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RESEARCH ARTICLE

Characterization of chemical components and cytotoxicity effects of indoor and outdoor fine particulate matter ($PM_{2.5}$) in Xi'an, China

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Abstract

The chemical and cytotoxicity properties of fine particulate matter ($PM_{2.5}$) at indoor and outdoor environment were characterized in Xi'an, China. The mass concentrations of $PM_{2.5}$ in urban areas (93.29~96.13 μg m^{-3} for indoor and 124.37~154.52 μg m^{-3} for outdoor) were higher than suburban (68.40 μg m^{-3} for indoor and 96.18 μg m^{-3} for outdoor). The $PM_{2.5}$ concentrations from outdoor environment due to fossil fuel combustion were higher than indoor environment. An indoor environment without central heating demonstrated higher organic carbon-to-elemental carbon (OC / EC) ratios and n-alkanes values that potentially attributed to residential coal combustion activities. The cell viability of human epithelial lung cells showed dose-dependent decrease, while nitric oxide (NO) and oxidative potential showed dose-dependent increase under exposure to $PM_{2.5}$. The variations of bioreactivities could be possibly related to different chemical components from different sources. Moderate ($0.4 < R < 0.6$) to strong ($R > 0.6$) correlations were observed between bioreactivities and elemental carbon (EC)/secondary aerosols (NO_3^-, SO_4^{2-}, and NH_4^+)/heavy metals (Ni, Cu, and Pb). The findings suggest $PM_{2.5}$ is associated with particle induced oxidative potential, which are further responsible for respiratory diseases under chronic exposure.

Keywords $PM_{2.5}$ · Indoor and outdoor · Oxidative stress · Inflammation

Introduction

The World Health Organization (WHO) considers air pollution is one of the key determinants of health. China is suffering from severe environmental degradation (e.g., frequent haze episodes, desertification) because of the country's robust economic growth (> 7.0% annual GDP growth over the past two decades) (Ma et al. 2012; Sun and Fang 2001; Zhu and Wang 1993; National Bureau of Statistics of China 2012). The Chinese national annual average of fine particulate matter (aerodynamic diameter < 2.5 μm: $PM_{2.5}$) concentration (e.g., 50 μg m^{-3}) regularly exceed the threshold limit assigned by the National Ambient Air Quality Standards (35 μg m^{-3}) from China (Report on the State of the Environment in China 2015). Nowadays, air pollution is responsible for approximately one in every nine deaths annually, and ambient air pollution alone can cost ~ 3 million people's life in each year (Lim et al. 2012; WHO 2016). High particulate matter concentration has demonstrated to be associated with various adverse cardiovascular and respiratory effects such as pulmonary inflammation, oxidative stress, blood coagulation, and endothelial dysfunction (Atkinson et al. 2014; Leung et al. 2014; WHO 2013; Lin et al. 2013). $PM_{2.5}$ can deposit in the lung periphery and elicit adverse inflammatory responses (Bitterle et al. 2006). The production of reactive oxygen species (ROS) in the human body is a prime concern. ROS comprise chemically reactive oxygen radicals or oxygen-derived

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species such as hydroxyl radical (•OH) and hydrogen peroxide (HOOH). Oxidative stress is an important underlying mechanism by which exposure to particulate matter may lead to adverse health effects when overproduction of oxidants (e.g., ROS and free radicals) counteracts anti-oxidative defenses (Charrier et al. 2014).

Past studies targeted health effects from exposure to ambient air pollution usually focused on outdoor environment only (Broich et al. 2012; Chuang et al. 2013; Ho et al. 2016a). Jalava et al. (2009) found that incomplete combustion and resuspended road dust are important fine particulate sources that are responsible for diverse toxicity. Previous studies showed that chemical composition of particles is an important factor that associated with oxidative stress and inflammatory responses (Ho et al. 2016b; Leung et al. 2014). Nevertheless, people spend over 80% of their lifespans in indoor (Morawska et al. 2013). A thorough understanding about the effects of indoor air pollutants on indoor air quality is essential for public health. A previous study showed that exposure to indoor air pollutants can be different from outdoor (Lim et al. 2011). Higher indoor $PM_{2.5}$ concentrations were observed in activities such as cooking, cleaning, smoking, and combustion-related emissions (Long et al. 2000; Zhang et al. 2018). There is currently a lack of knowledge about effects of indoor $PM_{2.5}$ on bioreactivity and inflammatory cell response, and information about indoor air pollutants distribution, together with indoor-outdoor $PM_{2.5}$ concentration relationship remains scarce (Perez-Padilla et al. 2010).

Xi'an is a sub-provincial city located in the center of the Guanzhong Plain in Northwestern China with a population of ~ 8 million. The city is the south of Qinling Mountains and north to the Loess Plateau, which leads to poor diffusion conditions (Shen et al. 2009). Previous studies in Xi'an suggested that emissions from fossil fuel combustion (coal burning and traffic) were the dominate factors for the $PM_{2.5}$ pollution (Cao et al. 2012b; Wang et al. 2015). The high emissions in winter and accumulation of pollutants can cause severe air pollution in Xi'an with serious health concerns. The aims of this study are to (1) investigate the chemical characteristics of $PM_{2.5}$ collected from different communities and conditions (indoor and outdoor environment) in Xi'an during winter; (2) evaluate oxidative stress and inflammatory responses of the samples; and (3) characterizes the relationships between chemical properties and bioreactivity of the $PM_{2.5}$.

Materials and methods

Sampling locations and sample collection

Three residential communities and different type of households were selected for the indoor and outdoor air quality investigation. These communities are representative for the major areas in the city. The sampling session was conducted from 26 January to 15 March 2016 with municipal/individual heating in full operation for most households (> 80%), and the sampling duration was lasted for 1 week in each community. The samples were collected from three different locations namely as (1) Qujiang village (QJ), the site is located in the southeast of Xi'an city and surrounded by densely populated residential buildings without any factories nearby, which is considered as an urban residential area; (2) Xiangyang community (XY), the site is located in the east of Xi'an and considered as a suburban area with relatively less population and commercial activities; (3) Hui street (H), the site is located in the center of inner city and considered as a tourist and residential area with dense population. The sampling locations are shown in Fig. 1 and the details of each sampling household can be referred to Table 1.

