrs before each measurement. The filters were weighed by microbalance (TG332A) (Qi *et al.*, 2008). A weighing error of less than 0.10 mg needed to be attained from multiple weighing of the filter before and after the sampling. The average value obtained was considered as the filter mass in the error range. Hence the particle mass concentration was calculated as the difference of the filter mass before and after the sampling divided by the sampling volume. The cut sizes were at 0.010, 0.018, 0.032, 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6 and 10 μm.

Measurement of Particle Number Concentration

The sampling of particle number concentration was performed using laser particle counters (LASAIR II 310B) with a flow rate of 28.3 L/min and cut sizes of 0.3, 0.5, 1.0, 3.0, 5.0, 10.0 μm. The data was logged at a time interval of 60 sec.

Measurement of WSOC, OC and EC Concentration

The WSOC was determined using a total organic carbon analyzer (TOC, Elementar) (Chow and Watson, 2002) to measure the water soluble total carbon (TC) and water soluble inorganic carbon. The difference between the water soluble TC and inorganic carbon was considered as WSOC concentration. Prior to the analysis, the filter mixed with 15 mL ultrapure water was sonicated over 40 mins followed by 12 hrs of oscillating at a low speed. Clear and transparent filtrate was obtained after being filtered with a PTEF filter (the diameter of the filter was 0.45 μm). The IMPROVE

thermal/optical reflectance (TOR) protocol (Liu *et al.*, 2012) was used for the carbon analysis.

Measurement of Particle Morphological and Elemental Information

Scanning (Ferro *et al.*, 2004) was performed as an effective methodology of micro-analysis to determine the morphological and elemental information of individual particles. The high and low vacuum resolution of SEM was 3.0 nm and 4.0 nm, respectively, and the magnification range of 5–100,000. The morphological features were observed using EOLJSM-6360 LV SEM. The elemental information was obtained by using X-ray detector and digital pulse processor to achieve the mass fraction and mole fraction converted into energy spectrum.

During the entire measurement period, the accuracy of sampling was ensured using a number of quality control protocols. The average value for each sampling was applied for the analysis. Outliers were judged by SPSS and removed prior to conducting the analysis.

RESULTS AND DISCUSSION

Particle Number Concentration

The Impact of Storey

As visitors entered all the exhibition halls according to the order from low to high, when they finished visiting all the exhibition halls on the lower floor level, then they will go upstairs to visit other halls. In addition, ultrafine size particles might enter each hall from the air-conditioning system installed on each floor. Hence the particle characteristics inside different exhibition halls located on different stories were investigated here. Fig. 1 illustrated the average half-hourly particle number concentration varied in 6 size intervals obtained inside the 9 display halls, respectively: 0.1–0.3, 0.3–0.5, 0.5–1.0, 1.0–3.0, 3.0–5.0 and 5.0–10.0 μm. We observed that particles ranging between 0.1 and 0.5 μm dominated the particle number concentration and nearly maintaining constant values throughout the entire monitoring period. PMs within this range might be originated from the ambient air entering from ventilation system which penetrated to indoors. Comparatively, in the case of particles within the other 3 size intervals, in addition to the specific feature appeared in all display halls on the same storey, the peak was observed again during the time period of 19:00–21:00. This was due to the cleaning time in the museum. It can be observed that on the same storey, particle number concentration presented the similar pattern for all size intervals, respectively. The museum was closed at 16: 50. Therefore, the staff would remind tourists to leave half an hour earlier. It was found that the concentration of large particles started to decrease at about 16:20, indicating that the large size particle concentration was significantly affected by the presence of tourists inside the museum. This was especially obvious for particles within the size range of 1.0–10.0 μm.

