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Indoor air quality at five site museums of Yangtze River civilization

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HIGHLIGHTS

- IAQ at site museums were surveyed for the first time in southern China.
- Unstable microclimate and acidic air pollutants were frequently observed.
- General hazards to collections and historical ruins were evaluated.

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ABSTRACT

The Yangtze River civilization, dating back to more than 7 thousand years ago, is one of the most historic culture aggregates in China. For long-term conservation of archaeological artifacts and historical ruins along the Yangtze River, indoor air quality at five site museums were investigated during summer and winter. Unstable microclimate conditions were observed at all five museums. The maximal seasonal variations in temperature and relative humidity were 25.7 °C and 40.0%, respectively. The mass concentration of $PM_{2.5}$ inside the museums remained at high levels, ranging from 33.9 to 79.6 μg/m^3 in winter and from 52.8 to 113.0 μg/m^3 in summer. Organic matter (OM) constituted a major fraction (39.3%−53.9% in summer, 22.1%−27.8% in winter) of total $PM_{2.5}$. The results showed that besides short-term fluctuation and seasonal variation in microclimate conditions, infiltration of gaseous and particulate air pollutants should be of increasing concern at museums in Southern China.

1. Introduction

The fluctuation of microclimate conditions inside museums as temperature and humidity plays an important role on the conservation and preservation of culture heritage. (Camuffo et al., 2001; Pavlogeorgatos, 2003). Furthermore, anthropogenic and secondary air pollutants, including airborne particulate matters (PM), acidic gases (e.g., sulfur dioxide and nitrogen dioxide), and strong oxidants (e.g., ozone) pose potential soiling hazard and atmospheric deterioration damages on most artworks in museums (Godoi et al., 2013; Krupinska et al., 2012). When deposited, particles may cover the details of collections, and cause further mechanical abrasion of surfaces and disfiguration of objects during cleaning. Some reactive species in airborne particles, including nitrate, sulfate, ammonium, and organic acids could also initiate or increase corrosion of materials (Nazaroff and Cass, 1991). Soot particles may cause the disfiguration of porous surfaces (painting, frescoes, statues, books, textiles, et al.), and increase the rate of metals corrosion (Tétreault, 2003). Damage can be intensifed through the synergistic effects from air pollutants, temperature and/or humidity (Camuffo et al., 1999).

The Yangtze River civilization is one of the most historic civilizations in China. The Yangtze River, together with the Yellow River, is known as "Mother River" in China. The earliest cultivation of rice in the world was also found along the Yangtze River drainage areas

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(Normile, 1997). The Yangtze River civilization flourished along the middle reaches of the river, dating back to 6400–6100 cal yr BP (calendar year before present) as Daxi culture (Yasuda et al., 2004). In the past 20 years, several cultural heritages from different historical periods of the Yangtze River civilization were excavated and subsequently protected by site museums.

All the site museums were built directly on the archaeological sites, promoting a sense of ownership of the past fortified settlements among present resident or nearby populations, as well as greater local interest in its preservation. Those museums are located in the most populated regions in China, which are experiencing extreme air pollution problems with the rapid urbanization and industrialization in recent years. In Emperor Qin's Terra-cotta Museum in northern China, high levels of indoor fine and coarse particles were observed in both summer and winter seasons and most of them were resulted from outdoor activities (Cao et al., 2011). Therefore, it is of great importance to understand the indoor air quality of an site museum by evaluating its ventilation conditions of buildings and measuring the microclimate conditions, particle intrusion, and penetration of gaseous pollutants from outdoor environment. In this study, it is the first time that a survey of indoor air qualities at five site museums along the Yangze River in Southern China were carried out during two seasons as summer and winter. Potential hazards due to exposure to the fluctuant microclimate, gaseous and secondary particulate acidic species in indoor air were investigated to ensure the long-term conservation of those valuable and vulnerable cultural properties.