Around 4–5 households were selected in each community to conduct the investigation simultaneously. Indoor air sampling was performed in living room with all instruments positioned at ~ 1.5 m above the floor. Outdoor air sampling was conducted at the open balcony unless stated otherwise and all instruments were positioned at 1–1.5 m above the floor. A mini-volume air sampler (Airmetrics, Eugene, OR, USA) was used for $PM_{2.5}$ collection at a flowrate of 5 L min^{-1}. The pre-fired (900 °C, 3 h) quartz filters (47 mm) (QM-A, Whatman Inc., Clifton, NJ, USA) and Teflon filters (47 mm) (Pall Life Sciences, Ann Arbor, MI, USA) used in the sampling sessions were collected for chemical and bioreactivity analysis, respectively. Filters were weighed before and after sampling by an electronic microbalance (Sartorius MC5, Göttingen, Germany) (± 1 μg sensitivity) after 24 h in equilibrium (20–23 °C, 35–45% relative humidity (RH)). Each filter was weighted in triplicate before and after each sampling procedure, and the differences should be less than 15 mg for blank and 20 mg for exposed filters. The temperature (T) of indoor and outdoor environment, together with relative humidity (RH) of each sampling location, was monitored by online gaseous pollutants instruments with electrochemical sensors (Alphasense B4) in continuous manner.

Chemical analysis

Four anions (SO_4^{2-}, NO_3^-, NO_2^-, and Cl^-) and five cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) were determined in the aqueous extract collected from filter extraction and analyzed by ion chromatography (IC, Dionex 500, Dionex Corp, Sunnyvale, CA). Further information can be referred to Zhang et al. (2011). The concentrations of elements in aerosol samples were determined by Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometry using an X-Ray Fluorescence (XRF) analyzer (PANalytical Epsilon 5, Almelo, The Netherlands). Further information about the procedure can be referred to Cao et al. (2012a). Organic carbon

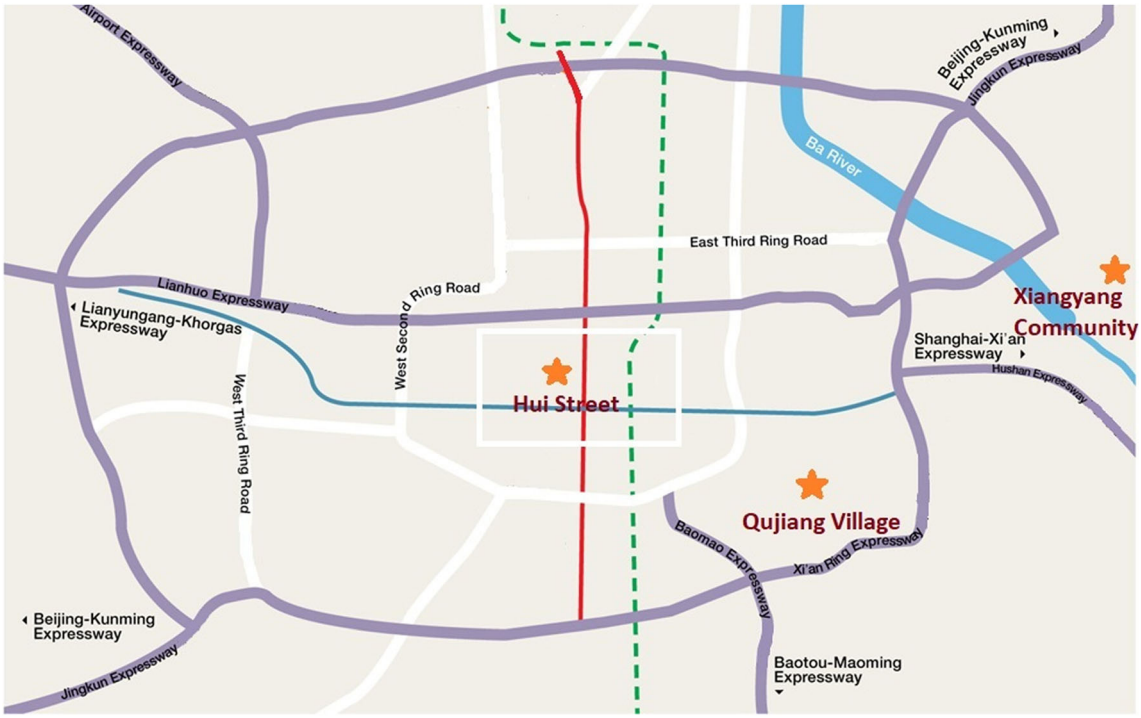

Fig. 1 Location of the sampling areas

(OC) and elemental carbon (EC) were analyzed by using the IMPROVE_A thermal optical reflectance (TOR) method (Cao et al. 2007) in a carbon analyzer (DRI model 2001; Atmoslytic, Inc., Calabasas, CA). The total carbon (TC) concentration was calculated as the sum of OC and EC.

The concentrations of 24 n-alkanes were analyzed in all samples. Dichloromethane and methanol (2:1, v/v) solution was used for filter extraction and the solution was dried and purified by Pasteur pipette anhydrous filled with sodium sulfate ($NaSO_4$) and glass wool. The extract was concentrated for two times (with toluene solvent added in the second extraction) by a rotary evaporator under vacuum to approximately 0.5 ml, and the remaining solution was spiked with 25 μl of 20 μg ml^{-1} Fluoranthene-D_{10} which served as an internal standard. A gas chromatography (Agilent GC-7890A) coupled with mass spectrometry (Agilent GC/MS 5975C) was used for the sample analysis. The analyzed compounds were separated in a fused silica capillary column (DB-5MS; 5% phenyl–95% methyl-polysiloxane, 30 m × 0.25 mm i.d., 0.5 μm film thickness). The above procedure was repeated for all samples.

