The chemical composition and toxicological effects of fine particulate matter ($PM_{2.5}$) emitted from different cooking styles

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Abstract

The mass, chemical composition and toxicological properties of fine particulates ($PM_{2.5}$) emitted from cooking activities in three Hong Kong based restaurants and two simulated cooking experiments were characterized. Extracts from the cooking $PM_{2.5}$ elicited significant biological activities [cell viability, generation of reactive oxygen species (ROS), DNA damage and inflammation effect (TNF-α)] in a dose-dependent manner. The composition of PAHs, oxygenated PAHs (OPAHs) and azaarenes (AZAs) mixtures differed between samples. The concentration ranges of the Σ30PAHs, Σ17OPAHs and Σ4AZAs and Σ7 carbonyls in the samples were 9627 – 23452 pg m^{-3}, 503 – 3700 pg m^{-3}, 33 – 263 pg m^{-3} and158 – 5328 ng m^{-3}, respectively. Cell viability caused by extracts from the samples was positively correlated to the concentration of benzo[a]anthracene, indeno[1,2,3-cd]pyrene and 1,4-naphthoquinone in the $PM_{2.5}$ extracts. Cellular ROS production (upon exposure to extracts) was positively correlated with the concentrations of $PM_{2.5}$, decaldehyde, acridine, Σ17OPAHs and 7 individual OPAHsTNF-α showed significant positive correlations with the concentrations of most chemical species (elemental carbon, 16 individual PAHs including benzo[a]pyrene, Σ30PAHs, SO_4^{2-}, Ca^{2+}, Ca, Na, K, Ti, Cr, Mn, Fe, Cu and Zn). The concentrations of Al, Ti, Mn, Σ30 PAHs and 8 individual PAHs including benzo[a]pyrene in the samples were positively correlated with DNA damage caused by extracts from the samples. This study demonstrates that inhalation of $PM_{2.5}$ emitted from cooking could result in adverse human health effects.

Keywords:

Cooking emissions; PAHs; Oxygenated PAHs; Azaarenes; Plasmid scission assay

1. **Introduction**

Emissions from cooking constitute an important source of particulate matter (PM) in the indoor and outdoor environment (Abdullahi et al., 2013; Cheng et al., 2016). Emissions from cooking has been identified as an important source of fine particulate matter (aerodynamic diameter < 2.5 µm: $PM_{2.5}$) in populated urban areas such as Hong Kong (Allan et al., 2010; Huang et al., 2011; Mohr et al., 2012). A previous study showed that commercial cooking restaurants in the South Coast Air Basin, USA emitted ~10.4 tons/day of $PM_{2.5}$ (Gysel et al., 2018). Such emissions represent a major source of exposure to $PM_{2.5,}$ which can adversely affect human health (Chiang et al., 1997; See and Balasubramanian, 2006; Zhong et al., 1999a).

Previous review articles have documented the emission of PM of various size ranges and > 300 chemicals species [e.g. organic carbon (OC), elemental/black carbon (EC/BC), metals/metalloids, water soluble ions (Cl^-, SO_4^{2-}, NO_3-, K^+, Ca^{2+}, Mg^{2+}), volatile organic compounds (VOCs), carbonyls, polycyclic aromatic hydrocarbons (PAHs)] from cooking activities (Abdullahi et al., 2013; Wang et al., 2017; Zhao and Zhao, 2018). The emitted PM, chemical species in gaseous phase and bound to PM are formed from a range of reactions (hydrolysis, thermal oxidation, Maillard reaction, recombination) between chemical components of oils, fats, solid food components, water etc. under high temperature (Abdullahi et al., 2013).

The amount, size distribution and composition profiles of PM and their bound chemicals emitted from cooking are driven by factors such as cooking styles, cooking methods, types of oil, ingredients, additives, indoor and ambient conditions (Buonanno et al., 2009; He et al., 2004; McDonald et al., 2003; See et al., 2006; Torkmahalleh et al., 2013; Ho et al., 2006; Huang et al., 2011; Nolte et al., 1999; Saito et al., 2014; Zhao et al., 2007). Cooking (particularly in Asian-style) often involves the application of high temperature oil (> 250 °C) which results in the enhanced formation of various organic compounds such as carbonyls and PAHs (Huang et al., 2011; See et al., 2006). The amount of PAHs released from cooking that involve deep frying (oil-based cooking) was found to be higher than boiling and steaming (water-based cooking) (See and Balasubramanian, 2008). In addition, frying and food ingredients (e.g. fat contents) could be important factors in the PAHs formation during cooking (Saito et al., 2014; Tanaka et al., 2012; Zhu and Wang, 2003). Oxygenated PAHs (OPAHs), nitrogen heterocyclic PAHs (azaarenes: AZAs) are generated together with PAHs from combustion/pyrolysis process. OPAHs can additionally be formed from the photolysis and (photo)chemical oxidation of PAHs (Clergé et al., 2019). The formation of AZAs and OPAHs during cooking have been documented (Blaszczyk and Janoszka, 2008; Szterk, 2015; Li et al., 2016) but these compounds are rarely characterized in cooking fumes (Sun et al., 2020). Several OPAHs (quinones) and AZAs are cytotoxic, involved in the generation reactive oxygen species (ROS), genotoxic, mutagenic, and carcinogens (Bolton et al., 2000; Jung et al., 2001; Sovadinová et al., 2006; Clergé et al., 2019). Some OPAHs and AZAs also been classified by the IARC as probable or possible human carcinogens (IARC, 2010; 2011a,b). Despite some OPAHs and AZAs being equally or even more toxic than the parent-PAHs, most previous studies on emissions from cooking did not characterize the OPAHs and AZAs and hence did not elucidate their possible contributions to the adverse effects of PM (Abdullahi et al., 2013; Ding et al., 2012).

Cooking activities also emit carbonyls and the sources were from cooking fuels combustion and heating of cooking oils (Ho et al., 2006; Lin and Liou, 2000; Zhang and Smith, 1999). Some of the aldehydes that are emitted from cooking activities are toxic themselves and also participate in atmospheric chemical reactions to form secondary pollutants that degrade air quality (Huang et al., 2011; Ho et al., 2006).

Epidemiological studies have demonstrated associations between exposure to cooking fumes and lung cancer risk (Seow et al., 2000; Zhong et al., 1999a). Several studies found that the risk of lung cancer in non-smoking women increases with increasing exposure to cooking fumes (Zhong et al., 1999a; Seow et al., 2000; Metayer et al., 2002). Some of the studies linked in lung cancer risk to exposure to fumes from cooking (frying) with oil at high temperature such as in Chinese-style cooking (Metayer et al., 2002; Shields et al., 1995; Zhong et al., 1999b). PM emitted from cooking was shown to have mutagenic activity (Chiang et al., 1997; Qu et al., 1992; Wu et al., 1998). The International Agency for Research on Cancer (IARC) has classified fumes originating from high temperature frying as a probable human carcinogen (Straif et al., 2006).

Upon inhalation, $PM_{2.5}$ and the chemicals bound to it such as quinones and transition metals it can trigger the overproduction of reactive oxygen species (ROS), which counteracts anti-oxidative defences (Gao et al., 2020; Charrier et al., 2014). The physiological effects of ROS imbalance can cause oxidative stress, inflammatory response, DNA and cell damage, which is the basis for several diseases (Kelly, 2003; Bitterle et al., 2006). Transition metals, secondary organic aerosols and OPAHs (quinones) were shown to induce ROS formation (Charrier et al., 2014; Bates et al., 2019). Polycyclic aromatic compounds (PAHs, OPAHs, AZAs) can cause oxidative DNA damage and DNA adduct formation (Clergé et al., 2019; Xue and Warshawsky., 2005; Yamada et al., 2004; Bolton et al., 2000) that can result in carcinogenic/mutagenic effects and cancers.

Knowledge of the amounts, chemical composition and toxicity of $PM_{2.5}$ emitted from cooking activities remain limited. The aims of this study are to: 1) characterize the chemical composition (including the particularly understudied OPAHs and AZAs) of $PM_{2.5}$ samples collected from different cooking operations; 2) determine the bioreactivity (cell viability, ROS production, DNA damage, and inflammation effects) of extracts of these $PM_{2.5}$ samples and 3) determine the relationship between $PM_{2.5}$ chemical components and bioreactivity.

2. Materials and methods

2.1 Sampling locations

Cooking fumes were collected from the exhausts of three commercial restaurants operating in Hong Kong and two simulated cooking experiments conducted in environmental chambers. The characteristics of the different sampling sites are detailed in Table 1. The studied restaurants cooked and served common cuisines in Hong Kong. The restaurants were selected based on several criteria. Factors such as roof top access availability, electricity supply during sampling, sampling space (minimum 2 m^2), floor plans, information of exhaust system and sampling safety were carefully considered prior to the sampling campaign. Each of the selected restaurants had to possess an independent exhaust system. This was to ensure that the collected samples were only generated from the target restaurant.

The simulated cooking by two common Chinese-style techniques (namely as stir-frying and deep-frying) were conducted in stainless steel environmental chamber of 19.1 m^3 (3.05 m × 3.05 m × 2.05 m) that was designed for measuring emissions from indoor sources.

2.2 Experimental procedures

2.2.1 Sampling from restaurant exhaust system

Sampling duration was synchronised to the peak hours of the restaurants. The peak hours in each restaurant were based on information provided by the owners. A particle sampler (DRI MEDVOL) was used for sampling in this study. The device consisted of a $PM_{2.5}$ cyclone, an inlet stilling chamber, a conical plenum, open-faced filter packs and differential pressure flow control together with a pump. The $PM_{2.5}$ cyclone (Bendix 240) was operated at 113 L/min, which removed particles (aerodynamic diameter > 2.5 μm) in the air stream. The air was purged through the cyclone and further diffused inside to the plenum. The plenum was coated with perfluoro alkoxyalkane (PFA) teflon. The PFA teflon open faced filter holder (Savillex 47-mm injection-moulded) was used throughout the sampling section. A Teflon-membrane (47mm), Nuclepore polycarbonate membrane and quartz-fiber filter (47 mm) were positioned in the filter holders separately.

Sampling was not performed in the kitchen area but at the rooftop, where sufficient space and access to exhaust for sampling were assured. The $PM_{2.5}$ samples were collected simultaneously during the cooking process. The sampling duration was set as 1.5 hours. Background samples from the kitchens were also collected when the kitchens were not in operation. The background samples were typically collected 1.5-3 hours after the restaurants have closed their operations for the day. Four cooking emission samples, in addition to background and field blank samples were collected at each restaurant for four consecutive sampling days.

2.2.2 Sampling from simulated cooking in an environmental test chamber

Induction hot plates were used for the cooking. Each cooking experiment was completed in 1.5 hours. The dishes were representative of typical Hong Kong cuisine including fried "Choy Sum" (a leafy vegetable commonly used in Chinese cuisine) and deep fried chicken breast. The portion of all dishes was enough for 4-5 members of a family. The same chef was responsible for the cooking experiments in order to ensure sample harmony. $PM_{2.5}$ emitted from these simulated cooking experiments (sample D*-E* from test chamber and sample A-C from commercial restaurant) were sampled with an identical sampling system as described in section 2.2.1. After each sampling, the filters were sealed in Petri dishes and frozen (-20 °C) until chemical and biological analysis.

The quartz filters were heated at 900 °C for 3 hours in order to remove any organic vapours on filters before being used to sample. All filters were pre-conditioned at 23±0.5 °C and 50±5% relative humidity (RH) for 48 hours before and after weighing. Each filter was weighed on a microbalance (±1 μg precision, Sartorius AG MC5, Germany) before and after $PM_{2.5}$ sample collection.

2.3 Analytical methods

2.3.1 Chemical components analysis

Detailed description of the analytical methods applied for the determination of the chemical components [OC, EC, inorganic elements, water-soluble ions (Cl^-, NO_3-, SO_4^{2-}, Na^+, Ca^{2+} and NH_4^+), PAHs, OPAHs, AZAs and carbonyls] in the sampled $PM_{2.5}$ can be found in Supplementary Material (Text S1-S4). In summary, the OC and EC were analysed by the thermal/optical carbon analyser. The inorganic elements and water soluble ions were analysed by the Energy Dispersive X-ray Fluorescence Analyser (ED-XRF) and ion chromatography system, respectively. The polycyclic aromatic compounds (PACs) in the filters were extracted with organic solvents and determined by gas chromatography–mass spectrometry (GC-MS) (Bandowe et al., 2016; Bandowe et al., 2014). The carbonyls were extracted from filters with methanol, derivatized using pentafluorobenzylhydroxyl amine (PFBHA) and analyzed by GC-MS. A list of the chemical species determined in the samples are shown in Table S1.