2. Methodology

2.1. Indoor and outdoor sampling

Field measurements and sampling were carried out at five institutions, among those Jinsha Museum (JS) in Chengdu city was located in the upper reaches of the Yangze River, and Xiongjia Mound Museum (XJM) in Jingzhou city was in the middle reaches of the Yangze River. The other three were distributed in the Yangze River Delta, including Tianluoshan Site Museum (TLS) in Yuyao city, Hongshan Ruin Museum (WXHS) in Wuxi city and Yuan Dynasty Water Gate Museum (SHWG) in Shanghai city. Detailed information of the sampling sites are listed in Table 1.

In all the five museums, the archaeological pits were covered by display halls to shield the pits and relics from direct solar illumination and precipitation. Besides, XJM also equipped with a huge glass chamber to separate the archaeological pit with tourist passway. Indoor sampling sites were placed in the center of the display buildings. Outdoor sampling sites were located about 5 m outside the main entrance of each display hall except in XJM museum the outdoor site was in the tourist passway and outside the glass chamber. All the samplers and real-time analyzers were settled at ground level.

Two portable Q-Trak Plus IAQ monitors (Model 7565, TSI Inc., Shoreview, MN, USA) were used to obtain the 1-min average air temperature (T) and relative humidity (RH) records indoors and outdoors, respectively. NO_2, SO_2, and O_3 concentrations were monitored using portable electrochemical gas analyzers (Model 4150, 4240 and 4480, Interscan Corporation, USA) at 1-min interval at a flow rate of 1 L/min. $PM_{2.5}$ samples were collected for about 24 h at 5 L/min flow rate with a mini-vol portable sampler (Airmetrics, Springfield, OR, USA) onto 47 mm quartz-fiber filters (Whatman, Clifton, NJ, USA) which were pre-heated at 900 °C for 3 h to remove the residual carbon before sampling. Two Dust-Trak Handheld Aerosol Monitors (Model 8520, TSI Inc., Shoreview, MN, USA) with PM2.5 inlets were placed at each indoor and outdoor location to measure $PM_{2.5}$ mass concentrations at 1-min intervals. The monitor can measure particulate matters based on the method of light scattering at a flow-rate of 3.0 L/min. Before sampling, a separate calibration test was carried out to convert the results given by the Dust-Trak monitor into corresponding concentrations obtained by gravimetric methods.

2.2. Sample analysis

The collected amount of $PM_{2.5}$ was calculated from averaged weight difference of each Quartz-fiber filter before and after sampling. The gravimetric analysis was done by using an electronic microbalance with 1 μg sensitivity (Model MC5, Sartorius, Göttingen, Germany) after 24-h equilibration at temperature between 20 and 23 °C and RH between 35 and 45%. Energy dispersive X-ray fluorescence spectrometry (Model Epsilon 5, PANalytical B.V., Almelo, Netherlands) was applied to determine the elemental concentrations collected on the filters. The ionic species

Table 1
Information of sampling sites.

Museum	Location	Surrounding	Culture	Historical eras	Collection	Ventilation	Area (m^2)	Date	Weather
JS	N30.67°, E104.01°	Urban	Ancient Ba-Shu culture	1250–650 BC	metal, stone, ivory, pottery	HVAC operated HVAC not operated	7588	15/Jul./2013 10/Jan./2014	Sunny Cloudy
XJM	N30.62°, E112.00°	Rural	Chu culture	476–400 BC	Jadeware, bone	Mechanical ventilation	1577	19/Jul./2013 13/Jan./2014	Sunny Sunny
TLS	N30.02° E121.38°	Suburban	Hemudu culture	5500–7000 BC	agricultural remains, wooden architectures	Natural ventilation	5000	22/Jul./2013 19/Jan./2014	Sunny Cloudy
WXHS	N31.48° E120.05°	Suburban	Yue culture	470 BC	Celadon, Pottery, jadeware, colored glaze	Natural ventilation	1100	25/Jul./2013 20/Jan./2014	Sunny Cloudy
SHWG	N31.26°, E121.43°	Urban	Yuan culture	1270–1370 AD	Wooden stake, stone and metal	HVAC operated HVAC not operated	2300	28/Jul./2013 22/Jan./2014	Sunny Sunny

Note: HVAC means Heating, Ventilation and Air Conditioning.