Extraction of $PM_{2.5}$ for bioreactivity investigation

The combined Teflon filters from each sampling site were extracted with high-purity methanol (5 ml) and the extractant was ultrasonicated in water bath (30 min). This step was repeated in triplicate and the extractants were combined to produce a final extractant after the extraction. Nitrogen gas (N_2 > 99.995%) was purged through the solution for ~ 2 h in order to completely remove the organic solvent. The extracted particles were re-dissolved in phosphate-buffered saline (PBS) with dimethyl sulfoxide (DMSO) (< 0.01%) in different concentrations (0 (control), 100, and 200 μg ml^{-1}).

Cell culture

The human alveolar epithelial cells (A549) were obtained from American Type Culture Collection (ATCC, Rockville, MD, USA) and cultured in cell culture medium (F-12; Thermo Fisher Scientific Inc., MA, USA) with fetal bovine serum (10%), penicillin (100 U ml^{-1}), and streptomycin (100 mg ml^{-1}) under the specific conditions (37 °C, 95% humidity and 5% CO_2). The cells were then incubated with the sample (50 μl) at different particle concentrations of (0 (control), 100, and 200 μg ml^{-1}) for 24 h.

Cell viability and intracellular ROS

The cell metabolic activities were identified by MTT (3-[4, 5-dimethylthiazol-2-yl]-2, 5 diphenyl tetrazolium bromide) assay. A549 cells with particles (50 μl) in different concentrations were seeded on 96-well transwells and filled up to a final volume of 200 μl with F-12 medium. A 100 μl of MTT solution (10%; Sigma Aldrich, St. Louis, MO, USA) was added for color development at 37 °C for 4 h, and the optical density was measured by a microplate reader (ELx800, BioTek, VT, USA) at 540 nm wavelength.

Table 1 Descriptions of sampling locations

Site	Age of house	Area (m^2)	Storage time after decoration	Material used for the floor	Wallpaper	Fuel for cooking	Fuel for heating	Ventilation	Cooking frequency	Smoking
QJ-1	5	100	6–12 months	Composite wood	Yes	Natural gas	Natural gas	Half open < 1 h	3 times/day	No
QJ-2	5	100	6–12 months	Composite wood	Yes	Natural gas	Natural gas	Half open > 1 h	> 3 times/day	No
QJ-3	5	100	6–12 months	Tile	No	Liquefied petroleum gas	Natural gas	Half open > 1 h	> 3 times/day	No
QJ-4	5	102	3–6 months	Wood	No	Natural gas Electricity	Natural gas	Half open < 1 h	1 time/day	No
QJ-5	5	148	3–6 months	Composite wood	No	Natural gas	Natural gas	Wide open < 1 h	3 times/day	No
XY-1	25	59	> 12 months	Composite wood	No	Natural gas	Coal (central heating)	Half open > 1 h	1 time/day	Yes
XY-2	9	136	> 12 months	Composite wood	Yes	Natural gas	Coal (central heating)	Wide open > 1 h	2 times/day	Yes
XY-3	7	127	6–12 months	Wood	Yes	Natural gas	Natural gas	Half open > 1 h	3 times/day	Yes
XY-4	16	90	> 12 months	Wood	No	Natural gas	Coal (central heating)	Half open > 1 h	3 times/day	No
XY-5	15	49	< 3 months	Composite wood	No	Natural gas Electricity	Coal (central heating)	Half open > 1 h	> 3 times/day	No
H-1	23	60	3–6 months	Tile	No	Liquefied petroleum gas	Electric radiator/Coal	Half open > 1 h	2 times/day	Yes
H-2	12	147	3–6 months	Tile	No	Electricity	Air-condition	Half open < 1 h	3 times/day	No
H-3	20	72	> 12 months	Cement	Yes	Coal Electricity	Coal	Wide open > 1 h	2 times/day	Yes
H-4	40	40	< 3 months	Cement	No	None	Electric radiator	Half open < 1 h	None	Yes

ROS production was determined by fluorogenic cell-based method using 2′,7′-dichlorodihydrofluorescein diacetate (DCFH-DA) as a probe. The highly fluorescent compound (2′,7′-dichlorofluorescin; DCF) was produced by oxidation of DCFH with ROS and the dominant cellular ROS was detected by the probe (Daher et al. 2014). After exposure, the fluorescence intensity was determined at an excitation wavelength of 485 nm and an emission wavelength of 530 nm by a Light Luminescence Plate Reader (VICTOR™ X; PerkinElmer, Waltham, USA). The cellular oxidative stress was represented by fluorescence intensity (FI). Four parallel wells for each sample were examined for cell viability and DCFH assay.

Determination of cytokines

The collected supernatant was used to determine cytokines levels. Nitric oxide (NO) production was determined by Nitric Oxide Synthase Assay Kit (Calbiochem, USA) according to the manufacturer's instructions. Each sample was analyzed in triplicate.

Statistical analysis

One-way analysis of variance (ANOVA) with Bonferroni method as post hoc test was applied to examine the variation of bioreactivities along with the blank samples as control. Pearson's correlation coefficient analysis was used to examine the correlations between chemical components and bioreactivity. All the data were analyzed using IBM SPSS statistics 22.0 (IBM ®, New York, NY). The value of $p < 0.05$ was considered as statistically significant.