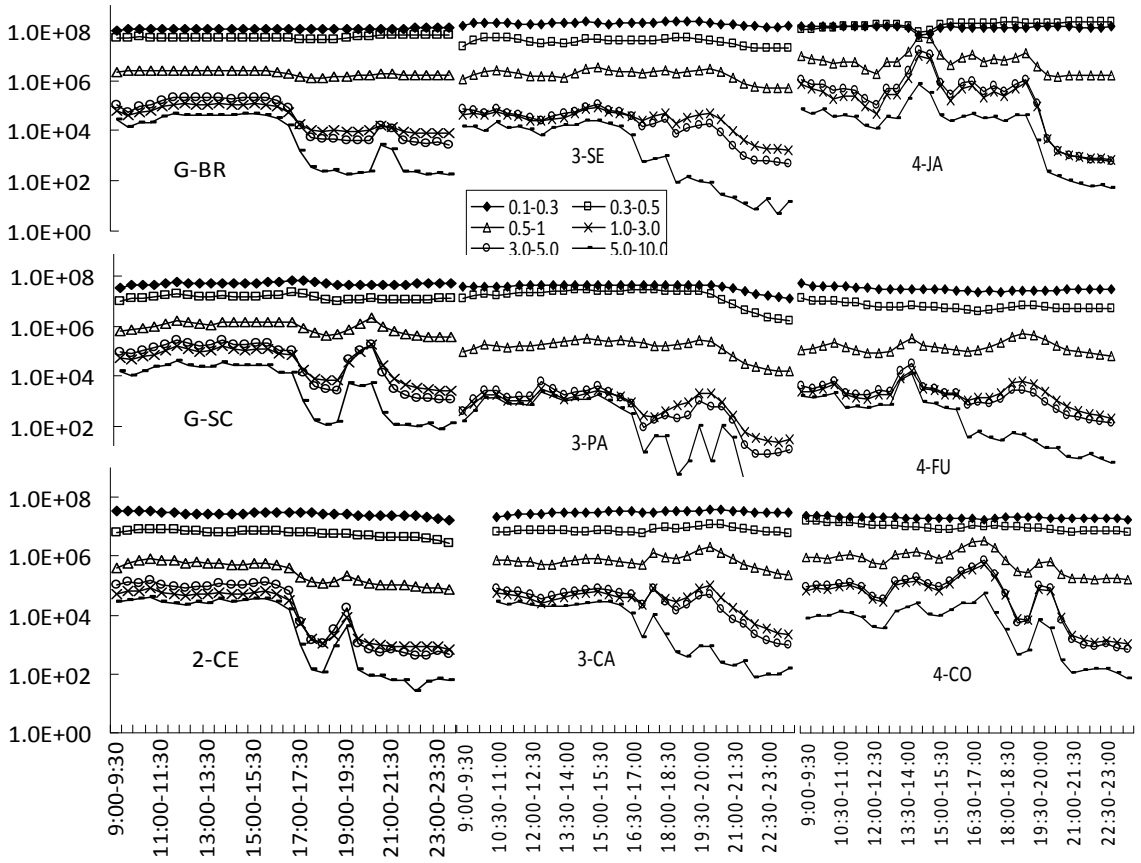


Fig. 1. The daily time-series trend of size-fractionated particle number concentration.

The total particle number concentration in each display hall was given in Fig. 1, exhibiting the identical trends. The contribution of the size-fractioned particles was further investigated. Particles ranging from 0.1 to 0.5 μm dominated over 95% of the total particle number concentration. The percentage of particles in this range was represented using column bars in Fig. 2. It was observed that comparatively greater particle number concentration occurred in the display hall of BR, PA, JA and CO with the percentage value of $PNC_{0.3–0.5}$ 31.6%, 35.5% , 52.3% and 30.9%, respectively. Accordingly, it can be observed that the larger percentage of the particles in the range of 0.3–0.5 μm, the higher particle number concentration appeared in the corresponding display hall. The results of elemental analysis of $PN_{0.3–0.5}$ showed that the mass concentration of S accounted for 16.26% of the total mass, suggesting that their precursors were vehicle emissions. The ratio of Al and Fe existing in $PN_{0.3–0.5}$ was calculated to further evaluate the likely source, which was about 0.13. This was an additional proof that $PN_{0.3–0.5}$ originate from vehicle emissions. This was because that the AL/Fe ratio in $PN_{0.3–0.5}$ produced from anthropogenic activity was less than 1, but about 1.9 for the $PN_{0.3–0.5}$ in soil.