($NH_4^+NH_4^+$, K^+, Na^+, Mg^{2+}, Ca^{2+}, NO_3^-, and SO_4^{2-}, Cl^-, F^-) was measured by an ion chromatography (Model DX600, Dionex Inc., Sunnyvale, CA, USA) (Chow and Watson, 1999). ACS12 column (150 × 4 mm) and an AS14 column (150 × 4 mm) were used for cation and anion analysis, respectively. One-fourth of each filter sample were removed and extracted in 6 ml of high-purity water for the ion analysis. Data on organic carbon (OC) and elemental carbon (EC) concentrations was obtained using a DRI Model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA) with the IMPROVE A thermal/optical protocol method by a punch of 0.526 cm^2 from each quartz filter (Cao et al., 2003).

3. Results and discussions

3.1. Microclimate

The air temperature and relative humidity inside and outside each site museum were summarized in Table 2. The weather conditions of the five cities along the Yangtze River in southern China were characterized with distinguishing seasonal variations in air temperature during summer and winter sampling campaigns. No obvious seasonal variations in outdoor humidity was observed. Microclimate in respective museums followed the fluctuations of outdoor weather conditions but varied between different museums due to their diverse location, building structure and display strategy, equipment and operation strategy of air conditioning and mechanical ventilation systems. In summer campaign, Shanghai city underwent the highest temperature (35.7 °C for daily average) of the year. However, indoor temperature in SHWG Museum reached the lowest (22.5 °C for daily average) among the five investigated museums due to the 24-h air conditioning operation with the location of archaeological site 10 m below the ground. Although XJM museum was equipped with mechanical ventilation system, the daily averaged relative humidity inside the closed glass chamber was still 7% higher than that in tourist passway. The American Society of Heating Refrigeration and Air-Conditioning Engineers (ASHRAE) handbook (ASHRAE, 2011) has recommended five classes of HVAC system setpoint in general museums for short-term fluctuations and seasonal variations according to the collection risks. All site museums investigated in this study have exceeded the AA class (no risk of mechanical damage to most artifacts and paintings) recommendation for seasonal variations (T: ±5 °C, RH: no change). Among those, the highest seasonal variation in air temperature was observed at naturally ventilated TLS museum, reaching 25.7 °C. At WXHS, another museum with natural ventilation conditions, daily averaged indoor relative humidity in summer season was 40.0% higher than that in winter. With the protection of closed glass chamber in XJM museum, although the lowest diurnal fluctuations in air temperature (1.1 °C) and relative humidity (11.0%) were obtained, obvious seasonal variations were still observed (T: 19.3 °C, RH: 22.6%).

Fig. 1 illustrated the fluctuations in microclimate conditions at five site museums during summer and winter season. ASHRAE (2011) AA class recommendation for short-term fluctuations (T: ±2 °C, RH: ±5%) were shown in red lines. In each season, air tightness of the diaplay hall and ventilation strategy played an important role on the short-term fluctuations of microclimate conditions. The highest diurnal fluctuations of indoor temperature among the five site museums was recorded at TLS museum with natural ventilation condition, reaching 7.9 °C in summer and 11.7 °C in winter, respectively. Inside the archaeological pits covered with glass chamber at XJM museum, the lowest fluctuations in microclimate conditions were achieved in both summer and winter. Two in five investigated site museums have exceeded ASHRAE indoor temperature fluctuation recommendation in summer, as well as the number of museums reached three in winter (Fig. 1a). As shown in Fig. 1b, indoor diurnal relative humidity at nearly all of the five site museums fluctuated dramatically during the summer and winter campaigns.