2.3.2 Cell culture and bioreactivity analysis

The human alveolar epithelial cells (A549) were obtained from American Type Culture Collection (ATCC, Rockville, MD, USA) and were cultured in RPMI medium containing 10% fetal bovine serum, penicillin, and streptomycin at 37 °C with 95% humidity and 5% CO_2. $PM_{2.5}$ samples were from Teflon filters removed according to previous reports (Wang et al., 2021), followed by resuspended in 0.01% dimethyl sulfoxide (DMSO) in serum-free minimum essential RPMI medium. Cells were exposed to the $PM_{2.5}$ samples at 0 (from blank filter), 100 and 200 μg/mL for 24 hrs. Cells were examined for cell viability and ROS activity, whereas the supernatant was collected for cytokine analyses.

The cell viability was identified by MTT (3-[4, 5-dimethylthiazol-2-yl]-2, 5 diphenyl tetrazolium bromide) assay. Cellular ROS production was determined by fluorogenic cell based method using 2′,7′-dichlorodihydrofluorescein diacetate (DCFH-DA) as a probe. Inflammation marker tumor necrosis factor-α (TNF-α) was detected through enzyme-linked immunosorbent assay (ELISA) kits (R&D systems, Inc., MN, USA) as per manufacturer guidelines. Details of the cell experiments can be found in Supplementary Material (Text S5-6).

2.3.3 Oxidative DNA damage

The plasmid scission assay (PSA) was used to determine the capability of each sample to induce oxidative DNA damage. The level of particle–DNA interaction and subsequent damage were measured by the three conformations of plasmid DNA topological states namely: supercoiled (no damage), relaxed (minor damage), and linear (severe damage) as shown in Figure S1-2 (Supplementary Material). Due to the amount of sample required for the analysis, $PM_{2.5}$ samples were pooled together for PSA analysis. Additional information about the procedure can be referred to in previous studies (Chuang et al., 2013; Shao et al., 2006). The $PM_{2.5}$ samples were run in suspension using molecular grade water over a range of concentrations. Twenty nanograms (20 ng) of ΦX174 RF DNA was added to the liquid and incubated for the analysis. The samples were conducted in triplicate. The final gel results were captured in images and determined by densitometric analysis (Genetools; Syngene system, UK). Molecular grade water and restriction enzyme PstI were used as control and positive control in this study, which caused 4.1% and 95.9% DNA damage, respectively.

2.4 Calculations and statistical analysis

The sum of the concentration of all analysed PAHs, parent-PAHs, 16 US EPA PAH, OPAHs, AZAs, carbonyls are referred to as Σ30PAHs, Σ21 parent-PAHs, ΣUS-EPA PAHs, Σ17OPAHs, Σ4AZAs and ΣCarbonyls, respectively. The sum of the concentration of parent-PAHs with 2-3 and 4-7 benzene rings are referred to as ΣLMW-PAHs and ΣHMW-PAHs, respectively. ΣCarci-PAHs refers to the sum of the concentration of eight carcinogenic-PAHs (benzo [a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, benzo[ghi]perylene). Statistical analysis was performed using SPSS 21.0 software. The significance level was set at $p < 0.05$. Due to the small sample size and non-parametric nature of the dataset, comparisons of the means of all samples types were done with Games-Howell test. Spearman's rank correlation was applied to identify relationships between chemical species and also between chemical species and biological end points [cell viability, ROS formation, TNF-α, oxidative DNA damage]. The DNA damage value at 1000 μg ml^{-1} dosage and ROS production and TNF-α values at extract concentration of 200 μg/ml were chosen for the correlation analysis.

3. Results and discussion

3.1 Concentration of $PM_{2.5}$, OC and EC

Mass concentrations of $PM_{2.5}$ in all samples are shown in Table 2. The highest and lowest mass concentrations of $PM_{2.5}$ were in samples C (711.5 ± 257.2 μg m^{-3}) and B (177.4 ± 58.4 μg m^{-3}). The concentration of $PM_{2.5}$ in sample C was ~1.45–4.01 times higher than in other samples. The high concentration of $PM_{2.5}$ in sample C could be attributed to cooking methods (stir-frying and deep-frying) in this restaurant which is supported by similar findings from other studies (Abdullahi et al., 2013; See and Balasubramanian, 2006). The concentrations of $PM_{2.5}$ in all samples are comparable to a recent study focused on outdoor char broiling and conventional Chinese cooking (Li et al., 2018). The concentrations of $PM_{2.5}$ in sample D* and E* (chamber) are also comparable to other Chinese cooking styles (Shandong and Hunan) as reported in a previous study (Wang et al., 2015).

The OC was the most abundant chemical component in the $PM_{2.5}$ (samples A-E*). The average OC concentration is in a range of 99.9–338.8 $\mu g/m^3$. The OC concentration in sample C (highest concentration) is ~3.39 times higher than in sample B (lowest concentration). The composition of the $PM_{2.5}$ mass was predominantly comprised of carbonaceous particles, particularly in OC at a proportion, which are higher than or comparable to previous studies (Li et al., 2018; Wang et al., 2015). The results are consistent with other findings that PM emissions from cooking operations were primarily organic in nature (Gysel et al., 2017; Li et al., 2015; Wang et al., 2015; Zhang et al., 2017). The lower OC composition (< 90%) compared to another study on emissions from charbroiling/grilling of chicken and beef could be due to relatively higher use of vegetables in Chinese cuisine (McDonald et al., 2003). A previous study also reported that OC constituted the highest fraction of the mass PM emissions from different cooking processes (See and Balasubramanian, 2008). The highest carbon fractions (> 60%) in sample D* and E* could possibly be due to high-fat content of the cooking materials and high amount of oils used in the cooking processes (Zhang et al., 2017). The EC fractions contribute < 5% of the total $PM_{2.5}$ mass in each of the five samples and is consistent with a previous study (Wang et al., 2015). The variations in the average concentrations of $PM_{2.5}$, OC and EC in the five samples could be due to the differences in cooking ingredients, cooking conditions and methods (Gysel et al., 2018) some of which are outlined in Table 1.

3.2 Concentration of inorganic elements and water extractable ions

The concentrations of elements and ions can be found in Table 2. The two most abundant elements in each sample were S and Cl. Other major elements found in the samples were Na, K, Fe, Mg, Al, Ca, Zn and Ba. Elements with medium range concentrations in the samples were Mn, Sb, Pb and Cu while four other elements (i.e. Ti, V, Cr, Co and Ni) were in low abundance (< 10 ng m-3) in all samples. The highest concentration of 16 of the 20 elements studied was in sample C. Most of the elements are components of food ingredients (vegetables, meat, cooking oils, salt, spices, water (Butnariu and Butu; Cobos and Diaz, 2014; Epstein, 1999) and can therefore be emitted as part of the PM in cooking fumes. Previous studies have also detected these elements in fumes from restaurants with different cooking styles and cooking of meat in simulated facilities (Gysel et al., 2018; Abdullahi et al., 2013; See and Balasubramian, 2008; McDonald et al., 2003). The presence of Fe, Ni, Cr and Cu in the samples could also be due to the release from cooking utensils (See et al., 2006; Taner et al., 2013; Gysel et al., 2018). A previous study showed that the Cr composition of stainless steel materials could vary from 11 to 30% (Kuligowski and Halperin, 1992). Some of the elements could have contaminated the food materials during their growing or processing because these elements are present in environmental compartments (air, soil) from anthropogenic (traffic) and natural sources crustal materials, dust, rocks, soils (Kebata-Pendias and Mukherjee, 2007; Louie et al., 2005). Some of the elements (e.g. Na, Mg, Ca, Fe, Zn, Cu, Co, Mn) that have been detected in the PM samples are essential for humans and are therefore harmless at the required concentrations, but others (e.g. Pb) have no known biological functions in humans and are only harmful to human health (Kebata-Pendias and Mukherjee, 2007). Upon inhalation, some of the transition metals (e.g. Fe, Cu, Mn) can catalyse ROS production leading to oxidative stress, inflammatory effects and oxidative DNA damage, which results in diseases such as cancers, respiratory and cardiovascular diseases (Gao et al., 2020; Pardo et al., 2015; Danielson et al., 2011; Gerlofs-Nijland et al., 2009).

Water extractable ions were also found in all sample extracts (Table 2), which is consistent with previous studies that also detected these substances in cooking fumes both from cooking test chambers and restaurants (Abdullahi et al., 2013; Schauer et al., 2002; See and Balasubramian, 2008). The highest concentrations of each of the ions can be found in Sample C. The concentration of the water soluble ions in sample C showed the following trend: NO_3^- > SO_4^{2-} > NH_4^+ > Cl^- > Na^+ > Ca^{2+}. The trends in other samples are slightly different. NO_3^-, SO_4^{2-} and NH_4^+ could be components of water and other cooking ingredients but could also be secondary products formed from SO_2, NOx and NH_3 emitted from heating of cooking ingredients (See and Balasubramanian, 2008; Schauer etal., 2002).

3.3 PAHs, OPAHs and AZAs in samples

The concentrations of PACs measured in the samples are shown in Table 3 and Figure S3. Samples C shows the highest concentrations of Σ30PAHs, Σ17OPAHs and Σ4AZAs. Sample C also showed the highest concentrations of 27 of 30 PAHs measured in the samples and the sums of PAHs sub-groups (Σ16 US-EPA PAHs, ΣCarci PAHs, ΣLMW-PAHs and ΣHMW-PAHs). Out of the 17 individual OPAHs measured, 13 had the highest average concentrations in Sample C, while the highest concentration of each of the individual AZAs was also in Sample C (Table 3). Benzo[a]pyrene is often used as a main indicator or marker of carcinogenic PAHs (Boström et al., 2002). The concentration (mean±standard deviation in pg/m^3) of benzo[a]pyrene in the samples increased in the order: D (135±32) > C (134.5±82.4) > B (124±67) > E (114±35) > A (94±34). The comparatively large amount of food processed and cooked in the kitchen of this commercial restaurant (Sample C) could be a reason for $PM_{2.5}$ samples to have the highest concentration of PAHs. Another unique feature of this restaurant is that it cooks and serves Cantonese and Hong Kong local cuisine that are prepared with methods such as deep frying, stir frying, pan frying and steaming (Table 1). Since the cooking methods and size of restaurant C is comparable to restaurant B, the main reason for the high concentration of the chemical species in C can be explained by higher emissions during cooking of Cantonese dishes than during the cooking of mixed cuisines and Western dishes prepared in restaurants B and A, respectively (Table 1). Such influence of various cultural cooking styles on the amount of emitted PAHs, as well as the fact that Asian cooking style emits higher PAHs than Western cooking has been reported in previous studies (See and Balasubramanian, 2008; Abdullahi et al., 2013). Our study reveals that different cultural cooking styles will also result in different concentrations of the PAH derivatives (AZAs and OPAHs) in cooking fumes. Specific studies have shown that cooking can result in the formation of AZAs and OPAHs (Blaszczyk and Janoszka, 2008; Li et al., 2016; Szterk, 2015). A combination of factors such as food components, relatively high usage of oil, type of oils, fat content of meat, spices and cooking methods applied (frying at high temperatures) during the cooking of Cantonese and local dishes results in the highest emitted concentrations of PAHs, OPAHs and AZAs (Li et al., 2018; See and Balasubramanian, 2008). Oil-based cooking methods could generally release more PAHs and their derivatives due to direct evaporation, oxidation, pyrolysis, and/or degradation of organic compounds from oils at higher temperature (Abdullahi et al., 2013; Moret and Conte, 2000). The higher concentration of Σ30PAHs, Σ17OPAHs and Σ4AZAs in sample E* compared to sample D* demonstrates that meat cooking by pan frying not only generates higher concentrations of PAHs (which has also been previously reported) but also OPAHs and AZAs than cooking of plant based food by stir frying (Abdullahi et al., 2013; Schauer et al., 1999a). The pyrolysis of animal fats and oils during the cooking of meat could be an explanation for the formation of PAHs and OPAHs. Azaarenes can be formed during cooking as a result of the pyrolysis of nitrogen containing organic compound components (e.g. aromatic amino acids) of food ingredients (Blaszczyk and Janoszka, 2008). The formed PACs could then subsequently be carried with the cooking fumes (Rogge et al., 1991). Food ingredients may also be contaminated with PAHs, which can subsequently be released with the cooking fumes (Martorell et al., 2010). The composition pattern of the PAHs were slightly different for the different samples (Figure 1). The most abundant PAHs in samples A were naphthalene (10.9%), phenanthrene (10.7%), acenaphthene (9.2%) with ΣLMW-PAHs/ΣHMW-PAHs of 1.4. Sample B was dominated by pyrene (10.6%), naphthalene (10.2%) and benzo[b+k+k]fluoranthene (8.1%) with ΣLMW-PAHs/ΣHMW-PAHs of 0.65. The most abundant PAHs in sample C were naphthalene (13%), cyclopenta[def]phenanthrene (11%) and pyrene (8%) with ΣLMW-PAHs/ΣHMW-PAHs of 1.08. Sample D* were dominated by acenaphthene (11.2%), phenanthrene (9.9%) and retene (9.8%) while sample E* was dominated by naphthalene (14.4%), retene (12.6%) and phenanthrene (9.6%). The ΣLMW-PAHs/ΣHMW-PAHs were 1.85 and 2.30 for samples D and E, respectively (Table S3). A previous study showed that the concentration of pyrene emitted from Chinese style cooking was higher than emitted from other cooking styles (e.g. Japanese) (He et al., 2004).