The Impact of Decorative Flooring

Aiming to further investigate the impact of decorative flooring and tourists' movements on the particle characteristics, the average particle number concentration of each size interval monitored in each display hall was summarized and shown in Fig. 3. Ferro *et al.* (2004) concluded that submicron particles only contribute to less than 1% of the suspended particle number concentration (Kim *et al.*, 2010). In this study it was observed that $PN_{0.1–0.3}$ and $PN_{0.3–0.5}$ were fairly stable throughout the day, which almost excluded the influence of resuspension from decorative flooring by tourists' movements on PNC. Consequently, only the particles in the size range from 0.5 to 10 μm were plotted to investigate of the floor impact. A substantially greater number concentration could be observed in JA and CO. In addition, BR, SC and CE exhibited relatively high particle number concentration in most of the cases (Except $CE_{0.5–1}$ and $SC_{5–10}$). To give an explicit interpretation, we summarized the decorative flooring in each display hall, listed in Table 1.

Table 1 illustrated that both JA and CO were covered with block carpet on the floor, in comparison to that BR, SC and CE were decorated with thick carpet.

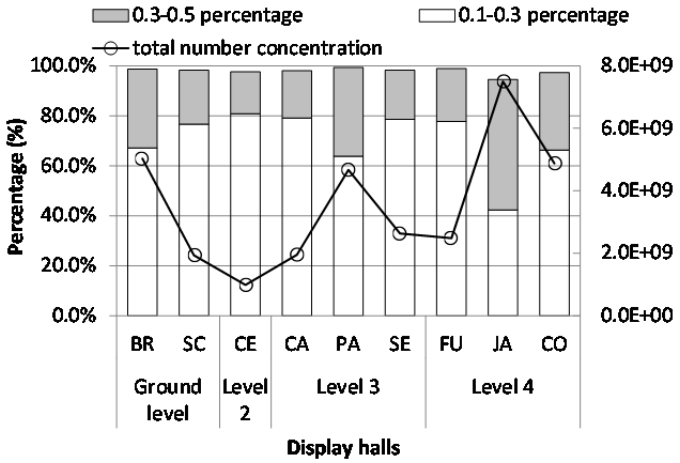


Fig. 2. The total particle number concentration of each display hall and the percentage of particles in the size range of 0.1–0.3 and 0.3–0.5 μm.

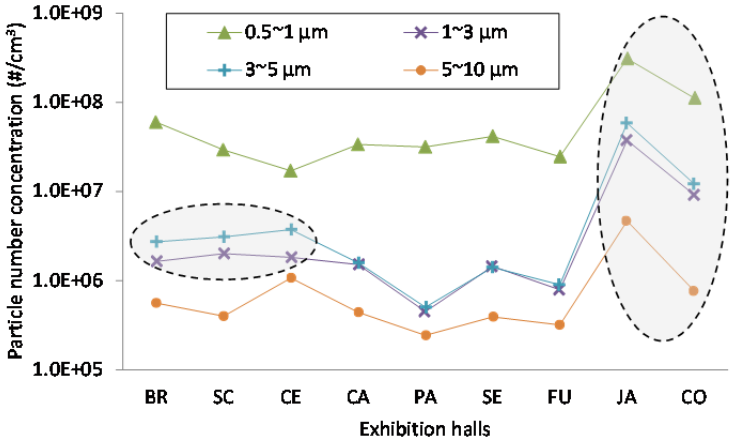


Fig. 3. Average size-fractionated particle number concentration in each display hall.

Table 1. Decorative flooring of each display hall.