3.2. Gaseous pollutants

The 24-h averaged concentrations of key acidic and oxidizing gases, including SO_2, NO_2, and O_3 inside and outside the five site museums are summerized in Table 3. The indoor concentrations of SO_2 at the investigated museums varied due to the various source emissions around the museums, ranging from 6 to 20 ppb in summer, and from 7 to 30 ppb in winter, respectively. Those SO_2 levels were several to a dozen times higher than the ASHRAE recommended limits for general collections in museums as 0.4–2 ppb. The I/O ratios of SO_2 concentrations ranged from 0.26 to 0.87 in summer, and from 0.47 to 0.96 in winter, implying obvious outdoor sources of indoor SO_2. Althrough some of the outdoor NO_2 concentrations reached several tens of ppb due to heavy traffic in the vicinity of the museums, the indoor concentrations of NO_2 remained at a relative low level, ranging from 1 to 11 ppb in summer, and from 1 to 9 ppb in winter, respectively. Only at two museums as JS and TLS in summer, the indoor concentrations of NO_2 have exceeded the ASHRAE recommended limits (2–10 ppb). The I/O ratio of NO_2 ranged from 0.03 to 0.92 in summer, and from 0.16 to 0.75 in winter. The indoor O_3 concentrations were found from 1 to 19 ppb in summer, and from 2 to 9 ppb in winter. Levels of O_3 at nearly all the five investigated museums were close to or beyond the ASHRAE recommended O_3 limits (0.5–5 ppb). The I/O ratio of O_3 ranged from 0.14 to 0.95 in summer, and from 0.46 to 1.5 in winter.

The average annual levels of SO_2, NO_2 in 31 provincial capitals of China between 2003 and 2010 ranged in 40–60 $\mu g/m^3$, 40–50 $\mu g/m^3$, respectively (Shang et al., 2013), suggesting serious gas pollution in outdoor environment. Most of the concentrations of the measured gaseous pollutants in this study were lower inside the museums than those outdoors, implying their obvious outdoor sources. Previous research also showed that without chemical filtration incorporated into modern HVAC system, gaseous pollutants can easily penetrate into building construction (Cass et al., 1989). Under high relative humidity inside the museums, the acidic and oxidizing gaseous pollutants could be absorbed onto and react with indoor surfaces or suspended particulate matters

Table 2
Daily averaged indoor and outdoor temperature and relative humidity with their standard diviations recorded during summer and winter seasons at different site museums.

Season	Location	JS		XJM		TLS		WXHS		SHWG	
		T (°C)	RH (%)	T (°C)	RH (%)	T (°C)	RH (%)	T (°C)	RH (%)	T (°C)	RH (%)
Summer	Indoor	26.4 ± 0.8	77.3 ± 6.7	28.0 ± 0.2	92.3 ± 2.1	30.1 ± 2.4	83.2 ± 7.8	29.7 ± 0.6	85.1 ± 4.3	22.5 ± 0.7	77.5 ± 6.8
	Outdoor	28.0 ± 1.9	72.0 ± 9.8	28.6 ± 0.3	85.3 ± 3.2	31.2 ± 3.8	74.2 ± 15.4	33.3 ± 3.0	68.9 ± 14.1	35.7 ± 2.6	50.8 ± 9.6
Winter	Indoor	9.0 ± 1.4	65.2 ± 6.7	8.7 ± 0.3	69.7 ± 2.8	4.4 ± 3.6	74.7 ± 13.3	4.8 ± 1.5	45.1 ± 5.6	8.3 ± 0.6	65.7 ± 11.4
	Outdoor	7.3 ± 1.4	70.0 ± 7.9	8.0 ± 0.4	69.8 ± 6.3	3.7 ± 4.4	76.6 ± 16.7	5.1 ± 4.4	37.1 ± 13.8	5.0 ± 1.6	46.5 ± 14.2

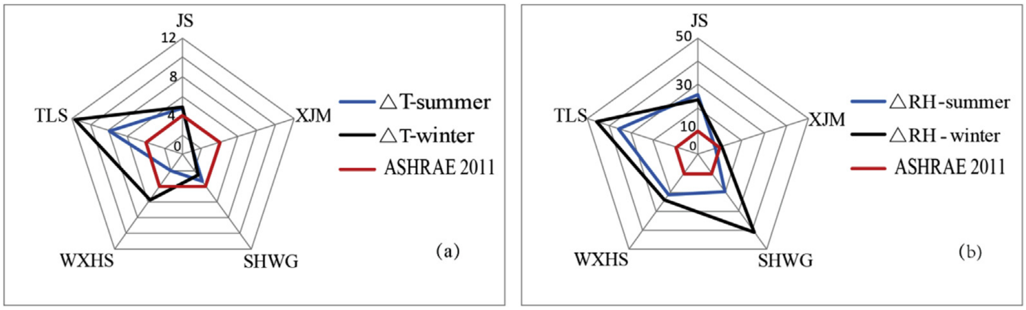

Fig. 1. Indoor fluctuation of temperature (a) and relative humidity (b) at five site museums during summer and winter.