The composition pattern of OPAHs mixtures differed in the various samples (Figure 1). Sample A was dominated by 1,4-chysenequinone (21%), 1-indanone (11.4%) and 9-fluorenone (9.5%), Sample B on the other hand was dominated by 1-indanone (14.4%), 1,4-chysenequinone (13.8%) and 6H-benzo(cd)pyren-6-one (8.3%). 9-fluorenone (14%), 9,10-anthraquinone (12%) and 2-methylanthracene-9,10-dione (9.9%) were the dominant OPAHs in sample C. Sample D* was dominated by 1-indanone (19%), 1,4-naphthoquinone (9.9%), 2-methyl-anthracene-9,10-dione (14%) while sample E* was dominated by 1-indanone (20.2%), 9-fluorenone (13.2%) and 1,4-anthraquinone (12.2%). Many of these found can also be found in other environmental compartments such as ambient air and soil (Clergé et al., 2019). The individual OPAH/parent-PAH ratio was in most cases highest in sample C. The 9,10-anthraquinone/anthracene ratio was >1 for some samples (Table S3). The concentrations of Σ17OPAHs was significantly correlated with Σ30PAHs ($r = 0.91$, $p < 0.05$). Many individual OPAHs were also significantly correlated with their related parent-PAHs. This can be explained by their similar sources and fate.

The four individual AZAs targeted in this study were identified and quantified in each of the five samples (Table 3). Sample C had the highest average concentration of the Σ4AZAs, which is similar to all the other PAC groups (Table 3). The contribution of individual AZAs to the Σ4AZAs concentration was slightly different for each of the samples (Figure 1). For sample A, the highest contributions were by carbazole (48%) and quinoline (32%). Quinoline (45%) and benzo[h]quinoline (24%) were the dominant AZAs in sample B. Sample C was dominated by quinoline (38.4%) and carbazole (32.4%). This same trend was in sample D* and E* in which the highest contribution was from quinoline, followed by carbazole (Figure 1). The substances are toxicologically relevant with carbazole being classified as possible human carcinogen (IARC, 2011a; Yamada et al., 2004). None of the samples had individual AZA/parent-PAH concentration ratio >1. The concentration ratios of individual AZA/parent-PAH were mostly < 10%, except in a few samples in which the acridine/anthracene and carbazole/fluorene concentration ratio were ≥ 10% (Table S3). These ratios were thus generally lower than the individual OPAH/parent-PAH and individual alkylated PAH/parent-PAH concentration ratios (Table S3). The Σ4AZAs were also significantly correlated with Σ30PAHs ($r=0.83$, $p <0.05$). This can be explained by the sources and fate of AZAs are similar to those of the PAHs and OPAHs (Bandowe et al., 2016).

3.4 Concentration of carbonyls

The average concentrations of high-molecular-weight (HMW) mono-carbonyl ($C \geq 6$) and di-carbonyl compounds (glyoxal and methylglyoxal) in $PM_{2.5}$ are shown in Table S4. Sample C (5327.6±1974.1 ng/m^3) showed the highest total concentration of HMW mono-carbonyl and di-carbonyl compounds, whereas sample B recorded the lowest (159.2±48.7 ng/m^3). Nonanaldehyde was the most abundant component in all samples, accounting for ~31-81% of total carbonyl compounds (Figure S4). The contribution of nonanaldehyde to the carbonyl mixtures was > 80% in samples A and C. The contribution of each of the other carbonyl compounds to the total mixture was < 10% in sample A and C. Nonaldehyde was also the most dominant contributor to the total carbonyl compounds in the other three sampling locations (B, D*, E*) but at these sites methylgloxal also made a high contribution (> 10%). Nonanaldehyde was typically identified as a dominant carbonyl component in cooking that involves usage of edible oils (Ho et al., 2006). The presence of nonanaldehyde could be due to the decomposition of 9-octadecenoic acid (oleic acid), a known fatty acid produced from cooking oil thermal decomposition (Schauer et al., 2002). A previous study showed that kitchens involved with frequent frying activities (e.g. western fast-food chain shops and Korean barbecue restaurant) could be more abundant in nonanaldehyde (Ho et al., 2006). The two dicarbonyl compounds were detected in all sampling locations, accounting for ~0.6-26.1% of total carbonyl compounds. Sample A and C showed similar glyoxal (0.7% and 0.6%) and methylglyoxal (3.3% and 2.9%) in their compositions. The highest contribution of decaldehyde to the carbonyl mixtures was observed in sample D* (10.5%) with much lower contributions (0.7-4.1%) in the other samples (Figure S4). This observation could be attributed to the types of oils usage in the cooking processes. A previous study showed that different types of seed oils (e.g. soybean and canola oil) could generate ~4.77 times difference of decaldehyde in emissions (Schauer et al., 2002), although further study is necessary. The results show that cooking activities are significant anthropogenic source of semi-volatile aldehydes.

3.5 Relationships between chemical species

There were significant correlations between the concentrations of many individual and sums of chemical species (Table S5). For example, the significant correlations between the Σ30PAHs, Σ17OPAHs and Σ4AZAs and the $PM_{2.5}$ mass, TC, EC, OC and Σcarbonyls can be attributed to their similar sources, the fact that they are co-sorbed with each other and hence have similar fates. Some ions like Cl^- and several metals were also strongly correlated with PAHs, OPAHs and AZAs, which might also be strong indication of their similarity in sources but especially similarity in their fates. Unlike Cl^- the correlations between the PACs and SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} were not statistically significant indicating that the sources and fate of these ions might be much more different to that of the PACs.

3.6 Bioreactivity of $PM_{2.5}$

The cell viability, oxidative potential (ROS generation), inflammatory reactions (TNF-α) and oxidative DNA damage elicited by $PM_{2.5}$ collected from different cooking sites are shown in Figures 2 and Table S2 (i.e. oxidative DNA damage under particle concentrations of 50, 100, 500 and 1000 µg/ml dosage). The cell viability of A549 cells demonstrates negative dose-response from all samples (Figure 2). Under the particle concentration of 200 µg/ml, it can be observed that sample C and D* further showed lowest cell viabilities. Positive dose-response was nevertheless identified in ROS generation and TNF-α (Figure 2) suggesting that oxidative and inflammatory reactions could be enhanced by the increase of particle concentration. Sample C shows the highest ROS generation followed by sample B, whereas sample D* and E* demonstrated higher oxidative potential (under 200 µg/ml dosage) in comparison with other samples.

A general increasing trend between particle dose concentration and DNA damage was observed in most sub-samples (except for the sub-sample 1-4 of sample A, Table S2 and Figure S6). The amount of damage to the plasmid DNA induced by $PM_{2.5}$ varied over the range of 2.3-65.8% (Figure S5) in the samples under 1000 µg/ml dosage. These findings are relatively low compared to the result from a study on PM_{10} derived from coal burning (0-55% under 500 µg/ml) (Shao et al., 2016).

Median lethal dose (LD_{50}) of the samples (Figure S5) were determined by dosage response analysis in Figure S6 (Supplementary Material). Sample A showed highest LD_{50} and the lowest LD_{50} was in sample E*. The results indicate that lower $PM_{2.5}$ concentration was required to cause 50% DNA damage for sample E* compared to the other samples. Both sample B and C show comparable DNA damage in this study. This observation could be due to similar cooking characteristics.

Chemical species and elements some of which have been quantified (e.g. heterocyclic amines, metals, and PACs) in emissions from cooking could cause oxidative stress, cell injury, DNA damage and mutations, which could result in the above observed effects and risk of diseases such as lung cancer, cardiovascular and pulmonary diseases (Pardo et al., 2015; Gerlofs-Nijland et al., 2009; Wei et al., 2009; Xue and Warshawsky, 2005; Bolton et al., 2000; Seow et al., 2000).

3.7 Correlation between chemical components and bioreactivities

Cell viability showed significant negative correlations with the concentrations of benzo[a]anthracene, indeno [1,2,3-cd]pyrene and 1,4-naphthoquinone in the samples (Table 4). These relationships suggest that these compounds might contribute to the toxicity of extracts of the $PM_{2.5}$ samples emitted from cooking activities.

Significant positive correlations were found between the ROS levels and the concentrations Σ17OPAHs in addition to several individual OPAHs (2-biphenlcarboxaldehyde, 1-acenaphthenone, 9-fluorenone, 9,10-anthraquinone, 2-methylanthraquinone, benzo[a]fluorenone), acridine and decaldehyde (Table 4 and Figure S7). The results of our study are consistent with other studies that show that OPAHs (and other quinones) are redox active chemical species in air PM that are associated with particle induced oxidative potential (Gao et al., 2020; Tuet et al., 2019; Sheng and Lu, 2017; Shang et al., 2013; Bolton et al., 2000).

None of the PAHs or alkyl-PAHs were significantly positively correlated to ROS generation (oxidative potential), in this study (Table 4). Positive correlations between the concentrations of PAHs in PM and cellular ROS activity (after exposure to extracts of the PM) has been reported (Tuet et al., 2019; Daher et al., 2012; Hu et al., 2008). PAHs and alkyl PAHs can only participate in reactions leading to ROS production after their biological transformation to redox active compounds (Verma et al., 2011; Ntziachristos et al., 2007; Bolton et al., 2000). The lack of significant positive correlation between ROS generation and the concentration of PAHs in our study might be explained by the mismatches between the PAHs extracted from the filters and the fraction of PAHs that is bioavailable/bioaccessible to the human alveolar epithelial cells (A549) for bioreactivity (Li et al., 2019; Baulig et al., 2004).

None of the (transition) metals which have also been identified as strong inducers of ROS formation were significantly positively correlated with ROS generation in this study (Gao et al., 2020; Bates et al., 2019; Daher et al., 2014; Verma et al., 2009; Hu et al., 2008). It is important to note that the method for determining the concentration of metals in our study (ED-XRF, see Text S2) measures the total concentration and not the water-soluble (bioavailable/bioaccessible fractions). It is the water-soluble concentrations (not the total concentrations) of some transition metals in fine particulate matter that are found to have positive correlation with ROS generation in cellular assays (Shafer et al., 2016; Daher et al., 2014; Verma et al., 2009; Hu et al., 2008).

Strong positive correlations were also found between TNF-α and the concentration of several chemical species (EC, Ca^{2+}, SO_4^{2-}, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Ni, 14 individual PAHs and Σ30PAHs; Table 4 and Figure S7). The TNF-α was however significantly negatively correlated with the concentration of more polar PAH derivatives (Σ4AZAs and Σ30OPAHs). This is despite the fact that the concentrations of PAH derivatives were positively correlated with the concentrations of PAHs. This suggests that the impact of PAHs extracted from the $PM_{2.5}$ on the generation of pro-inflammatory cytokines was opposite to the impact of PAH derivatives (OPAHs and AZAs). Most of the PAHs showing strongest positive correlations with TNF-α are those with 4-6 ring sizes (HMW-PAHs) with several of them classified as probable human carcinogens. Our study suggests that inflammation is a strong pathway for the toxicity of $PM_{2.5}$ emitted from cooking emissions and that several chemical species including PAHs, transition metals might be the triggers for the inflammatory response. Relationships between inflammation and metal/PAH contents of PM has been reported in other studies (Pardo et al., 2015; Gerlofs-Nijland et al., 2009).