Display halls	BR	SC	CE	CA	PA	SE	FU	JA	CO
Decorative flooring	Thick carpet	Thick carpet	Thick carpet	Solid wood	Solid wood	Solid wood	Solid wood	Block carpet	Block carpet

Resuspension rate is the net result of adhesion forces and removal forces acting concurrently on particles deposited on a surface. Regarding the adhesion force, it includes the van der Waals force, the electrostatic force, and the capillary force, a among of which, van der Waals force was the main force impacted by the visitor movement, and worked between the particles and the nearby surface (Kim *et al.*, 2010). It is decided by the Hamaker constant (Ge *et al.*, 2013) and the separation distance between the particle and the surface. The greater the Hamaker constant, the stronger the Van der Waals forces are. Contrary, the greater the separation distance, the smaller the Van der Waals force will be. The Hamaker constant between particles and wood is larger compared to that between particles and carpet. Opposite trend is expected for the separation distance. Consequently, weak van der Waals force existing between carpet and particles was found in this study. Regarding the removal force, it was concluded that occupant thermal plume may play a significant role in transporting pollutants from floor to the breathing zone (Rim and Novosela, 2009). The friction force between the tourists' shoes and the carpet would get the flooring charged in comparison with solid wood. For this reason, it was likely to expect an enhanced thermal plume microenvironment, leading to particle resuspension. Generally, museums use carpets for the decorative flooring for a purpose of good appearance and sound insulation. However, such flooring can cause the resuspension of particles in space. Therefore, intense physical activities of visitors inside museum lead to the resuspension of previously deposited particles. Human activity seems to be the most important factor influencing the particles in the size range of 0.5 and 10 μm. We would advise minimising carpet usage inside museums or implementation of some other precautius measures, such as wearing shoe covers prior to entering.

Particle Number Size Distribution

The particle number size distributions in all display halls were characterized using multi-lognormal fittings, which has been commonly applied to characterize the particle number size distribution indoors and outdoors, following the principles introduced in Hussein *et al.* (2005).

$$\frac{dN}{d\log(\mathrm{D_p})} = \sum_{i=1}^{n} \frac{N_i}{\sqrt{2\pi}\log(\sigma_{\mathrm{g,i}})} \times \exp\left[-\frac{(\log(\mathrm{D_p}) - \log(\overline{\mathrm{D}}_{\mathrm{pg,i}}))^2}{2\log^2(\sigma_{\mathrm{g,i}})}\right] \quad (1)$$

in which, D_p was the particle diameter. N denoted the number of the individual log-normal modes. The three items capable of obtaining an individual log-normal particle number size distribution were the mode number concentration N_i, geometric variance $\sigma^2_{g,I}$ plus the geometric mean diameter (GMD) $D_{pg,i.}$

The simulated curve obtained from Eq. (1), the corresponding measured curve, the predicted separated coarse and accumulation curves in each display hall were compared. It was found that the particle number concentration in all display halls was maintained at fairly similar order of magnitude. As expected, the decomposed particle number size distribution exhibited two modes among all the display halls, attributing to similar originated particulate pollutant sources. The curve patterns of a representative display hall were shown in Fig. 4.

The results obtained from Eq. (1) were presented in Table 2. The geometric mean diameter (GMD) $D_{pg,i.}$ of the accumulation mode was 0.17 μm and the coarse mode was 3.87 μm. The sampling conducted in winter in a park nearby the museum by Yang *et al.* (2006) also observed two modes of particles in the size range of 10 nm–10 μm: 37 nm and 115 nm. The absent nuclei mode in this study was due to the instrument limitation. There was no significant difference of σ (given on the figure) in relation to the coarse and accumulation mode, in the case of all monitored display halls. The value ranged between 1.6 and 1.9 for the accumulation mode, and ranged from 1.3 to 1.6 for the coarse mode.

Aiming to further differentiate between the two modes and trace the sources, the particle morphology was analyzed via SEM photomicrograph, shown in Fig. 5. PMs were classified into 4 types based on their morphology and elemental composition such as aggregated soot, coal fly ash, mineral matter and other types. It was observed that soot aggregates and mineral matter dominated the composition of coarse particles. The contribution of coal fly ash was negligible. However, fine particles were found to be mainly composed of soot aggregates. Regarding the fine particles, by taking into consideration the *impact of particle size* discussed above, a possible solution for preventing them of entering the museum building would be implement of measures such as proper closing of doors and windows, prevention of leaks and draft on the envelop etc. In addition, an effective mechanical system with filtration should be provided, with exclusion of electrostatic filters. Electrostatic filters have the ability of bringing the ozone, which further promotes the conversion from sulphur dioxide to sulphuric acid.