Results and discussion

Chemical concentration of $PM_{2.5}$

Mass concentrations of $PM_{2.5}$ and chemical compositions obtained from indoor/outdoor environment are shown in Table 2. The daily variations of T and RH at different sampling sites are shown in Figure S1. The temperatures in location XY and H were higher than QJ due to different sampling period. In these communities, the outdoor $PM_{2.5}$ concentration was highest in location QJ and followed by H. The $PM_{2.5}$ concentration in location XY was lowest and ~ 37.7% lower than QJ. The sampling location in QJ is termed "village-in-cities" (villages become engulfed in urban centers as the population and geographic parameters of cities grow) and without central heating. This situation demonstrated that high consumption of coal and liquefied petroleum gas use for heating and cooking purpose in individual homes could further lead to higher $PM_{2.5}$ emissions. The sampling location H is considered as a tourist area with high population density, large number of restaurants, and

Table 2 Chemical compositions of the samples in indoor and outdoor environment

Chemical species	QJ-in	QJ-out	XY-in	XY-out	H-in	H-out
Inorganic compositions (μg m^{-3})						
$PM_{2.5}$	93.29 ± 57.27	154.52 ± 75.97	68.40 ± 25.64	96.18 ± 24.49	96.13 ± 50.43	124.37 ± 54.92
OC	32.07 ± 11.04	40.79 ± 15.33	17.71 ± 6.83	15.57 ± 3.97	20.10 ± 8.81	22.45 ± 7.16
EC	7.67 ± 3.45	10.49 ± 4.66	4.19 ± 1.67	4.92 ± 1.56	5.16 ± 2.79	6.33 ± 2.61
Total carbon	39.7 ± 14.3	51.3 ± 19.9	21.9 ± 8.3	20.5 ± 5.4	25.3 ± 11.1	28.8 ± 9.7
SO_4^{2-}	2.89 ± 1.7	4.64 ± 2.79	1.83 ± 0.61	2.64 ± 0.67	2.79 ± 1.6	3.64 ± 1.84
NO_3^-	3.62 ± 2.46	7.73 ± 4.71	1.94 ± 1.43	3.13 ± 2.17	4.36 ± 3.21	6.24 ± 4.04
NH_4^+	1.20 ± 1.08	2.92 ± 2.09	0.30 ± 0.41	0.55 ± 0.67	1.44 ± 1.28	2.16 ± 1.59
K^+	0.20 ± 0.13	0.34 ± 0.19	0.12 ± 0.12	0.11 ± 0.07	0.14 ± 0.08	0.17 ± 0.1
Mg^{2+}	0.15 ± 0.04	0.17 ± 0.03	0.18 ± 0.06	0.21 ± 0.02	0.13 ± 0.03	0.15 ± 0.03
Ca^{2+}	0.39 ± 0.15	0.57 ± 0.31	0.70 ± 0.33	0.95 ± 0.25	0.24 ± 0.25	0.31 ± 0.15
Cl^-	0.69 ± 0.31	1.54 ± 0.81	0.51 ± 0.17	0.70 ± 0.24	0.73 ± 0.44	0.92 ± 0.49
Na^+	1.47 ± 0.39	1.40 ± 0.2	1.25 ± 0.26	1.43 ± 0.13	0.71 ± 0.25	0.78 ± 0.17
NO_2^-	0.11 ± 0.04	0.10 ± 0.03	0.12 ± 0.05	0.13 ± 0.01	0.11 ± 0.03	0.10 ± 0.01
Total ions	10.63 ± 5.68	19.35 ± 10.56	6.90 ± 2.88	9.70 ± 3.93	10.49 ± 6.57	14.31 ± 8.09
Ti	0.018 ± 0.004	0.025 ± 0.007	0.034 ± 0.014	0.051 ± 0.019	0.023 ± 0.007	0.030 ± 0.007
Mn	0.018 ± 0.005	0.023 ± 0.005	0.021 ± 0.006	0.033 ± 0.009	0.018 ± 0.008	0.025 ± 0.008
Fe	0.224 ± 0.046	0.347 ± 0.101	0.427 ± 0.159	0.672 ± 0.193	0.283 ± 0.089	0.417 ± 0.07
Ni	0.004 ± 0.002	0.012 ± 0.02	0.004 ± 0.002	0.004 ± 0.001	0.003 ± 0.002	0.004 ± 0.002
Cu	0.021 ± 0.004	0.061 ± 0.093	0.022 ± 0.007	0.022 ± 0.003	0.02 ± 0.005	0.023 ± 0.005
Zn	0.077 ± 0.028	0.125 ± 0.043	0.083 ± 0.043	0.100 ± 0.058	0.110 ± 0.077	0.128 ± 0.078
Sb	0.048 ± 0.012	0.046 ± 0.013	0.048 ± 0.014	0.053 ± 0.013	0.049 ± 0.016	0.051 ± 0.018
Ba	0.185 ± 0.037	0.195 ± 0.029	0.197 ± 0.046	0.189 ± 0.017	0.192 ± 0.033	0.204 ± 0.049
Pb	0.053 ± 0.021	0.065 ± 0.028	0.045 ± 0.016	0.051 ± 0.014	0.056 ± 0.023	0.067 ± 0.020
Total elements	0.644 ± 0.118	0.895 ± 0.160	0.876 ± 0.212	1.171 ± 0.221	0.749 ± 0.193	0.994 ± 0.163
Alkanes (ng m^{-3})						
C17	2.48 ± 0.48	1.84 ± 0.93	3.02 ± 0.86	3.54 ± 1.45	3.27 ± 0.24	3.61 ± 1.59
C18	2.31 ± 1.53	1.8 ± 1.03	5.6 ± 1.88	3.77 ± 1.36	4.18 ± 0.46	3.94 ± 1.82
C19	3.66 ± 2.07	2.75 ± 1.56	3.23 ± 1.24	1.97 ± 0.56	2.3 ± 0.59	5.54 ± 2.56
C20	2.95 ± 1.06	3.68 ± 1.42	3.04 ± 0.65	3.45 ± 1.74	3.65 ± 1.11	4.28 ± 2.07
C21	8.32 ± 1.35	18.01 ± 5.36	9.96 ± 2.1	25.88 ± 9.21	13.86 ± 3.21	24.94 ± 10.71
C22	15.04 ± 5.19	31.97 ± 7.94	15.06 ± 3.73	41.43 ± 10.96	20.44 ± 6.34	33.09 ± 14.63
C23	22.81 ± 7.14	34.3 ± 9.41	20.84 ± 6.05	73.1 ± 24.34	41.46 ± 12.2	66.43 ± 25.14
C24	67.96 ± 34.37	104.0 ± 35.33	55.96 ± 16.55	161.23 ± 52.31	92.66 ± 20.03	122.21 ± 49.32
C25	40.88 ± 18.33	80.93 ± 20.54	38.26 ± 9.28	95.61 ± 28.45	69.05 ± 17.19	93.81 ± 35.86
C26	22.31 ± 9.75	39.42 ± 8.24	16.01 ± 4.51	45.96 ± 18.42	28.08 ± 12.42	38.73 ± 14.27
C27	17.53 ± 5.17	32.15 ± 6.16	18.83 ± 3.79	47.84 ± 15.32	31.52 ± 19.4	35.77 ± 16.7
C28	8.76 ± 2.75	15.96 ± 2.71	10.49 ± 3.49	35.92 ± 19.57	16.99 ± 6.17	21.44 ± 8.25
C29	13.92 ± 5.53	20.03 ± 4.65	14.93 ± 3.83	33.54 ± 17.21	21.09 ± 11.56	24.8 ± 9.14
C30	4.9 ± 1.5	2.82 ± 0.78	5.11 ± 1.21	15.58 ± 4.28	7.52 ± 3.69	7.36 ± 3.17
C31	7.24 ± 3.23	3.62 ± 1.87	8.44 ± 1.62	20.24 ± 8.51	17.58 ± 14.49	12.79 ± 5.48
C32	2.22 ± 0.96	1.75 ± 0.59	2.93 ± 0.47	5.26 ± 2.06	4.26 ± 2.13	4.91 ± 1.27
C33	1.85 ± 0.65	1.37 ± 0.73	3.2 ± 0.68	4.47 ± 1.67	4.69 ± 3.05	3.49 ± 1.06
C34	1.79 ± 1.14	0.52 ± 0.27	1.25 ± 0.74	4.21 ± 1.34	1.82 ± 0.66	1.36 ± 0.61
C35	0.89 ± 0.44	0.33 ± 0.16	0.8 ± 0.47	3.77 ± 1.68	0.96 ± 0.37	1.44 ± 0.84
C36	0.46 ± 0.07	0.18 ± 0.06	0.55 ± 0.32	2.02 ± 0.46	0.46 ± 0.18	0.73 ± 0.37
C37	0.92 ± 0.84	0.26 ± 0.09	0.42 ± 0.25	1.52 ± 0.82	0.59 ± 0.12	0.67 ± 0.39
C38	0.54 ± 0.31	0.2 ± 0.06	0.27 ± 0.13	1.76 ± 0.71	0.48 ± 0.26	0.46 ± 0.19
C39	0.27 ± 0.16	0.22 ± 0.11	0.29 ± 0.17	0.62 ± 0.42	0.36 ± 0.06	0.17 ± 0.05
C40	0.27 ± 0.24	0.23 ± 0.01	0.17 ± 0.08	0.67 ± 0.26	0.12 ± 0.07	0.22 ± 0.05
∑Alkanes	250.14 ± 94.98	398.20 ± 82.65	238.53 ± 49.55	633.22 ± 142.62	387.27 ± 132.15	512.07 ± 184.12