Significant positive correlation was observed between DNA damage and concentrations of several elements (Al, Ti, Cr and Mn), Σ30PAHs and individual PAHs (1,2,3,4-tetrahydronaphthalene, naphthalene, acenaphthene, dibenzo[a,h]anthracene, perylene, benzo[a]pyrene, benzo[b+j+k]fluoranthene and benzo[ghi]perylene) (Table 4). PAHs are known mutagens and carcinogens and damage to DNA is a known mechanism for their carcinogenic effect (Wei et al., 2009; IARC 2005; Bolton et al., 2000). A previous study showed cytotoxicity of Cr in Chinese commercial cooking $PM_{2.5}$ emissions (Sun et al., 2020).

4. Conclusions

This study investigated $PM_{2.5}$ emissions generated from different cooking conditions. The concentrations of nearly all chemical species were highest in one restaurant that served Cantonese dishes and applied oil frying methods, suggesting the cooking ingredients and conditions could be the determining factors for higher emissions. The extracts from the samples elicited toxicologically relevant responses (cell death, ROS generation, inflammation activity and DNA damage). The responses were in dose dependent manner and demonstrated higher responses under higher $PM_{2.5}$ concentrations. These responses were correlated to concentrations of specific chemical species in the $PM_{2.5}$ suggesting that some of the determined chemical species might play roles in the toxicity of $PM_{2.5}$ emitted from cooking activities.

Acknowledgments

This study was supported by the Research Grants Council of the Hong Kong Special Administrative Region China (General Research Fund, Project number: 14205318).

References

Abdullahi, K.L., Delgado-Saborit, J.M., Harrison, R.M., 2013. Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: a review. Atmospheric Environment 71: 260-294. https://doi.org/10.1016/j.atmosenv.2013.01.061.

Allan, J.D., Williams, P.I., Morgan, W.T., Martin, C.L., Flynn, M.J., Lee, J., Nemitz, E., Phillips, G.J., Gallagher, M.W., Coe, H., 2010. Contributions from transport, solid fuel burning and cooking to primary organic aerosols in two UK cities. Atmospheric Chemistry and Physics 10: 647-668. https://doi.org/10.5194/acp-10-647-2010.

Bandowe, B.A.M., Meusel, H., Huang, R., Ho, K., Cao, J., Hoffman, T., Wilcke, W., 2014. PM2.5-bound oxygenated PAHs, 2014. PM2.5-bound oxygenated PAHs, nitro-PAHs and parent-PAHs from the atmosphere of a Chinese megacity:seasonal variation, sources and cancer risk assessment. Science of the Total Environment 473-474, 77-87

Bandowe, B.A.M., Meusel, H., Huang, R., Hoffmann, T., Cao, J., Ho, K., 2016. Azaarenes in fine particulate matter from the atmosphere of a Chinese megacity. Environmental Science and Pollution Research 23, 16025-16036 https://doi.org/10.1007/s11356-016-6740-z.

Baulig, A., Poirault, J.J., Ausset, P., Schins, R., Shi, T., Baralle, D., Dorlhene, P., Meyer, M., Lefevre, R., Baeza-Squiban A., Marano, F., 2004. Physicochemical characteristics and biological activities of seasonal atmospheric particulate matter sampling in two locations of Paris. Environmental Science and Technology 38: 5985-5992. https://doi.org/10.1021/es049476z.

Bein, K.J., Wexler, A.S., 2015. Compositional variance in extracted particulate matter using different filter extraction techniques. Atmospheric Environment 107: 24-34. https://doi.org/10.1016/j.atmosenv.2015.02.026.

Bitterle, E., Karg, E., Schroeppel, A., Kreyling, W.G., Tippe, A., Ferron, G.A., Schmid, O., Heyder, J., Maier, K.L., Hofer, T., 2006. Dose-controlled exposure of A549 epithelial cells at the air–liquid interface to airborne ultrafine carbonaceous particles. Chemosphere 65: 1784-1790. https://doi.org/10.1016/j.chemosphere.2006.04.035.

Blaszczyk, U., Janoszka, B., 2008. Analysis of azaarenes in pan fried meat and its gravy by liquid chromatography with fluorescence. Food Chemistry 109, 235-242. https://doi.org/10.1016/j.foodchem.2007.12.038.

Bolton, J.L, Trush, M.A., Penning, T.M., Dryhurst, G., Monks, T.J., 2000. Role of quinones in toxicology. Chemical Research in Toxicology 13, 135-160. https://doi.org/10.1021/tx9902082.

Boström, C-E., Gerde, P., Hanberg, A., Jernström, B., Johansson, C., Kyrklund, T., Rannug, A., Törnqvist, M., Victorin, K., Westerholm, R., 2002. Cancer risk assessment, indicators, and guidelines for polycyclic aromatic hydrocarbons in the ambient air. Environmental health perspectives 110: 451. https://doi.org/10.1289/ehp.110-1241197.

Buonanno, G., Morawska, L., Stabile, L., 2009. Particle emission factors during cooking activities. Atmospheric Environment 43: 3235-3242. https://doi.org/10.1016/j.atmosenv.2009.03.044.

Butnariu, M., Butu, A, 2014. Chemical composition of vegetables and their products. Handbook of Food Chemistry. Springer-Verlag, Berlin Heidelberg. pp. 1-49. https://link.springer.com/content/pdf/10.1007%2F978-3-642-41609-5_17-1.pdf

Charrier, J.G., McFall, A.S., Richards-Henderson, N.K., Anastasio, C., 2014. Hydrogen peroxide formation in a surrogate lung fluid by transition metals and quinones present in particulate matter. Environmental Science & Technology 48: 7010-7017. https://doi.org/10.1021/es501011w.

Cheng, S., Wang, G., Lang, J., Wen, W., Wang, X., Yao, S., 2016. Characterization of volatile organic compounds from different cooking emissions. Atmospheric Environment 145: 299-307. https://doi.org/10.1016/j.atmosenv.2016.09.037.

Chiang, T-A., Wu, P-F., Wang, L-F., Lee, H., Lee, C-H., Ko, Y-C., 1997. Mutagenicity and polycyclic aromatic hydrocarbon content of fumes from heated cooking oils produced in Taiwan. Mutation Research/Fundamental and Molecular Mechanisms of Mutagenesis 381: 157-161. https://doi.org/10.1016/S0027-5107(97)00163-2.

Chuang, H-C., BéruBé, K., Lung, S-CC., Bai, K-J., Jones, T., 2013. Investigation into the oxidative potential generated by the formation of particulate matter from incense combustion. Journal of hazardous materials 244: 142-150. https://doi.org/10.1016/j.jhazmat.2012.11.034.

Clergé, A., Le Goff, J., Lopez, C., Ledauphin, J., Delépée, R., 2019. Oxy-PAHs: occurrence in the environment and potential genotoxic/mutagenic risk assessment for human health. Critical Reviews in Toxicology 49, 302-328. https://doi.org/10.1080/10408444.2019.1605333.

Cobos, A., Diaz, O, 2014. Chemical composition of meat and meat products. Handbook of Food Chemistry. Springer-Verlag, Berlin-Heldelberg pp 1-22. https://link.springer.com/content/pdf/10.1007%2F978-3-642-41609-5_6-1.pdf.

Daher, N., Ruprecht, A., Invernizzi, G., De Marco, C., Miller-Schulze, J., Heo, J.B., Shafer, M.M., Shelton, B.R., Schauer, J.J., Sioutas. C., 2012. Characterization, sources and redox activity of fine and coarse particulate matter in Milan, Italy. Atmospheric Environment 49: 130-141. https://doi.org/10.1016/j.atmosenv.2011.12.011.

Daher, N., Saliba, N.A., Shihadeh, A.L., Jaafar, M., Baalbaki, R., Shafer, M.M., Schauer, J.J., Sioutas. C., 2014. Oxidative potential and chemical speciation of size-resolved particulate matter (PM) at near-freeway and urban background sites in the greater Beirut area. Science of the Total Environment 470-471: 417-426. https://doi.org/10.1016/j.scitotenv.2013.09.104.

Danielsen, P.G., Møller P., Jensen, K.A., Sharma, A.K., Wallin, H., Bossi, R., Autrup, H., Mølhave, L,. Ravanat, J.L, Briedé, J.J., de Kok, T.M., Loft, S., 2011. Oxidative stress, DNA Damage, and inflammation induced by Air and wood smoke particulate matter in Human A549 and THP-1 Cell lines. Chemical Research in Toxicology 24, 168-184. https://doi.org/10.1021/tx100407m.

De Oliveira, L.C.C., Vieira, M.A., Ribeiro, A.S., Lisboa, M.T., Gonçalves, R.A., De Campos, R.C., 2012. Determination of silicon in vegetable oil and biodiesel by high-resolution continuum source flame atomic absorption spectrometry using sample dilution with xylene. Energy & Fuels 26: 7041-7044. https://doi.org/10.1021/ef3012867.

Ding, J., Zhong, J., Yang, Y., Li, B., Shen, G., Su, Y., Wang, C., Li, W., Shen, H. Wang, B., Wang, R., Huang, Y., Zhang, Y., Cao, H., Zhu, Y., Simonich, S.L.M., Tao, S., 2012. Occurrence and exposure to polycyclic aromatic hydrocarbons and their derivatives in a rural Chinese home through biomass fuelled cooking. Environmental Pollution 169: 160-166. https://doi.org/10.1016/j.envpol.2011.10.008.

Epstein, E., 1999. Silicon. Annual Review of Plant Physiology and Plant Biology 50, 641-664. https://doi.org/10.1146/annurev.arplant.50.1.641.

Fortmann, R., Kariher, P., Clayton, R., 2001. Indoor air quality: residential cooking exposures final report. https://hero.epa.gov/hero/index.cfm/reference/details/reference_id/90689.

Fullana, A., Carbonell-Barrachina, Á.A., Sidhu, S., 2004. Comparison of volatile aldehydes present in the cooking fumes of extra virgin olive, olive, and canola oils. Journal of Agricultural and Food Chemistry 52: 5207-5214. https://doi.org/10.1021/jf035241f.

Fullana, A., Carbonell ‐ Barrachina, Á.A., Sidhu S., 2004. Volatile aldehyde emissions from heated cooking oils. Journal of the Science of Food and Agriculture 84: 2015-2021. https://doi.org/10.1002/jsfa.1904.

Gao, D., Ripleyb, S., Weichenthalb, S., Godri Pollitt, J.K., 2020. Ambient particulate matter oxidative potential: chemical determinants, associated health effects, and strategies for risk management. Free Radical Biology and Medicine 151, 7-25. https://doi.org/10.1016/j.freeradbiomed.2020.04.028.

Gerlofs-Nijland, M.E., Rummelhard, M., Boere, A.J.F., Leseman, D.L.A.C., Duffin, R., Schins, R.P.F., Borm, P.J.A., Sillanpää, M., Salonen, R.O., Cassee, F.R., 2009. Particle induced toxicity in relation to transition metal and polycyclic aromatic hydrocarbon content. Environmental Science and Technology 43, 4729-4736. https://doi.org/10.1021/es803176k.

Gysel, N., Dixit, P., Schmitz, D.A., Engling, G., Cho, A.K., Cocker, D.R., Karavalakis, G., 2018. Chemical speciation, including polycyclic aromatic hydrocarbons (PAHs), and toxicity of particles emitted from meat cooking operations. Science of The Total Environment 633: 1429-1436. https://doi.org/10.1016/j.scitotenv.2018.03.318.

Gysel, N., Welch, W.A., Chen, C-L., Dixit, P., Cocker III, D.R., Karavalakis, G., 2017. Particulate matter emissions and gaseous air toxic pollutants from commercial meat cooking operations. Journal of Environmental Sciences 65: 162-170. https://doi.org/10.1016/j.jes.2017.03.022.