The mineral particles identified in this study were counted as taken inside by tourists. This was the result of the observation that secondary organic aerosols appear with larger particle sizes and due to the fact that mineral soil source was supposed to be absent inside the museum.

The corresponding EDS figure of coarse (a) and fine (b) particles were given in Fig. 6, respectively. Enriched elements on coarse particles included Ca, Si, Al, Na, C, O, S and Mg, in comparison with that significant S content detected

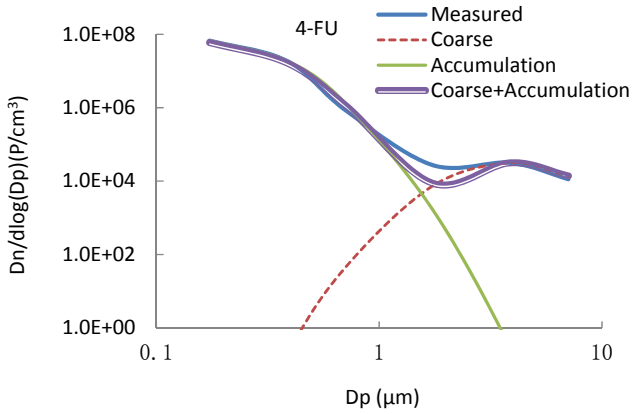
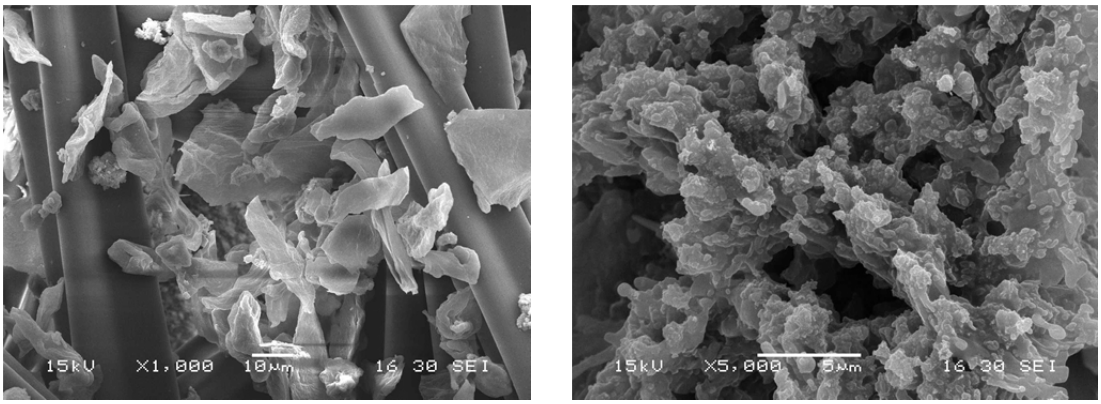


Fig. 4. Decomposing the particle number size distribution using multi-lognormal fitting.

Table 2. Obtained parameter values from Eq. (1).

		BR	SC	CE	CA	PA	SE	FU	JA	CO
N_i	Accumulation	1.2×10^8	4.9×10^8	2.7×10^7	5.6×10^7	9.9×10^7	6.9×10^7	6.7×10^7	1.3×10^8	1.1×10^8
	Coarse	9.1×10^4	1.1×10^5	1.3×10^5	6.4×10^4	1.68×10^4	4.82×10^4	3.14×10^4	1.96×10^6	4.1×10^5
$\sigma_{g,I}$	Accumulation	1.9	1.8	1.6	1.61	1.8	1.6	1.65	1.9	1.9
	Coarse	1.4	1.35	1.5	1.5	1.5	2.7	1.60	1.3	1.3
$D_{pg,i.}$	Accumulation	0.17	0.17	0.17	0.17	0.17	0.17	0.17	0.17	0.17
	Coarse	3.87	3.87	3.87	3.87	3.87	3.87	3.87	3.87	3.87



(a) Coarse particles (b) Fine particles

Fig. 5. SEM photomicrographs of coarse (a) and fine (b) particles.