Table 3
Concentrations of gaseous pollutants at five site museums during summer and winter.

Museum	Location	SO_2 (ppb)		NO_2 (ppb)		O_3 (ppb)	
		Summer	Winter	Summer	Winter	Summer	Winter
JS	Indoor	20 ± 20	30 ± 13	11 ± 11	4 ± 5	12 ± 8	4 ± 2
	Outdoor	23 ± 22	31 ± 13	15 ± 13	25 ± 12	26 ± 15	8 ± 4
XJM	Indoor	20 ± 11	21 ± 5	1 ± 1	1 ± 1	1 ± 1	2 ± 1
	Outdoor	34 ± 15	44 ± 13	5 ± 3	2 ± 4	7 ± 4	2 ± 2
TLS	Indoor	6 ± 6	7 ± 6	23 ± 8	5 ± 3	19 ± 9	6 ± 2
	Outdoor	24 ± 11	11 ± 11	25 ± 36	17 ± 12	20 ± 9	13 ± 4
WXHS	Indoor	12 ± 3	18 ± 10	5 ± 5	9 ± 5	9 ± 8	9 ± 3
	Outdoor	28 ± 22	34 ± 19	32 ± 22	33 ± 16	30 ± 28	6 ± 3
SHWG	Indoor	19 ± 5	8 ± 9	1 ± 1	6 ± 7	6 ± 3	8 ± 4
	Outdoor	35 ± 20	12 ± 17	30 ± 15	8 ± 5	44 ± 19	9 ± 4

(Tétreault, 2003). The production of those reactions may pose a potential chemical or salt weathering hazard to all acid-sensitive materials, including metals, carbonate minerals or calcareous materials, textiles and fibers, dyestuffs and pigments. As a strong oxidant, ozone can accelerate the formation of indoor gaseous and particulate acidic species. When absorbed onto building materials, including paintings, carpet, plasterboard, and pinewood, ozone can also react with and damage surfaces or be re-released into indoor environment (Druzik et al., 1990; Salmon et al., 2000).

3.3. $PM_{2.5}$ mass concentration and chemical composition

The 24-h averaged $PM_{2.5}$ mass concentrations at five site museums during summer and winter campaigns are shown in Fig. 2. ASHRAE recommended $PM_{2.5}$ limits was 1–10 μg/m^3 for general collections in museums. Inside the five investigated museums $PM_{2.5}$ levels were measured 3.4–11.3 times higher than the recommendation value. Seasonal and diurnal variation in indoor $PM_{2.5}$ mass concentrations did not follow the pattern of those outdoors, implying that besides outdoor intrusion, tourist flow and indoor activities, including cleaning and construction work, also played an important role on indoor fine particulate levels. All the I/O ratio of $PM_{2.5}$ mass concentrations in summer (ranging from 0.61 to 0.94) were lower than 1, while most of those in winter (ranging from 0.86 to 1.09) were larger than 1. Furthermore, indoor $PM_{2.5}$ mass concentrations in summer were lower than those in winter in respective museums. During the haze pollution in 2013, $PM_{2.5}$ in Shanghai reached 90.7 μg/m^3 (Huang et al., 2014) which exceeded the Chinese national standard of 75 μg/m^3. Besides serious PM pollution often occurred in winter in Southern China, most of the