He, L-Y., Hu, M., Huang, X-F., Yu, B-D., Zhang, Y-H., Liu, D-Q., 2004. Measurement of emissions of fine particulate organic matter from Chinese cooking. Atmospheric Environment 38: 6557-6564. https://doi.org/10.1016/j.atmosenv.2004.08.034.

Ho, S.S.H., Yu, J.Z., Chu, K.W., Yeung, L.L., 2006. Carbonyl emissions from commercial cooking sources in Hong Kong. Journal of the Air & Waste Management Association 56: 1091-1098. https://doi.org/10.1080/10473289.2006.10464532.

Hu, S., Polidori, A., Arhami, M., Shafer, M.M., Schauer, J.J., Cho, A., Sioutas, C., 2008. Redox activity and chemical speciation of size fractionated PM in the communities of the Los Angeles-Long Beach harbor. Atmospheric Chemistry and Physics 8: 6439-6451. https://doi.org/10.5194/acp-8-6439-2008.

Huang, Y., Ho, S.S.H., Ho, K.F., Lee, S.C., Yu, J.Z., Louie, P.K.K., 2011. Characteristics and health impacts of VOCs and carbonyls associated with residential cooking activities in Hong Kong. Journal of hazardous materials 186: 344-351. https://doi.org/10.1016/j.jhazmat.2010.11.003.

IARC, 2010. Some non-heterocyclic polycyclic aromatic hydrocarbons and some related exposures. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Volume 92, Lyon France. https://monographs.iarc.who.int/wp-content/uploads/2018/06/mono92.pdf

IARC, 2011a. Bitumens and bitumen emissions, and some N- and S-heterocyclic aromatic hydrocarbons. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans 103, 1-311.https://publications.iarc.fr/Book-And-Report-Series/Iarc-Monographs-On-The-Identification-Of-Carcinogenic-Hazards-To-Humans/Bitumens-And-Bitumen-Emissions-And-Some-Em-N-Em---And-Em-S-Em--Heterocyclic-Polycyclic-Aromatic-Hydrocarbons-2013

IARC, 2011b. Some chemicals present in industrial and consumer products, food and drinking water. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Volume 101, Lyon France. https://monographs.iarc.who.int/wp-content/uploads/2018/06/mono101.pdf

Jung, D.K., Klaus, T., Fent, K., 2001. Cytochrome P450 induction by nitrated polycyclic aromatic hydrocarbons, azaarenes, and binary mixtures in fish hepatoma cell line PLHC-1. Environmental Toxicology and Chemistry 20, 149-159. https://doi.org/10.1002/etc.5620200117.

Kebata-Pendias, A., Mukherjee, A.B., 2007. Trace elements from Soil to Humans. Springer-Verlag Berlin Heildelberg. https://link.springer.com/content/pdf/10.1007%2F978-3-540-32714-1.pdf.

Kelly, F.J., 2003. Oxidative stress: its role in air pollution and adverse health effects. Occupational and Environmental Medicine 60: 612-616. http://dx.doi.org/10.1136/oem.60.8.612.

Kuligowski, J., Halperin, K.M., 1992. Stainless steel cookware as a significant source of nickel, chromium, and iron. Archives of environmental contamination and toxicology 23: 211-215. https://doi.org/10.1007/BF00212277

Li, C-T., Lin, Y-C., Lee, W-J., Tsai, P-J., 2003. Emission of polycyclic aromatic hydrocarbons and their carcinogenic potencies from cooking sources to the urban atmosphere. Environmental Health Perspectives 111: 483. https://doi.org/10.1289/ehp.5518.

Li, Y-C., Qiu, J-Q., Shu, M., Ho, S.S.H., Cao, J-J., Wang, G-H., Wang, X-X., Zhao, X-Q., 2018. Characteristics of polycyclic aromatic hydrocarbons in PM 2.5 emitted from different cooking activities in China. Environmental Science and Pollution Research 25: 4750-4760. https://doi.org/10.1007/s11356-017-0603-0.

Li Y-C., Shu, M., Ho, S.S.H., Wang, C., Cao, J-J,, Wang, G-H., Wang, X-X. Wang, K., Zhao, X-Q.,2015 Characteristics of PM2. 5 emitted from different cooking activities in China. Atmospheric Research 166: 83-91. https://doi.org/10.1016/j.atmosres.2015.06.010.

Li, G., Wu, S., Zeng, J., Wang, L., Yu, W., 2016. Effect of frying and aluminium on the levels and migration of parent and oxygenated PAH in a popular Chinese fried bread youtiao. Food Chemistry 209, 123-130. https://doi.org/10.1016/j.foodchem.2016.04.036.

Lin, J-M., Liou, S-J., 2000. Aliphatic aldehydes produced by heating Chinese cooking oils. Bulletin of environmental contamination and toxicology 64: 817-824. https://doi.org/10.1007/s0012800076.

Louie, P.K.K., Chow, J.C., Antony Chen, L.-W., Watson, J.G., Leung, G., Sin, D.W.M, 2005. PM2.5 chemical composition in Hong Kong: urban and regional variations. Science of the Total Environment 338, 267-281. https://doi.org/10.1016/j.scitotenv.2004.07.021.

Lui, K.H., Bandowe, B.A.M et al., 2016. Characterization of the chemical components and bioreactivity of fine particulate matter ($PM_{2.5}$) during incense burning. Environmental Pollution 213, 524-533.

Martorelli, I., Perello, G., Martí-Cid, R., Castell, V., Llobet, J.M., Domingo, J.L., 2010. Polycyclic aromatic hydrocarbons (PAHs) in foods and estimated intake by the population of Catalonia, Spain Temporal trend. Environment International 36, 424-432. https://doi.org/10.1016/j.envint.2010.03.003

McDonald, J.D., Zielinska, B., Fujita, E.M., Sagebiel, J.C., Chow, J.C., Watson, J.G.,2003. Emissions from charbroiling and grilling of chicken and beef. Journal of the Air & Waste Management Association 53: 185-194. https://doi.org/10.1080/10473289.2003.10466141.

Metayer, C., Wang, Z., Kleinerman, R.A., Wang, L., Brenner, A.V., Cui, H., Cao J., Lubin, J.H.,2002. Cooking oil fumes and risk of lung cancer in women in rural Gansu, China. Lung Cancer 35: 111-117. https://doi.org/10.1016/S0169-5002(01)00412-3.

Mohr, C., DeCarlo, P.F., Heringa, M.F., Chirico, R., Slowik, J.G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J.L., Crippa, M., Zimmermann, R., Baltensperge, U., Prévôt, A.S.H., 2012. Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data. Atmospheric Chemistry and Physics 12: 1649-1665. https://doi.org/10.5194/acp-12-1649-2012

Moret, S., Conte, L.S., 2000. Polycyclic aromatic hydrocarbons in edible fats and oils: occurrence and analytical methods. Journal of chromatography A 882: 245-253. https://doi.org/10.1016/S0021-9673(00)00079-0.

Ntziachristos, L., Froines, J.R., Cho, A.K., Sioutas, C., 2007. Relationship between redox activity and chemical composition of size-fractionated particulate matter. Particle and Fibre Toxicology 4:5.

Nolte, C.G., Schauer, J.J., Cass, G.R., Simoneit, B.R.T., 1999. Highly polar organic compounds present in meat smoke. Environmental Science & Technology 33: 3313-3316. https://doi.org/10.1021/es990122v.
Pardo, M., Shafer, M.M., Rudich, A., Schauer, J.J., Rudich, Y, 2015. Single exposure to near Roadway particulate matter leads to confined inflammatory and defence responses: possible role of metals. Environmental Science and Technology 49, 8777-8785. https://doi.org/10.1021/acs.est.5b01449
Qu, Y.H., Xu, G.X., Zhou, J.Z., Chen, T.D., Zhu, L.F., Shields, P.G., Wang, H.W., Gao, Y.T., 1992. Genotoxicity of heated cooking oil vapors. Mutation Research/Genetic Toxicology 298: 105-111. https://doi.org/10.1016/0165-1218(92)90035-X.
Robberect, H., Van Dyck, K., Bosscher, D., Van Cauwenbergh, R., 2008. Silicon in Foods: content and bioavailability. International Journal of Food Properties 11, 638-645. https://www.tandfonline.com/doi/full/10.1080/10942910701584252.
Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T., 1991. Sources of fine organic aerosol. 1. Charbroilers and meat cooking operations. Environmental Science & Technology 25: 1112-1125. https://doi.org/10.1021/es00018a015.
Roper, C., Chubb, L.G., Cambal, L., Tunno, B., Clougherty, J.E., Fattman, C., Mischler, S.E., 2017. Association of IL-6 with PM 2.5 Components: Importance of Characterizing Filter-Based PM 2.5 Following Extraction. Water, Air, & Soil Pollution 228: 43. https://doi.org/10.1007/s11270-016-3219-y.
Roper, C., Delgado, L.S., Barrett, D., Massey Simonich, S.L., Tanguay, R.L., 2018. PM2. 5 Filter Extraction Methods: Implications for Chemical and Toxicological Analyses. Environmental science & technology 53: 434-442. https://doi.org/10.1021/acs.est.8b04308.
Saito, E., Tanaka, N., Miyazaki, A., Tsuzaki, M., 2014. Concentration and particle size distribution of polycyclic aromatic hydrocarbons formed by thermal cooking. Food chemistry 153: 285-291. https://doi.org/10.1016/j.foodchem.2013.12.055.
Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 1999a. Measurement of emissions from air pollution sources. 1. C1 through C29 organic compounds from meat charbroiling. Environmental Science & Technology 33: 1566-1577. https://doi.org/10.1021/es980076j
Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 2002. Measurement of emissions from air pollution sources. 4. C1− C27 organic compounds from cooking with seed oils. Environmental Science & Technology 2002; 36: 567-575. https://doi.org/10.1021/es002053m.
See, S.W., Balasubramanian, R., 2006. Risk assessment of exposure to indoor aerosols associated with Chinese cooking. Environmental research 102: 197-204. https://doi.org/10.1016/j.envres.2005.12.013.
See, S.W., Balasubramanian, R., 2008. Chemical characteristics of fine particles emitted from different gas cooking methods. Atmospheric Environment 42: 8852-8862. https://doi.org/10.1016/j.atmosenv.2008.09.011.
See, S.W., Karthikeyan, S., Balasubramanian, R., 2006. Health risk assessment of occupational exposure to particulate-phase polycyclic aromatic hydrocarbons associated with Chinese, Malay and Indian cooking. Journal of Environmental Monitoring 8: 369-376. https://doi.org/10.1039/B516173H
Seow, A., Poh, W.T., Teh, M., Eng, P., Wang, Y.T., Tan, W.C., Yu, M.C. Lee, H.P., 2000. Fumes from meat cooking and lung cancer risk in Chinese women. Cancer Epidemiology and Prevention Biomarkers 2000; 9: 1215-1221. https://cebp.aacrjournals.org/content/9/11/1215.
Shafer, M.M., Hemming, J.D.C., Antkiewicz, D.S., Schauer, J.J., 2016. Oxidative potential of size-fractionated atmospheric aerosol in urban and rural sites across Europe. Faraday Discussions 189: 381-405. https://doi.org/10.1039/C5FD00196J.
Shang, Y., Fan, L., Feng, J., Lv, S., Wu, M., Li, B., Zang, Y.S., 2013. Genotoxic and inflammatory effects of organic extracts from traffic-related particulate matter in human lung epithelial A549 cells. The role of quinones. Toxicology in Vitro 27: 922-931. https://doi.org/10.1016/j.tiv.2013.01.008.
Shao, L., Hou, C., Geng, C., Liu, J., Hu, Y., Wang, J., Jones, T., Zhao, C., BéruBé, K., 2016. The oxidative potential of PM10 from coal, briquettes and wood charcoal burnt in an experimental

domestic stove. Atmospheric Environment 127: 372-381. https://doi.org/10.1016/j.atmosenv.2015.12.007.

Shao, L., Hu, Y., Shen, R., Schäfer, K., Wang, J., Wang, J., Schnelle-Kreis, J., Zimmermann, R., BéruBé, K., Suppan, P., 2017. Seasonal variation of particle-induced oxidative potential of airborne particulate matter in Beijing. Science of The Total Environment 579: 1152-1160. https://doi.org/10.1016/j.scitotenv.2016.11.094.