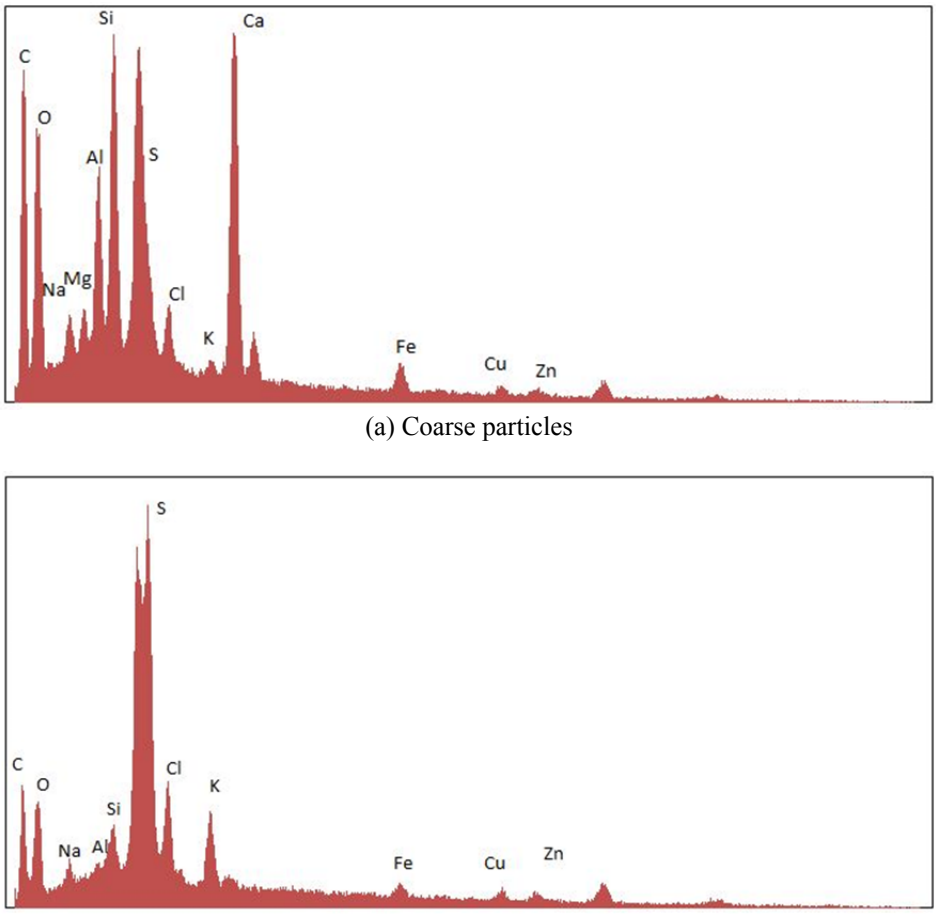
on the fine particles. As we know, increased amounts of Ca, Mg, Na and Si generally came from soil dust. The enhanced presence of S, especially on fine particles, was attributed to vehicle emissions due to the museum's exposure to heavy traffic. Therefore, the presence of tourists contributed to number of coarse particles monitored in this study while fine particles originated from the outside atmospheric environment. The comparatively higher C and O proved the existence of biological particles, such as fragments of pollen or spores.

Particle Matters

The American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) and Canadian Conservation Institute advised that $PM_{0.1}$ concentration limit should be 1–10 μm/m^3 for sensitive materials and general collections. The measured $PM_{0.1}$ in this study reached up to 220 μg/m^3 in the hall measurement. It is noteworthy that although the $PM_{0.1}$ value was not within the recommended value, it appeared in the hall area where the air quality requirement was not as strict as in the areas with sensitive materials exhibited in the display halls and display windows.