-in stand for indoor environment; -out stand for outdoor environment

The concentrations of metals were kept 3 decimals due to the low values

high traffic flow, together with self-built houses occupied by the residents. Due to the higher temperature in location H, the $PM_{2.5}$ concentrations were slightly lower than location QJ with less heating activities. Heating, cooking, and vehicles were the major sources for location H. The sampling location XY is in suburban area of the city with central heating. Less anthropogenic $PM_{2.5}$ emissions were noticed when comparing to other sampling locations. Infiltration from outdoor to indoor air could be a possible factor affecting the indoor air quality.

The carbonaceous fractions showed similar trends to the $PM_{2.5}$ concentrations. The OC and EC concentration were highest at location QJ in indoor and outdoor environment (Table 2), which could be attributed to coal combustion emissions (Cao et al. 2004). The outdoor concentrations were

higher than indoor environment except for the OC fraction in sampling location XY. This could be due to additional indoor combustion sources (i.e., smoking). The organic carbon-to-elemental carbon (OC/ EC) ratio is an indicator to analyze emission sources, secondary organic carbon (SOC) formations, and transformation/deposition process (Cao et al. 2005). The average OC/EC values were in a range of 3.2–6.0 (Supplementary Materials: Table S1) and the values indicate that the emissions from carbonaceous fraction in communities were possibly attributed to a combination of coal combustion, vehicle exhaust, and biomass burning in origin. The OC/EC ratio in location XY at outdoor environment was lower than the other sampling locations. This could be possibly due to higher contribution of vehicle emissions, and the high OC/EC ratio in location H in indoor environment was potentially attributed to higher contribution from residential coal combustion (Watson et al. 2001). The indoor SOC accounted for 30.4–31.6% of the OC, whereas in outdoor environments, SOC only contributed in a range of 9.8–15.2% to the OC (Supplementary Materials: Table S1). This could be due to higher temperature and relatively more enclosed indoor environment that favored SOC formation.