Shao, L., Shi, Z., Jones, T.P., Li, J., Whittaker, A.G., Berube, K.A., 2006. Bioreactivity of particulate matter in Beijing air: results from plasmid DNA assay. Science of the Total Environment 367: 261-272. https://doi.org/10.1016/j.scitotenv.2005.10.009.

Sheng, K., Lu, J., 2017. Typical airborne quinones modulate oxidative stress and cytokine expression in lung epithelial A549 cells. Journal of Environmental Science and Health, Part A 52: 127-134. https://doi.org/10.1080/10934529.2016.1237127.

Shields, P.G., Xu, G.X., Blot, W.J., Fraumeni Jr, J.F., Trivers, G.E., Pellizzari, E.D., Qu, Y.H., Gao, Y.T., Harris, C.C., 1995. Mutagens from heated Chinese and US cooking oils. Journal of the National Cancer Institute 87: 836-841. https://doi.org/10.1093/jnci/87.11.836.

Sovadinová, I., Bláha, L., Janošek, J., Hilcherová, K., Giessy, J.P., Jones, P.D., Holoubek, I., 2006. Cytotoxicity and aryl hydrocarbon receptor-mediated activity of N-heterocyclic polycyclic aromatic hydrocarbons: structure-activity relationships. Environmental Toxicology and Chemistry 25, 1291-1297. https://doi.org/10.1897/05-388R.1.

Sun, J., Shen, Z., Niu, X., Yu, J., Zhang, Y., Liu, S., Niu, X., Zhang, Y., Xu, H., Li, X.J.A.E., 2020. PM2. 5 source profiles from typical Chinese commercial cooking activities in northern China and its influences on bioreactivity of vascular smooth muscle cells (VSMCs). Environmental Pollution 239: 117750.

Svendsen, K., Jensen, H.N., Sivertsen, I., Sjaastad, A.K., 2002. Exposure to cooking fumes in restaurant kitchens in Norway. Annals of Occupational Hygiene 46: 395-400.

Szterk, A., 2015. Acridine derivatives (PANH, azaarenes) in raw, fried or grilled pork from different origins, and PANH formation during pork thermal processing. Journal of Food Composition and Analysis 43, 18-24. https://doi.org/10.1016/j.jfca.2015.04.011.

Tanaka, N., Ohtake, K., Tsuzaki, M., Miyazaki, A., 2012. Analysis of polycyclic aromatic hydrocarbons in oil-mist emitted from food grilling. Bunseki Kagaku 2012; 61: 77-86. https://doi.org/10.2116/bunsekikagaku.61.77.

Taner, S., Pekey, B., Pekey, H., 2013. Fine particulate matter in the indoor air of barbeque restaurants: Elemental compositions, sources and health risks. Science of the Total Environment 454: 79-87. https://doi.org/10.1016/j.scitotenv.2013.03.018

Torkmahalleh, M.A., Zhao, Y., Hopke, P.K., Rossner, A., Ferro, A.R., 2013. Additive impacts on particle emissions from heating low emitting cooking oils. Atmospheric environment 74: 194-198. https://doi.org/10.1016/j.atmosenv.2013.03.038.

Tuet, W.Y., Liu, F., de Oliveira Alves, N., Fok, S., Artaxo, P., Vasconcellos, P., Champion, J.A., Ng, N.L., 2019. Chemical oxidative potential and cellular oxidative stress from open biomass burning aerosol. Environmental Science and Technology Letters 6: 126-132. https://doi.org/10.1021/acs.estlett.9b00060.

Van Winkle, L.S., Bein, K., Anderson, D., Pinkerton, K.E., Tablin, F., Wilson, D., Wilson, D., Wexler, A.S., 2014. Biological dose response to PM2. 5: effect of particle extraction method on platelet and lung responses. Toxicological Sciences 143: 349-359. https://doi.org/10.1093/toxsci/kfu230.

Verma, V., Pakbin, P., Cheung, K.L., Cho, A.K., Schauer, J.J., Shafer, M.M., Kleinman, M.T., Sioutas, C., 2011. Physicochemical and oxidative characteristics of semi-volatile components of quasi-ultrafine particles in an urban atmosphere. Atmospheric Environment 45: 1025-1033. https://doi.org/10.1016/j.atmosenv.2010.10.044.

Wang, G., Cheng, S., Wei, W., Wen, W., Wang, X., Yao, S., 2015. Chemical characteristics of fine particles emitted from different Chinese cooking styles. Aerosol Air Qual. Res 15: 2357-2366. https://doi.org/10.4209/aaqr.2015.02.0079.

Wang, L., Xiang, Z., Stevanovic, S., Ristovski, Z., Salimi, F., Gao, J., Wang, H., Li, L., 2017. Role of Chinese cooking emissions on ambient air quality and human health. Science of the Total Environment 589: 173-181. https://doi.org/10.1016/j.scitotenv.2017.02.124.

Wei, Y., Han, I.-K., Shao, M., Hu, M., Zhang, J., Tang, X., 2009. PM2.5 constituents and oxidative DNA damage in Humans. Environmental Science and Technology 43, 4757-4762. https://doi.org/10.1021/es803337c.

Wu, P.F,, Chiang, T.A., Wang, L.F., Chang, C.S., Ko, Y.C., 1998. Nitro-polycyclic aromatic hydrocarbon contents of fumes from heated cooking oils and prevention of mutagenicity by catechin. Mutation Research/Fundamental and Molecular Mechanisms of Mutagenesis 403: 29-34. https://doi.org/10.1016/S0027-5107(98)00015-3.

Xue, W., Warshawsky, D., 2005. Metabolic activation of polycyclic and heterocyclic aromatic hydrocarbons and DNA damage: A review. Toxicology and Applied Pharmacology 206, 73-93. https://doi.org/10.1016/j.taap.2004.11.006.

Yamada, K., Suzuki, T., Kohara, A., Hayashi, M., Mizutani, T., Saeki, K-I., 2004. In vivo mutagenicity of benzo[f]quinoline, benzo[h]quinoline, and 1,7-phenanthroline using *lacZ* transgenic mice. Mutation Research 559, 83-95 https://doi.org/10.1016/j.mrgentox.2003.12.012.

Yang, A., Jedynska, A., Hellack, B., Kooter, I., Hoek, G., Brunekreef, B., Kuhlbusch, T.A.J., Cassee, F.R., Janssen, N.A.H., 2014. Measurement of the oxidative potential of PM2. 5 and its constituents: The effect of extraction solvent and filter type. Atmospheric environment 83: 35-42. https://doi.org/10.1016/j.atmosenv.2013.10.049.

Zhang, J., Smith, K.R., 1999. Emissions of carbonyl compounds from various cookstoves in China. Environmental Science & Technology 33: 2311-2320. https://doi.org/10.1021/es9812406.

Zhang, N., Han, B., He, F., Xu, J., Zhao, R., Zhang, Y., Bai, Z., 2017. Chemical characteristic of PM2. 5 emission and inhalational carcinogenic risk of domestic Chinese cooking. Environmental pollution 227: 24-30. https://doi.org/10.1016/j.envpol.2017.04.033

Zhao, Y., Hu, M., Slanina, S., Zhang, Y., 2007. Chemical compositions of fine particulate organic matter emitted from Chinese cooking. Environmental Science & Technology 41: 99-105. https://doi.org/10.1021/es0614518.

Zhong, L., Goldberg, M.S., Gao, Y-T., Jin, F., 1999a. Lung cancer and indoor air pollution arising from Chinese-style cooking among nonsmoking women living in Shanghai, China. Epidemiology 488-494. https://pubmed.ncbi.nlm.nih.gov/10468420/.

Zhong, L., Goldberg, M.S., Parent, M-É., Hanley, J.A., 1999b. Risk of developing lung cancer in relation to exposure to fumes from Chinese-style cooking. Scandinavian journal of work, environment & health 309-316. https://www.sjweh.fi/show_abstract.php?abstract_id=440.

Zhu, L., Wang, J., 2003. Sources and patterns of polycyclic aromatic hydrocarbons pollution in kitchen air, China. Chemosphere 50: 611-618. https://doi.org/10.1016/S0045-6535(02)00668-9.

List of Tables

Table 1: Characteristic of cooking operations (n = 4 for each type of operation).

Table 2: Average concentrations ± standard deviations of $PM_{2.5}$ (µg/m^3), TC (µg/m^3), OC (µg/m^3) and EC (µg/m^3) of inorganic elements (ng/m^3), water extractable ions (µg/m^3) in cooking emission samples.

Table 3: The average concentrations ± standard deviations (pg/m^3) of PAHs, OPAHs and AZAs in five sampling locations

Table 4: Spearman's rank correlation coefficients (R) of the cell viability, ROS generated (DCFH) and inflammatory activity (TNF-α & IL-6), and oxidative DNA damage elicited by extracts and the concentration chemical species

List of Figures and Figure Legends

Figure 1: Mean contributions of individual PAHs, OPAHs, and AZAs to the Σ30PAHs, Σ17OPAHs, and Σ4AZAs concentrations in different sampling locations. The y-axis was broken at 15% to enlarge the scale before the break. Error bars indicate standard deviations for each sample.

Figure 2: Cell viability, ROS generation (fluorescence intensity) and TNF-α induced by extracts of $PM_{2.5}$ from five sampling locations (*$p < 0.05$). Bars are the mean ±standard deviation.

Table 1: Characteristic of cooking operations (n = 4 for each type of operation).

Sample	Sampling location name	Seating capacity (seats)	Peak hour	Fuel type	Cooking style	Cooking method	Relative humidity[d] (%)	Temperature[d] (°C)
A	A core (theatre lounge)	80	12:30-14:30	Town gas, Electricity	Western cuisine (e.g. pasta, salad and rice)	Baking, Frying (pan frying, stir frying), Grilling, Steaming	77.1	21.6
B	Student canteen (communal student canteen)	520	12:00-14:00	Town gas, Electricity	Mixed cuisine (e.g. siu mei[b], hamburger and vegetarian diet)	Frying (pan frying, stir frying, deep frying), Roasting, Steaming	63.9	28.4
C	Chinese restaurant (communal student & staff restaurant)	450	12:00-13:00	Town gas, Electricity	Cantonese and local cuisine (noodles, congee, steamed rice and Chinese dim sum[c]	Frying (pan frying, stir frying, deep frying), Steaming	83.2	29.1
D*[a]	Environmental test chamber	Not available	Not available	Electricity	Stir-fried rice noodle	Stir frying	60.0	27.0
E*[a]	Environmental test chamber	Not available	Not available	Electricity	Fried chicken	Pan frying	60.0	27.0

[a] represents samples collected from stainless steel environmental constant temperature and humidity test chamber that mimic residential kitchen hood condition.
[b] Siu mei is the generic name in Cantonese cuisine given to meats roasted on spits over an open fire or a huge wood burning rotisserie oven.
[c] Dim sum is a style of Chinese cuisine prepared as small bite-sized portions of food served in small steamer baskets or on small plates.
[d] represents the sampling condition of exhaust system in restaurant and the test chamber.

Table 2: Average concentrations ± standard deviations of $PM_{2.5}$ (μg/m^3), TC (μg/m^3), OC (μg/m^3) and EC (μg/m^3) of inorganic elements (ng/m^3), water extractable ions (μg/m^3) in cooking emission samples.