OC, EC and WSOC

Fig. 7 showed that the correlation between EC and OC were not significant during both working day and National day holiday measurements, with R^2 = 0.4001 and 0.0323, respectively. The interception of the two straight lines on OC axis indicated the existence of other particulate sources contributing to OC except the common sources for both EC and OC (He *et al.*, 2011). The ratio of OC/EC greater than 2 for all the size-fractioned particles demonstrated the same conclusion (Chow *et al.*, 1993). The total mass concentration



(a) Coarse particles

(b) Fine particles

Fig. 6. The corresponding EDS figure of coarse (a) and fine (b) particles.

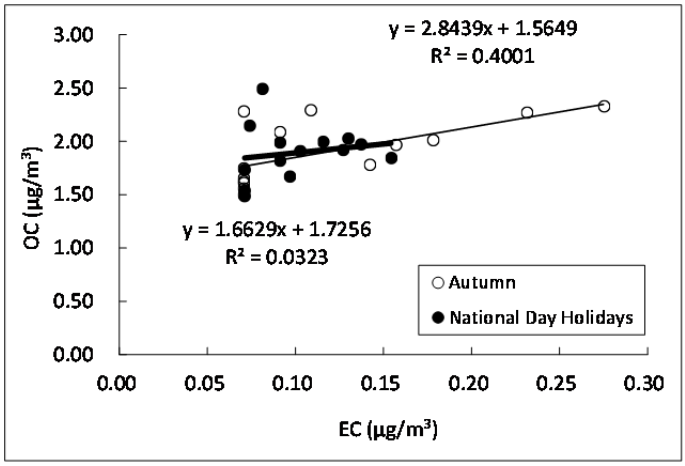


Fig. 7. The correlation between EC and OC during working day and National day holiday measurements.

of EC was 1.68 and 1.42 μg/m^3 obtained from the working day and National day holiday campaign, respectively. The corresponding values for OC were 26.68 and 26.51 μg/m^3, respectively. There was no significant difference of size-fractionated OC existing between working day and National day holiday monitoring ($P > 0.05$), which was observed occurring for EC ($P = 0.01$). This was due to the strong decrease of EC during the National day holiday monitoring for the particles with mode of 240 nm. As EC mainly originates from the direct discharge of combustion processes, reduced EC was interpreted as cutting down the number of vehicles during National day holiday. Regarding OC, in addition to the associated production from combustion processes, a portion of OC comes from non-combustion

processes, such as soil dust, construction dust etc. It was also previously reported that that dust resuspension features greater OC/EC ratios. Considering the large number of visitors each day, it was very likely that the significantly greater amount of OC originates from the soil dust brought in the museum by visitors. Fig. 6 also confirmed high Si concentration was detected from coarse particle, which was very possible brought in by visitors.

The WSOC concentration monitored was 17.9 and 13.8 μg/m^3 during the working day and National day holiday. The greatest mass concentration appeared at 17.3 μm. The correlation coefficient between WSOC and OC was 0.28 and 0.17 for working day and National day holiday measurement, respectively. The weak correlations implied primary emission which contributed less to OC. The WSOC/OC ratio for the entire size span was 0.69 and 0.58 on working day and National Day holiday, which was within the expected range of 20–80% reported by previous findings (Kondo *et al.*, 2007). High WSOC/OC ratio indicated stronger secondary carbon formation inside the museum.