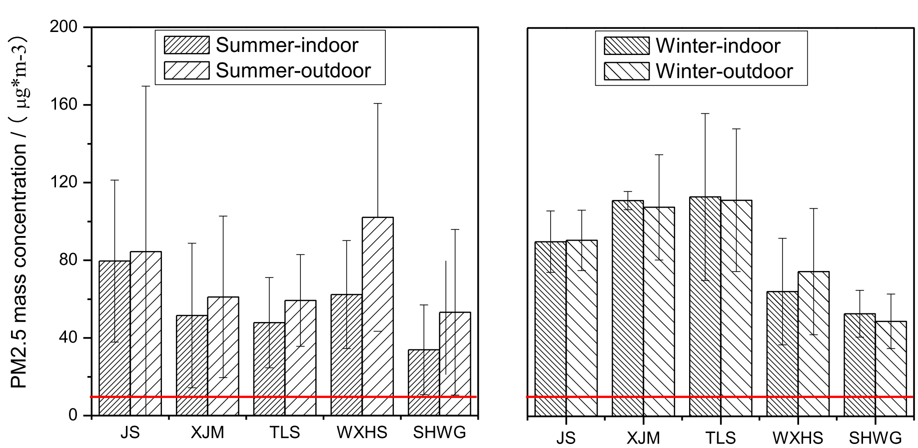

Fig. 2. Summer and winter indoor $PM_{2.5}$ mass concentrations were compared to those outdoors at five site museums. ASHRAE recommendation limit for $PM_{2.5}$ (10 μg/m^3) is shown by the red lines. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

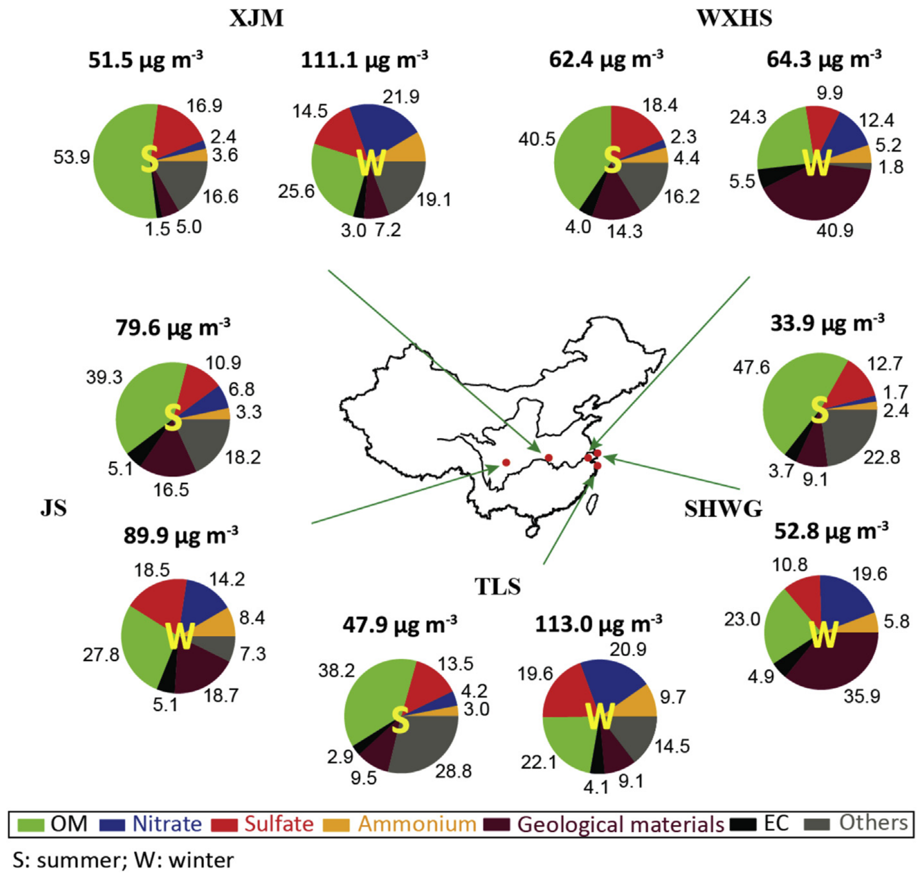

Fig. 3. Material balance charts for indoor $PM_{2.5}$ in summer and winter at five site museums. The average mass concentrations of $PM_{2.5}$ were also listed above each pie in the figure.

HVAC and particle filtration systems did not operate during winter time.