Component[a]	Sample A	Sample B	Sample C	Sample D*	Sample E*
$PM_{2.5}$	234.5±22.1	177.4±50.6	711.5±222.6	354.1±60.8	492.0±257.9
TC	113.8±4.2	105.2±32.6	353.1±124.1	217.9±25.5	319.3±164.9
OC	108.3±4.2	99.9±31.5	338.8±121.1	215.0±25.1	315.5±164.4
EC	5.5±0.7	5.3±1.5	14.4±4.0	2.9±0.5	3.7±0.9
Elements[a]					
Na	181.9±21.8	349.8±106.2	1046.7±315.1	189.0±14.4	191.7±33.5
Mg	93.7±76.0	44.6±78.1	156.8±147.5	64.2±111.2	49.0±84.9
Al	73.5±49.5	89.7±65.1	171.7±80.3	84.4±73.5	45.2±48.2
Si	169.9±65.2	163.1±94.3	279.9±60.5	267.4±89.4	312.2±123.1
S	1276.1±676.8	1100.5±262.5	3459.0±796.3	1575.7±915.0	476.5±293.6
Cl	329.9±55.6	616.3±210.4	2882.9±855.0	1078.6±401.9	3497.1±2024.4
K	224.5±99.5	258.7±131.4	467.1±139.1	107.4±42.9	47.9±22.4
Ca	88.9±19.9	114.0±74.6	129.0±50.4	67.5±40.0	31.7±13.6
Ti	8.0±1.6	7.2±5.8	5.3±4.6	4.8±3.0	2.8±2.8
V	2.9±2.9	1.7±0.7	10.3±10.5	3.8±3.1	5.3±7.0
Cr	4.0±0.3	4.1±1.1	7.6±1.0	5.4±0.6	6.9±4.0
Mn	14.8±3.6	22.2±9.6	27.4±14.6	13.7±3.5	21.1±5.2
Fe	147.2±20.5	203.4±127.4	330.0±191.5	94.6±27.1	91.4±22.1
Co	1.6±1.8	1.5±1.8	0.3±0.6	1.4±1.4	0.4±0.7
Ni	1.6±1.1	3.0±2.1	6.6±5.1	3.6±0.7	3.2±2.2
Cu	14.7±4.6	12.3±5.0	28.1±11.4	11.3±3.4	11.4±3.1
Zn	87.1±21.7	99.4±68.9	132.8±60.6	45.0±6.3	74.3±16.7
Sb	16.6±6.1	22.9±7.8	39.4±12.1	33.4±12.5	48.7±30.3
Ba	48.5±8.9	38.8±16.2	144.3±37.3	72.6±18.0	107.9±62.0
Pb	28.5±14.3	20.3±10.5	38.6±4.8	30.3±11.1	36.5±5.2
Ions[a]					
Cl^-	0.2±0.0	0.4±0.1	3.0±0.8	0.5±0.3	2.7±1.4
NO_3^-	4.8±0.9	2.4±0.7	23.9±8.5	0.3±0.0	0.4±0.1
SO_4^{2-}	3.7±1.6	2.2±0.6	10.0±2.6	2.5±1.1	0.5±0.5
Na+	0.3±0.3	0.4±0.1	1.1±0.7	0.6±0.5	0.2±0.2
NH_4^+	1.9±0.5	1.1±0.3	8.0±2.4	0.7±0.4	0.5±0.1
Ca^{2+}	0.3±0.0	0.1±0.0	0.3±0.1	0.1±0.1	0.1±0.1

[a]Name of the individual component can be referred to Table S1 (Supplementary Material).

Table 3: The average concentrations ± standard deviations (pg/m^3) of PAHs, OPAHs and AZAs and carbonyls in five sampling locations

Compound	Sample A	Sample B	Sample C	Sample D*[b]	Sample E*
PAHs (pg/m^3)					
1,2,3,4-Tetrahydronaphthalene	224.4±43.8	135.8±50.6	435.4±158.6	242.9±38.9	330.8±149.7
Naphthalene	1049.9±147.9	1021.7±317.1	2983.5±923.7	1694.9±289.8	2363.3±736.2
2-Methylnaphthalene	212.6±48.5	193.8±52.7	542.7±194.0	342.8±55.1	409.1±135.8
1-Methylnaphthalene	256.5±62.5	218.1±53.7	617.6±216.5	412.3±24.1	921.2±432.8
Biphenyl	242.3±55.1	201.0±58.4	681.1±208.9	333.3±39.6	477.8±146.0
1,3-Dimethylnaphthalene	527.1±129.1	294.2±102.9	912.8±348.4	490.0±30.1	571.4±210.2
Acenaphthylene	178.5±42.1	92.5±53.5	364.2±119.2	205.4±128.8	243.1±139.2
Acenaphthene	883.1±204.0	630.5±237.4	966.0±398.4	1335.8±264.2	2096.3±795.8
Fluorene	217.8±29.3	192.8±55.1	734.5±311.9	346.3±101.4	481.7±290.0
Phenanthrene	1028.6±144.0	603.4±102.9	2150.5±871.9	1187.9±131.1	1615.2±625.7
Anthracene	127.9±23.6	82.0±20.7	273.8±123.6	190.6±99.0	129.8±93.8
4H-Cyclopenta(d,e,f)phenanthrene	692.6±56.7	550.8±189.7	2662.8±1126.2	847.5±258.8	1125.7±437.3
1-Methylphenanthrene	179.1±56.4	99.0±32.1	481.3±206.6	192.1±32.9	315.9±89.2
3,6-Dimethylnaphthalene	94.3±19.3	63.7±21.5	246.5±100.5	136.9±4.4	251.6±105.8
Fluoranthene	249.4±49.2	285.3±155.6	888.7±271.1	273.9±47.4	296.7±126.8
Pyrene	475.1±114.0	1190.9±1105.9	1917.8±696.3	617.4±86.8	814.9±341.6
Retene	638.3±148.9	439.5±132.3	1185.8±445.5	1175.2±118.9	2210.6±1465.4
Benzo[a]anthracene	66.3±17.7	174.8±68.6	275.2±162.1	248.7±6.8	210.8±70.0
Chrysene[a]	324.9±73.2	212.6±81.3	551.1±176.5	210.1±11.9	238.9±80.5
Benzo[b+j+k]fluoranthenes[b]	625.7±199.0	823.7±363.5	1404.4±541.3	472.1±104.8	611.9±208.3
Benzo[e]pyrene	230.1±78.0	387.0±243.0	645.6±346.5	139.9±46.6	177.4±60.1
Benzo[a]pyrene	94.1±33.8	123.8±67.2	134.5±82.4	135.0±32.0	114.1±35.3
Perylene	23.7±11.8	42.1±29.0	1.8±0.7	19.4±12.0	6.7±2.1
Indeno[1,2,3-cd]pyrene	255.2±43.0	664.0±171.7	972.4±920.1	456.6±253.2	89.8±36.4
Dibenzo[a,h]anthracene	88.4±45.1	98.8±67.8	127.2±62.6	97.7±18.3	31.2±10.3
Benzo[g,h,i]perylene	332.0±26.0	832.1±477.7	712.3±355.7	145.5±127.4	655.1±538.9
Coronene	309.0±114.1	675.5±419.6	582.8±373.9	81.7±44.1	81.2±22.3
ΣLMW-PAHs[c]	3485.8±341.3	2622.8±632.7	7472.6±2320.5	4960.8±653.4	6929.3±2174.9
ΣHMW-PAHs[d]	2511.0±435.2	4406.1±1580.9	6983.6±2056.0	2657.1±76.4	3063.4±1079.1
ΣCarci-PAHs[e]	1786.5±297.0	2929.9±930.3	4177.1±1245.3	1765.8±37.1	1951.8±724.5
ΣUS-EPA PAHs	5996.8±752.3	7029.0±2100.8	14456.2±4183.6	7617.9±713.6	9992.7±3253.2
Σ21 Parent-PAHs	6559.6±888.1	8133.6±2608.7	15686.4±4242.0	7858.9±775.0	10258.1±3283.9
Σ30PAHs	9626.7±1078.7	10329.7±3166.4	23452.4±6498.8	12031.9±1219.4	16872.1±5753.7
OPAHs (pg/m^3)					
1-Indanone	65.3±12.9	68.1±32.8	264.5±136.8	226.9±51.3	642.4±428.9
1,4-Naphthoquinone	23.7±5.9	28.2±14.0	305.9±257.3	116.2±27.4	45.6±26.1
1-Naphthaldehyde	22.1±10.6	17.5±8.0	95.8±68.8	51.8±16.6	78.9±40.0
2-Biphenylcarboxaldehyde	8.1±1.6	7.8±3.4	71.7±39.1	23.9±5.0	236.2±126.3
1-Acenaphthenone	23.5±4.1	14.0±5.7	168.1±84.4	38.2±5.9	91.7±55.2
9-Fluorenone	54.6±7.8	35.7±14.9	524.7±243.3	112.6±5.1	423.1±264.4
9,10-Anthraquinone	41.5±7.0	38.4±17.8	413.1±165.9	103.6±29.7	261.6±185.6
1,8-Naphtalic anhydride	41.9±2.3	25.9±14.9	216.8±82.6	23.8±6.2	105.0±66.9
1,4-Anthraquinone	25.5±5.3	42.2±24.6	217.4±111.4	96.3±28.4	359.6±116.3
4H-Cyclopenta[d,e,f]phenanthrenone	7.8±1.5	10.2±5.9	101.3±46.9	12.6±2.1	38.5±29.5
2-Meth-9,10-anthraquinone	47.7±13.5	26.4±16.1	317.1±133.1	165.7±18.6	355.8±243.3
Benzo[a]florenone	22.5±4.2	16.8±7.8	216.1±98.8	38.0±7.1	84.9±36.2
7H-Benzo[d,e]anthracene-7-one	17.8±2.3	32.1±14.2	259.9±121.6	22.8±7.0	52.4±27.3
Benzo[a]anthracene-7,12-dione	15.2±2.6	10.1±4.2	112.4±45.5	12.2±2.1	25.2±8.2
1,4-Chrysenequinone	140.7±106.6	80.1±60.9	291.1±152.6	114.9±28.0	149.3±6.3
5,12-naphthacenequinone	14.7±3.2	8.4±5.3	57.1±24.9	18.6±1.7	105.3±108.8
6H-Benzo[c,d]pyran-6-one	12.5±3.1	40.7±21.7	66.8±42.1	13.2±6.2	58.6±65.1
Σ17OPAHs	585.2±135.6	502.7±249.4	3700.1±1735.3	1191.1±190.8	2944.5±1494.4
AZAs (pg/m^3)					
Quinoline	15.3±3.5	14.1±5.8	97.9±62.9	51.9±14.2	83.0±44.8
Benzo(h)quinoline	5.9±0.7	7.0±2.4	41.9±28.0	28.8±14.5	33.6±21.7
Acridine	3.3±1.5	4.4±3.2	36.0±20.6	18.2±6.0	35.3±24.5
Carbazole	26.8±17.4	7.2±5.8	87.5±52.3	38.9±24.7	49.0±45.5
Σ4AZAs	51.3±19.3	32.7±15.7	263.3±160.0	137.8±39.3	200.8±133.0