The concentrations of total ions in location QJ and H were higher than XY, and the concentration in outdoor was higher than indoor environment at all locations. NO_3^-, SO_4^{2-}, and NH_4^+ were the most abundant ions and the contribution of NO_3^-, SO_4^{2-}, and NH_4^+ were in a range of 25.8–41.3%, 24.5–28.7%, and 3.3–13.6%, respectively. NO_3^- ions in location H and QJ at outdoor environment showed higher contributions and these observations could be potentially attributed to higher coal combustion and vehicle emissions, and the high contribution of NO_3^- (~ 39%) at location H in indoor could be related to the use of honeycomb briquette for heating purpose (Shen et al. 2008). The total concentration of elements was highest in location XY at outdoor environment (1.17 ± 0.22 μg m^{-3}) with possible high contribution from mineral dust. Location H with more anthropogenic activities showed the second highest total concentration of elements in outdoor environment (0.99 ± 0.16 μg m^{-3}), and the corresponding indoor environment was also in relatively high level. Fe and Ba were the two dominant elements accounting for 34.7–57.4% and 16.1–28.7% of the total elements, respectively.

The total concentrations of n-alkanes in outdoor environment were ~ 1.3–2.7 times higher than indoor. Location XY at outdoor environment showed highest concentration (633.22 ± 142.62 ng m^{-3}) compared with the other locations. In indoor environment, the total concentration of n-alkanes in location H (387.27 ± 13.15 ng m^{-3}) was higher than communities with central heating, indicating possible influences from coal burning for residential heating purpose (Bi et al. 2003). C24 showed highest concentrations (55.96~161.23 ng m^{-3}) in all n-alkanes components followed by C25 (38.26~95.61 ng m^{-3}). Low molecular weight (LMW) components (< C27) were the main composition in n-alkanes and fossil fuel combustion could be attributed to the main sources in indoor and outdoor environment. The concentrations of high molecular weight (HMW) components (> C27) were higher than other locations at location XY in outdoor environment. This could be due to more biogenic emissions in suburban area. The ratio of HMW/LMW and carbon preference index (CPI, the relative quantities of odd/even carbon number n-alkanes) (Supplementary Material: Table S2) were commonly used to identify the potential sources of n-alkanes. The HMW/LMW and CPI values were all close to 1.0, suggesting possible dominant anthropogenic emissions (Hong et al. 2017).

Relationship between indoor and outdoor air quality

The indoor-to-outdoor (I/O) ratios for the chemical components shown in Fig. 2 were calculated to evaluate the relationships between indoor and outdoor pollutions. All I/O ratios were < 1.0 with large variations due to different emission sources and indoor conditions. The I/O ratios were in a range of 0.60–0.77, suggesting possible infiltration from outdoor to indoor environment and relatively less emissions from indoor activities (e.g., heating, cooking, and cleaning).

The I/O ratios of total carbon ranged from 0.77 to 1.07 and the higher ratio was contributed from the OC (Supplementary Materials: Figure S1). Indoor cooking and heating activities could potentially contribute to the OC emissions, and the present of EC was possibly due to infiltration to indoor air (lower I/O ratio). The I/O ratios of total ions ranged from 0.55 to 0.73 and the influences of indoor emissions were relatively low in QJ. In Figure S2 (Supplementary Materials), the I/O ratios of NO_2^- were ≥ 1.0, this observation could be due to favorable indoor conditions and beneficial to the instable constituents. Ions such as SO_4^{2-}, NO_3^-, and NH_4^+ all showed lower I/O ratios compared with the others, indicating possible further contributions from coal combustion and vehicle emissions in outdoor environments (secondary formation reactions). The I/O ratios of crustal origin elements (Ti and Fe) ranged from 0.64 to 0.68, which were lower than the values observed in anthropogenic elements (0.77–0.93). The crustal elements were mainly contributed from outdoor dust, whereas other elements could be emitted from human activities in indoor environment and location XY and H were substantially influenced by indoor effects (Lui et al. 2017). The I/O ratios of n-alkanes were in a range of 0.38–0.76, indicating outdoor environment was more influenced by fossil fuel emissions in winter and the n-alkanes in indoor environment were transported from outdoor environment in origin. Various I/O ratios of individual n-alkane (Supplementary Materials: Figure S3) were attributed to different emission sources. The LMW components were mainly originated from fossil fuels combustion and higher concentrations were observed in outdoor environment, whereas several HMW components (e.g.,

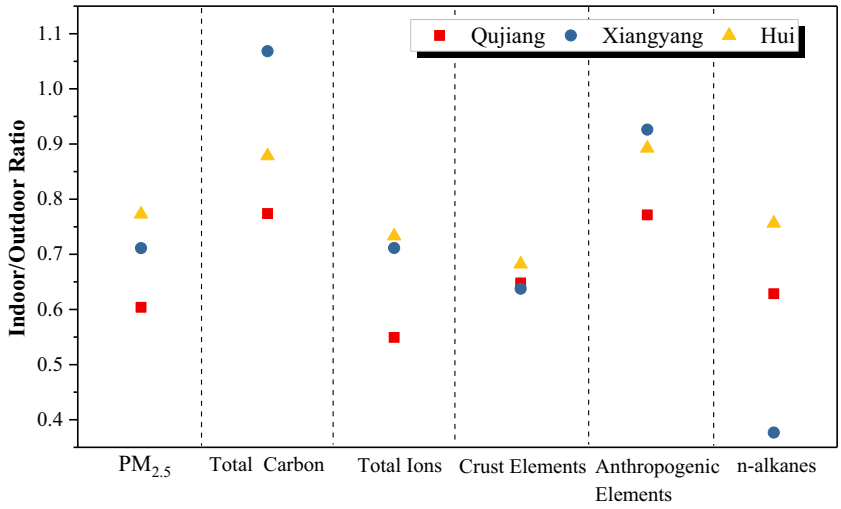

Fig. 2 Indoor/outdoor ratios of mass and chemical components in the sampling areas

C18, C30, C31, C33, C34) showed higher levels in indoor environment at location QJ and H. This observation could be due to emissions from plants or cooking/heating activities in families. The aforementioned I/O values were all comparable to a previous study in Rome (Italy) (Albinet et al. 2007).