The material balances for geological material (GM), organic matter (OM), elemental carbon (EC), ammonium (NH_4^+), sulfate (SO_4^{2-}), nitrate (NO_3^-), and others (as the difference between the measured mass and the sum of the major components) for indoor $PM_{2.5}$ in summer and winter were shown in Fig. 3. Details of the reconstruction methodology which was used to estimate mass closure refer to the literature (Cao et al., 2011). As shown in Fig. 3, OM dominated (39.3%–53.9% in summer, 22.1%–27.8% in winter) in $PM_{2.5}$ mass at most investigated museums, partly due to the vegetations around each site museum. SO_4^{2-} and GM were also the major chemical components after OM. SO_4^{2-} accounted for 10.9%–18.4% and 9.9%–18.5% of $PM_{2.5}$ mass in summer and winter, as well as the percentages of GM were 5.0–16.5% and 7.2–35.9% in summer and winter, respectively. NO_3^- accounted for 1.7%–6.8% in summer and increased to 12.4%–21.9% in winter, which may be due to more particulate NO_3^- resided in fine particles under lower temperature in winter. NH_4^+ accounted for 3.0%–4.4% in summer and 5.2%–9.7% in winter. The maximal mass concentrations of NH_4^+ of 11.0 $\mu g/m^3$ occurred at TLS museum, which is located at a rural environment and surrounded by rice fields.

Although OM was abundant in most of the winter $PM_{2.5}$ samples, the sum of major secondary water soluble inorganic ions, including SO_4^{2-}, NO_3^-, and NH_4^+ (SNA) predominated at all the five museums, accounting for 16.7%–25.1% and 27.5–50.1% in summer and winter, respectively. In European museums, similar trends were found that secondary particles, ranging from 2% to 53% in fine fractions, made the smallest contribution in summer than in other seasons (Krupinska et al., 2012). Under high relative humidity, on various surfaces, SO_4^{2-} and NO_3^- could form acid conditions which will affect any collections in particular organic materials (Krüger and Diniz, 2011). SNA were easily dissolved in humidity, forming soluble salts, which could penetrate deep into the porous surfaces. With the fluctuations of temperature and humidity, the dissolution and recrystallization of soluble salts may potentially lead to a physical weathering.

EC is a catalyst to enhance the chemical reaction and a soiling contributor (such as soot) to the esthetic values of most culture relics (Van Grieken et al., 2000). In the three southern California museums, elemental carbon was found to yield perceptible soiling after accumulated on vertical surfaces (Nazaroff et al., 1990). Element carbon accounted for a relatively lower percentage (1.5%–5.5%) in $PM_{2.5}$ at the investigated museums during summer and winter campaigns. The indoor EC mass concentrations varied from 0.8 $\mu g/m^3$ (XJM) to 4.1 $\mu g/m^3$ (JS) in summer and from 2.7 $\mu g/m^3$ (SHWG) to 4.6 $\mu g/m^3$ (TLS) in winter. The EC concentrations at most museums in the corresponding season were lower than those measured at Emperor Qin's Terra-cotta Museum (3.9 $\mu g/m^3$ in summer and 7.7 $\mu g/m^3$ in winter) in Northern China (Cao et al., 2011).

4. Conclusions

In this study it was found that most of the buildings at the investigated museums were not able to provide an effective protection for collections against changes in microclimate, and potential hazards from gaseous pollutants and fine particulate matters. Besides being equipped with mechanical ventilation systems, SHWG museum adopted air conditioning and XJM museum had a glass enclosure to achieve better controls in microclimate conditions among the five museums. However, even those two underground museums were facing significant seasonal variations in temperature and relative humidity. A natural ventilation condition made TLS museum the most easily affected by the outdoor weather and air pollution. The penetration of acidic and oxidizing gaseous pollutants, intrusion and resuspension of fine particulate matters at all the site museums posed potential physical and chemical hazards to indoor materials. Damage to artifacts is cumulative and irreversible. All effects to minimize exposing susceptible collections and historical ruins to pollutants and microclimate fluctuation are beneficial to the long-term conservation of cultural heritages.

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