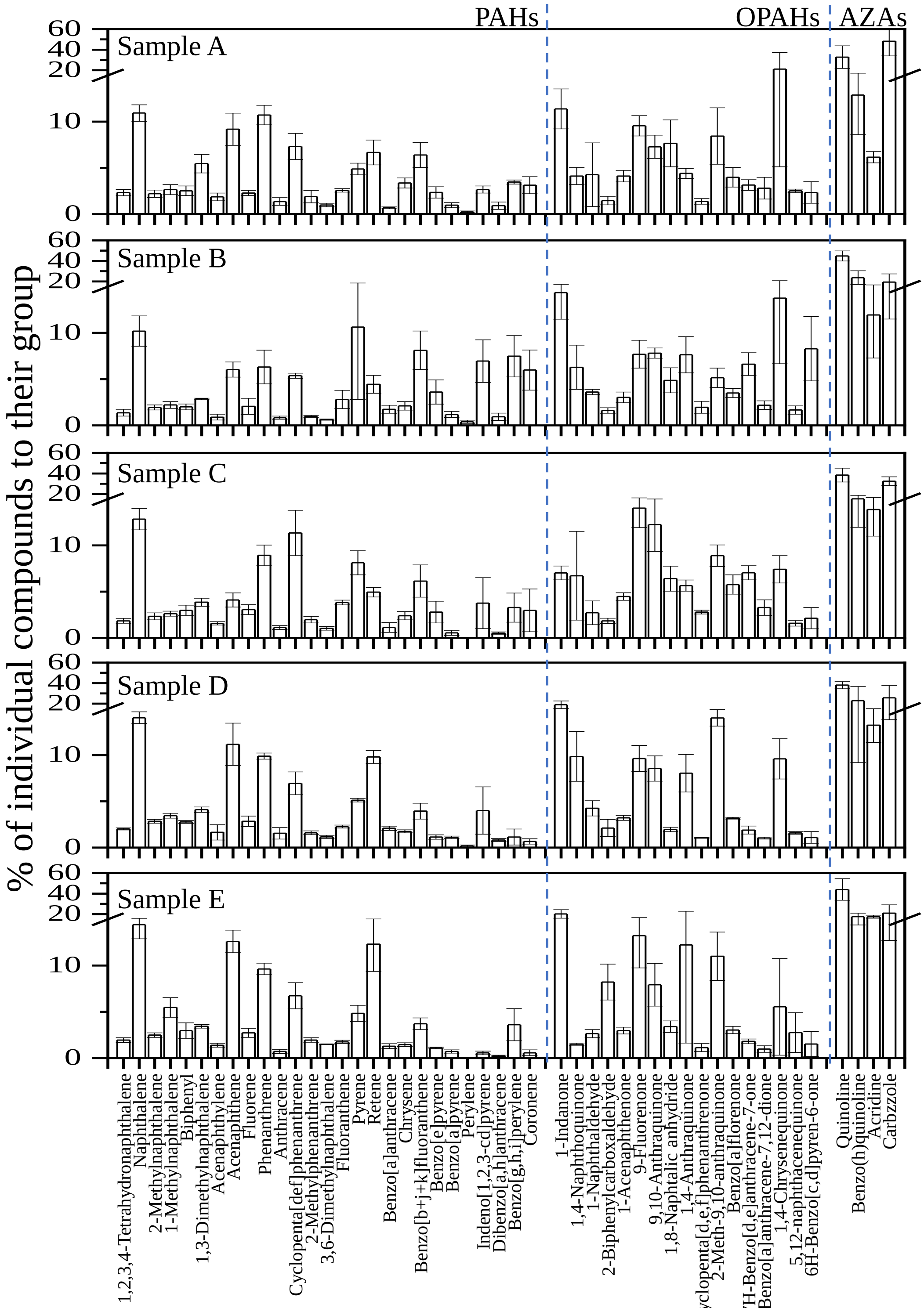
[a] represents chrysene + triphenylene; [b] represents benzo[b]fluoranthene + benzo[j]fluoranthene + benzo[k]fluoranthene;. [c]ΣLMW-PAHs: low molecular weight PAHs with 2-3 aromatic rings; [d]ΣHMW-PAHs: high molecular weight PAHs with 4-7 aromatic rings; [e]ΣCarci-PAHs: benzo[a]anthracene + chrysene +benzo[b+j+k]fluoranthenes + benzo[a]pyrene + indeno[1,2,3-cd]pyrene + dibenzo[a,h]anthracene + benzo[g,h,i]perylene.

Table 4: Spearman's rank correlation coefficients (R) of the biological responses [cell viability, ROS generated (DCFH), inflammatory activity (TNF-α), and oxidative DNA damage] elicited by extracts with the concentrations of chemical species.

Chemical Species	Cell viability	ROS generated (DCFH)	TNF-α	DNA damage
TC	-0.102	-0.239	-0.336	0.307
OC	-0.101	-0.158	-0.423	0.233
EC	-0.028	*-0.502*^{*b}	**0.824****	0.325
Cl^-	0.327	0.367	-0.386	-0.292
NO_3^-	0.216	0.151	0.437	-0.239
SO_4^{2-}	-0.148	-0.176	**0.671****	-0.046
Na^+	-0.346	-0.238	0.389	0.393
NH_4^+	0.165	0.158	0.431	-0.345
Ca^{2+}	0.122	-0.427	**0.676****	0.279
Na	-0.332	-0.287	**0.799****	0.137
Mg	-0.056	-0.156	0.231	-0.164
Al	-0.112	*-0.689***	**0.624***	**0.643****
Si	-0.150	*-0.590**	0.286	0.357
S	-0.360	-0.227	**0.644****	-0.056
Cl	0.214	0.300	*-0.479**	-0.185
K	-0.252	*-0.582**	**0.930****	0.274
Ca	-0.275	*-0.711***	**0.906****	0.434
Ti	0.087	*-0.742***	**0.748****	**0.540***
V	0.191	-0.014	0.035	-0.024
Cr	-0.250	*-0.622***	**0.565***	**0.570***
Mn	-0.140	*-0.715***	**0.641****	**0.543***
Fe	-0.240	*-0.592***	**0.912****	0.298
Co	-0.315	-0.202	0.118	-0.068
Ni	-0.334	-0.359	0.444	0.203
Cu	-0.082	*-0.543**	**0.720****	0.287
Zn	0.076	*-0.646***	**0.693****	0.430
Sb	-0.198	*-0.575**	0.255	0.427
Ba	-0.057	0.008	-0.004	0.317
Pb	-0.017	*-0.491**	0.360	0.316
Hexaldehyde	0.277	0.393	0.106	-0.405
Heptaldehyde	0.307	0.398	0.116	-0.334
Octaldehyde	0.186	0.399	0.197	-0.384
Nonaldehyde	0.342	0.285	0.268	-0.256
Decaldehyde	0.285	**0.595****	-0.329	-0.432
Glyoxal	-0.085	0.414	-0.382	-0.306
Methylglyoxal	-0.061	0.379	-0.387	-0.319
1,2,3,4-Tetrahydronaphthalene	0.177	*-0.470**	0.301	**0.472***
Naphthalene	-0.117	-0.397	0.096	**0.485***
2-Methylnaphthalene	-0.293	-0.421	0.244	0.261
1-Methylnaphthalene	0.327	-0.374	-0.231	0.359
Biphenyl	0.082	-0.164	0.101	0.025
1,3-Dimethylnaphthalene	0.051	-0.449	**0.550***	0.335
Acenaphthylene	0.300	-0.102	0.100	0.189

Acenaphthene	0.193	-0.445	-0.117	**0.533***
Fluorene	-0.267	0.034	0.105	0.046
Phenanthrene	0.364	-0.396	0.244	0.366
Anthracene	-0.343	-0.392	**0.602****	0.361
Cyclopenta[def]phenanthrene	0.000	0.069	0.297	-0.015
1-Methylphenanthrene	0.463	0.082	-0.091	0.112
3,6-Dimethylphenanthrene	0.451	-0.074	-0.360	0.367
Fluoranthene	-0.212	-0.176	**0.683****	0.089
Pyrene	-0.195	-0.041	**0.511***	0.060
Retene	0.297	-0.257	-0.446	0.457
Benzo[a]anthracene	**-0.602****	-0.368	0.234	0.285
Chrysene + Triphenylene	0.054	*-0.593***	**0.822****	0.447
Benzo [b+j+k] fluoranthene	-0.153	*-0.660***	**0.873****	**0.474***
Benzo[e]pyrene	-0.210	*-0.549**	**0.896****	0.346
Benzo[a]pyrene	-0.336	*-0.789***	**0.580***	**0.763****
Perylene	-0.308	*-0.888***	**0.688****	**0.740****
Indeno [1,2,3-cd] pyrene	**-0.615****	*-0.478**	**0.743****	0.248
Dibenzo[ah]anthracene	-0.456	*-0.677***	**0.818****	**0.599****
Benzo[ghi]perylene	0.081	*-0.696***	**0.660****	**0.690****
Coronene	-0.152	*-0.587**	**0.853****	0.465
Quinoline	0.005	0.378	*-0.603***	-0.247
1-Indanone	0.104	0.178	*-0.631***	0.024
1,4-Naphthoquinone	**-0.665****	0.187	0.082	-0.315
1-Naphthaldehyde	0.073	0.243	-0.426	-0.291
2-Biphenylcarboxaldehyde	0.234	**0.594****	*-0.796***	-0.339
1-Acenaphthenone	0.435	**0.711****	*-0.637***	*-0.516**
9-Fluorenone	0.409	**0.739****	*-0.756***	*-0.495**
Benzo[h]quinoline	-0.144	0.266	-0.445	-0.219
Acridine	0.044	**0.581***	*-0.733***	-0.358
Carbazole	0.174	0.424	-0.283	-0.196
9,10-Anthraquinone	0.234	**0.661****	-0.577*	*-0.471**
1,8-Naphthalic anhydride	*0.524**	0.446	-0.072	-0.382
1,4-Anthraquinone	0.173	0.335	*-0.621***	-0.114
4H-Cyclopenta[def]phenanthrenone	0.046	0.444	-0.133	-0.314
2-Methylanthracene-9,10-dione	0.187	**0.686****	-0.884**	-0.418
Benzo[a]fluorenone	0.359	**0.719****	-0.591**	-0.432
7H-Benz[de]anthracene-7-one	0.089	0.281	0.075	-0.184
Benz[a]anthracene-7,12-dione	0.347	0.328	0.164	-0.280
1,4-Chrysenequinone	0.320	0.011	-0.208	0.149
Naphthacene-5,12-dione	*0.665***	0.415	*-0.541**	-0.232
6H-benzo[cd]pyrene-6-one	-0.136	-0.256	0.439	0.171
Σ7Carbonyls	0.233	0.348	0.239	-0.293
Σ30PAHs	-0.048	*-0.876***	**0.745****	**0.763****
Σ17OPAHs	0.371	**0.474***	*-0.617***	-0.223
Σ4AZAs	0.067	0.419	*-0.558**	-0.124

[a]**Correlation is significant at the 0.01 level (2-tailed).
[b]* Correlation is significant at the 0.05 level (2-tailed).



923

924 Figure 1: Mean contributions of individual PAHs, OPAHs, and AZAs to the Σ30PAHs, Σ15OPAHs, and Σ4AZAs concentrations in different sampling locations. The y-axis was broken at 15% to enlarge the scale before the break. Error bars indicate standard deviations for each sample.

928

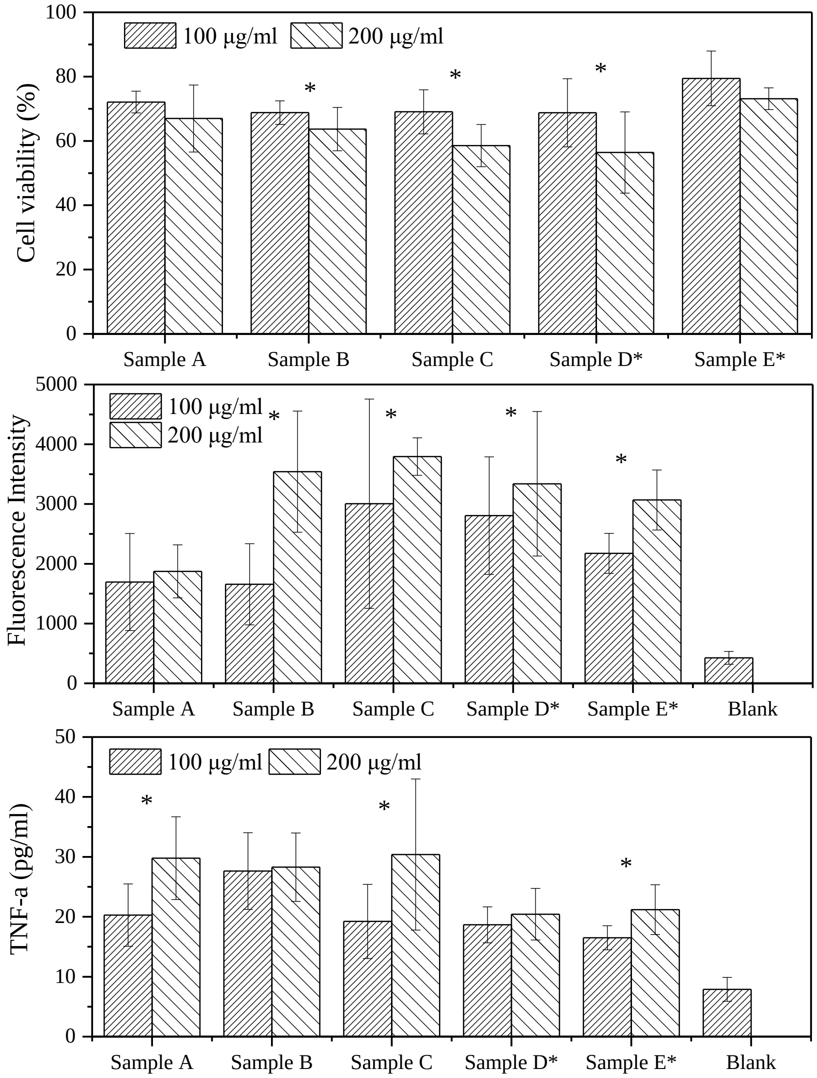


Figure 2: Cell viability, ROS generation (fluorescence intensity) and TNF-α induced by extracts of $PM_{2.5}$ from five sampling locations (*$p < 0.05$). Bars are the mean ±standard deviation