Bioreactivities of cells exposed to particles

The bioreactivities of A549 cells exposed to $PM_{2.5}$ are shown in Fig. 3. The cell viability of A549 exhibited dose-dependent decrease in all conditions. Lower cell viability (~ 45% at 200 μg ml^{-1} of $PM_{2.5}$ exposure) was observed at location QJ and H in outdoor environment. The results of cell viability in outdoor environment were lower than indoor, indicating possible variations on chemical compositions from different emission sources and sampling conditions could lead to different toxicity to human cells. The NO and oxidative potential demonstrated dose-dependent increase with $PM_{2.5}$ concentrations. And the $PM_{2.5}$ could possibly cause cytotoxicity in human alveolar epithelial cells. The NO and oxidative potential levels in outdoor environment were all higher than indoor, indicating potentially higher oxidative and inflammatory reactions. The level of oxidative potential is in descending order of H > QJ > XY at indoor environment. For outdoor environment, the oxidative potential is in descending order of QJ > H > XY. In location XY, the $PM_{2.5}$ in indoor environment released highest NO concentration and the production of NO could further induce inflammation. All bioreactivities showed significant differences with the control groups, whereas only trends were observed for different sampling sites.

Various extracellular sources could induce oxidative stress and disrupt the balance between ROS production and antioxidant defenses in biological environment (Deng et al. 2013). ROS could be generated from different environmental sources, such as anthropogenic combustion-derived sources (Chuang et al. 2011). EC and metals were identified as important parameters in ROS formation. The higher surface area of soot and transition metals could induce ROS formation under Fenton-like reactions/inhibition of anti-oxidative processes (Dilger et al. 2016; Limbach et al. 2007). The location QJ in outdoor environment was influenced by strong coal combustion emissions and the higher EC concentrations comparing to the other sampling sites could lead to high oxidative potential. Location H in indoor environment was influenced by self-heating activities, high levels of metals from combustion emissions, and poor ventilation. All of these could contribute to the overall oxidative stress. NO was identified to play important roles in mediating inflammation and cytotoxicity of tissue by generating peroxynitrite anion via superoxide radicals reactions (Nam et al. 2004). NO can also protect cells against oxidative damage from hydroxyl radicals by converting them to less toxic compounds (Chuang et al. 2012). Previous studies showed that NO was released in human alveolar epithelial cells through catalytic activities with isoform of NO synthase (iNOS) and the expression of iNOS was induced by various cytokines (IL-1β, TNF-α, and IFN-γ) and endotoxin (lipopolysaccharide) (Alvarez and Evelson 2007; Kwon and George 1999). The high expressions of NO in location XY at indoor environment (51.2 mmol ml^{-1}) indicate higher potential of inflammatory cascade in the sampling location, and possibly due to immunocompetent cells production to the recruitment of leukocytes (Kwon et al. 2001). The overproduction of NO is considered as cytotoxic (Diociaiuti et al. 2001). At location QJ and H in outdoor environment showed high levels of NO, suggesting combustion activities could possibly induce higher cellular toxicity, although further investigation will be required in future analysis.

Correlation between chemical compounds and bioreactivity

To identify the concentrations of chemical compounds per unit volume (m^3) of $PM_{2.5}$ and the association with