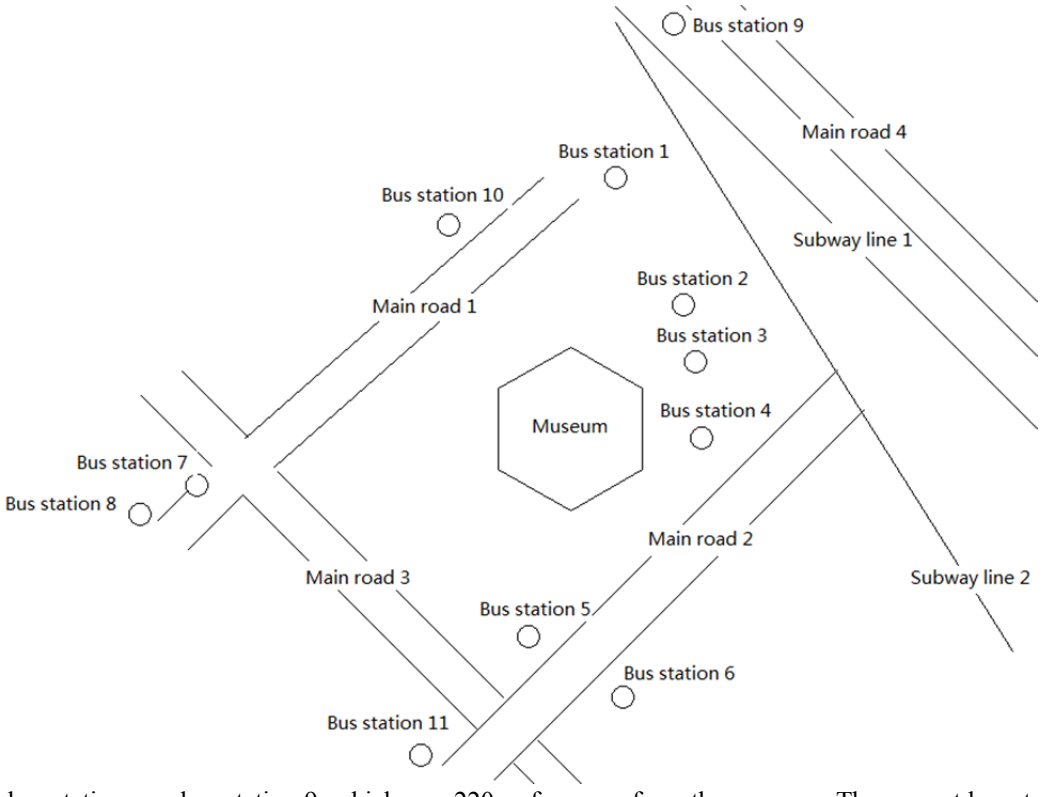
CONCLUSION

Investigating the characteristics of airborne particles inside museums is of urging interest nowadays. This study conducted a multiple-parameter monitoring in the museum located in Shanghai CBD. The impact of storey, particle size and decorative flooring on particle characteristics was analyzed. It was found that particles in each display hall on the same storey exhibited similar patterns of daily time-series trend. Particles in the size range of 0.3–0.5 μm dominated the largest amount of number concentrations. The flooring of carpet easily caused the resuspension of particles in large size spans, affecting the heritage conservation as well as human health. The result of multi-lognormal fitting applied in museum showed an accumulation mode of 0.17 μm and a coarse mode of 3.87 μm, respectively. SEM photomicrograph analysis showed that soot aggregates and mineral matter accounted for the majority of coarse particles with a small contribution of coal fly ash, in comparison with the absolute domination of soot aggregates on fine particles. The corresponding EDS results showed that enriched elements on coarse particles included Ca, Si, Al, Na, C, O, S and Mg, while S was mainly detected on fine particles. It was also found that enhanced OC concentrations were partially brought in by moving tourists.

ACKNOWLEDGEMENTS

This study was sponsored by the grants of 13PJD013 of Shanghai Pujiang Plan, B200-2-13043 of Shanghai Natural Science fund, Innovation Program of Shanghai Municipal Education Commission 12ZZ054, 2-7 of Government of Bilateral Cooperation projects between China and Serbia, WB1113005 and WB1314017 from East China University of Science and Technology. The author would like to thank our colleague and staff from the museum for their contribution to this work.

APPENDIX 1:



(The farthest bus station was bus station 9, which was 220 m far away from the museum. The nearest bus station was bus station 4, which was 60 m far away from the museum.)

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Received for review, December 18, 2013
Revised, March 29 2014
Accepted, May 5, 2014