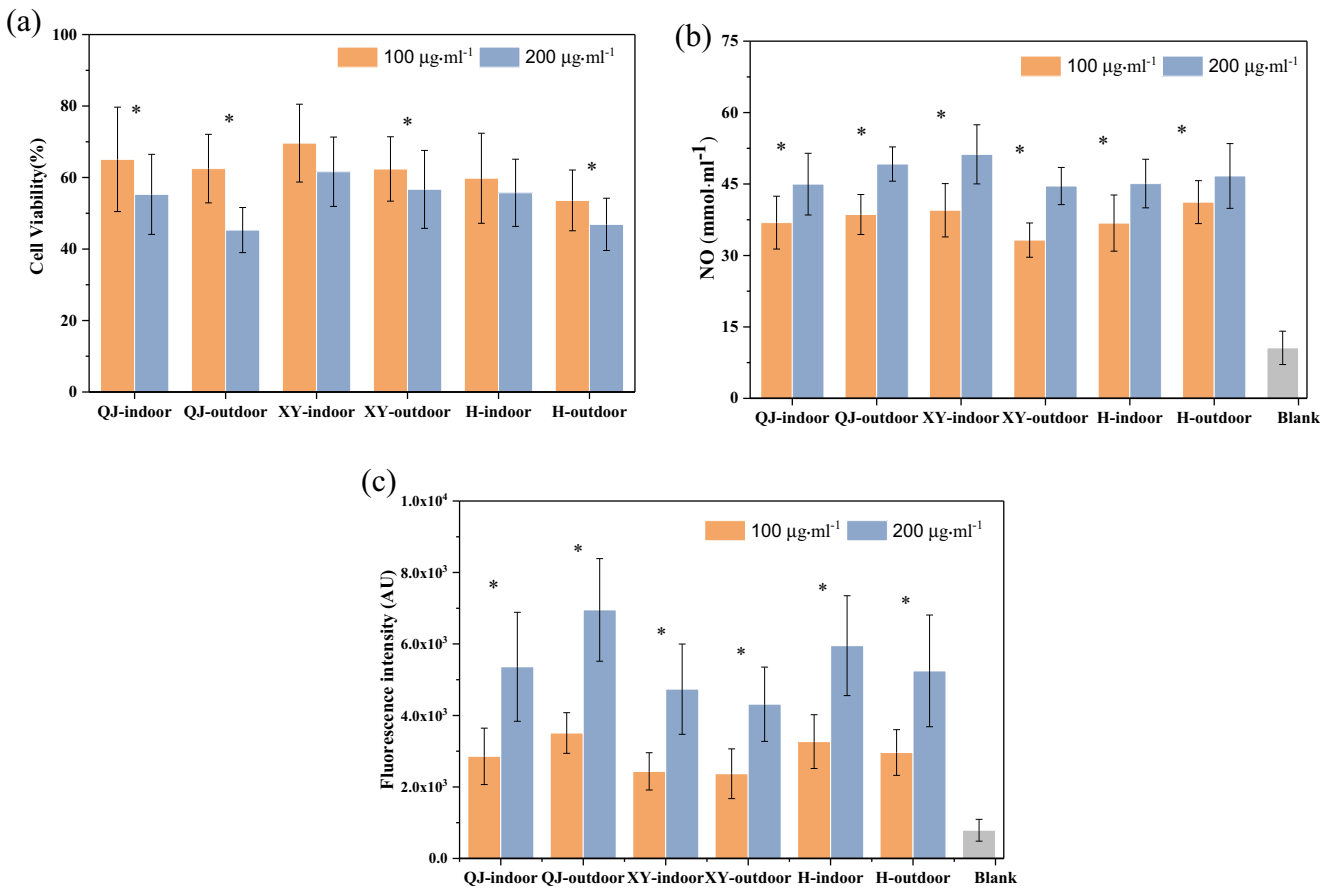

Fig. 3 The bioreactivities of A549 cells in exposure to different conditions: **a** cell viability; **b** nitric oxide; **c** oxidative potential (*$p < 0.05$ compared with control)

bioreactivity, Pearson's correlation coefficients (R) were calculated between ROS-inflammatory activity and the normalized PM compounds (μg ml^{-1}) as shown in Table 3. Only those chemical components that demonstrated moderate ($0.4 < R < 0.6$) to strong correlations ($R > 0.6$) with ROS-inflammatory responses are presented in this study. EC and secondary inorganic aerosols (NO_3^-, SO_4^{2-}, and NH_4^+) were negatively correlated with NO production ($|R| > 0.50$). Strong correlations were identified between ions (Cl^- and K^+) and cell viability ($R = -0.69$, $R = -0.63$)/oxidative potential ($R = 0.70$, $R = 0.65$), these two ions were mainly emitted from fossil fuel combustion. Moderate positive correlation was identified between Ni and oxidative potential ($R = 0.54$). Moderate positive correlations were also identified between Cu and Pb against cell viability ($R = -0.53$, $R = -0.58$)/oxidative potential ($R = 0.58$, $R = -0.63$). No significant correlations were identified between alkanes and bioreactivity. It was proved that NO can protect the cells by inhibition of NOS reactions or trigger toxic peroxynitrite anion via reaction with superoxide radicals. Both negative and positive correlations with the chemical species could be identified for NO. In our study, the negative correlations between NO and carbon/ions may indicate the inhibition effect of $PM_{2.5}$ to NO expression in low concentrations. Different physical and chemical composition of $PM_{2.5}$ could lead to different oxidative stress and inflammatory reactions in human lung and cardiovascular system (Kroll et al. 2013). Hence, the $PM_{2.5}$ collected from different sampling locations could lead to different cytotoxicity and bioreactivity. A past study showed that high OC and EC concentration could be associated with pro-inflammatory activity, and the inflammatory response was driven by the transition metals and organic compounds that coated on carbon surface (Dilger et al. 2016). Associations between sulfate, nitrate, and ammonium against oxidative-inflammatory responses and cardiovascular reaction were all identified in previous studies (Ho et al. 2016b; Chuang et al. 2007). Several heavy metals (Pb, Fe, and Ni, and Cu) were used as toxicity indicator (Chuang et al. 2012; Shen and Anastasio 2012). A past study showed that ROS generation was linked to combustion emission (Verma et al. 2009). Therefore, the outdoor environment in winter and specific indoor environment with coal combustion demonstrated higher bioreactivity potential.

Table 3 Correlations coefficients of chemical compositions and bioreactivity tests

	Cell viability	Nitride oxide	Oxidative potential
OC	− 0.30	− 0.46	0.50
EC	− 0.33	− 0.53*	0.43
SO_4^{2-}	− 0.34	− 0.64*	0.41
NO_3^-	− 0.30	− 0.63*	0.42
NH_4^+	− 0.35	− 0.62*	0.49
K^+	− 0.63*	− 0.08	0.65**
Mg^{2+}	0.37	0.16	− 0.38
Ca^{2+}	0.41	0.23	− 0.35
Cl^-	− 0.69**	− 0.04	0.70**
Na^+	0.21	− 0.16	− 0.26
Ti	0.30	0.18	− 0.43
Mn	0.04	− 0.05	− 0.35
Fe	0.16	0.29	− 0.35
Ni	− 0.48	0.09	0.54*
Cu	− 0.53*	0.17	0.58*
Zn	− 0.41	0.13	0.44
Sb	− 0.12	0.27	− 0.04
Ba	0.35	0.11	− 0.02
Pb	− 0.58*	− 0.19	0.63*

** Correlation is significant at the 0.01 level (2-tailed)

* Correlation is significant at the 0.05 level (2-tailed)

Conclusions

The characteristics of $PM_{2.5}$ at indoor/outdoor environment from different locations in Xi'an city was investigated in this study. Higher concentrations of $PM_{2.5}$ were observed in outdoor environments compared to indoor and this observation was possibly attributed to fossil fuel combustions. Lower $PM_{2.5}$ concentrations were observed for the rural compared to urban areas in all conditions (indoor and outdoor environment). The collected $PM_{2.5}$ was shown to induce oxidative stress and inflammatory response. The trends of bioreactivities were potentially due to different $PM_{2.5}$ emissions at indoor/outdoor environment, in addition to the variations of chemical components at different locations. In this study, the extraction solvent for ions and alkanes were different from $PM_{2.5}$ extraction in the cell experiments. This limitation can be an important contributing factor to the overall correlation analysis results. Further studies will be necessary especially for the associations between organic components and cytotoxicity. The results suggest the need to minimize indoor pollutants emissions from various sources (e.g., central heating and coal burning) in a hope to curb any related respiratory diseases.

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