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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)

78 which results in the enhanced formation of various organic compounds such as carbonyls and
79 PAHs (Huang et al., 2011; See et al., 2006). The amount of PAHs released from cooking that
80 involve deep frying (oil-based cooking) was found to be higher than boiling and steaming
81 (water-based cooking) (See and Balasubramanian, 2008). In addition, frying and food
82 ingredients (e.g. fat contents) could be important factors in the PAHs formation during cooking
83 (Saito et al., 2014; Tanaka et al., 2012; Zhu and Wang, 2003). Oxygenated PAHs (OPAHs),
84 nitrogen heterocyclic PAHs (azaarenes: AZAs) are generated together with PAHs from
85 combustion/pyrolysis process. OPAHs can additionally be formed from the photolysis and
86 (photo)chemical oxidation of PAHs (Clergé et al., 2019). The formation of AZAs and OPAHs
87 during cooking have been documented (Blaszczyk and Janoszka, 2008; Szterk, 2015; Li et al.,
88 2016) but these compounds are rarely characterized in cooking fumes (Sun et al., 2020).
89 Several OPAHs (quinones) and AZAs are cytotoxic, involved in the generation reactive oxygen
90 species (ROS), genotoxic, mutagenic, and carcinogens (Bolton et al., 2000; Jung et al., 2001;
91 Sovadinová et al., 2006; Clergé et al., 2019). Some OPAHs and AZAs also been classified by
92 the IARC as probable or possible human carcinogens (IARC, 2010; 2011a,b). Despite some
93 OPAHs and AZAs being equally or even more toxic than the parent-PAHs, most previous
94 studies on emissions from cooking did not characterize the OPAHs and AZAs and hence did
95 not elucidate their possible contributions to the adverse effects of PM (Abdullahi et al., 2013;
96 Ding et al., 2012).

97 Cooking activities also emit carbonyls and the sources were from cooking fuels combustion
98 and heating of cooking oils (Ho et al., 2006; Lin and Liou, 2000; Zhang and Smith, 1999).
99 Some of the aldehydes that are emitted from cooking activities are toxic themselves and also
100 participate in atmospheric chemical reactions to form secondary pollutants that degrade air
101 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

126 production, DNA damage, and inflammation effects) of extracts of these $PM_{2.5}$ samples and 3)
127 determine the relationship between $PM_{2.5}$ chemical components and bioreactivity.

128

129 ## 2. Materials and methods

130 ### 2.1 Sampling locations

131 Cooking fumes were collected from the exhausts of three commercial restaurants operating in
132 Hong Kong and two simulated cooking experiments conducted in environmental chambers.
133 The characteristics of the different sampling sites are detailed in Table 1. The studied
134 restaurants cooked and served common cuisines in Hong Kong. The restaurants were selected
135 based on several criteria. Factors such as roof top access availability, electricity supply during
136 sampling, sampling space (minimum 2 m^2), floor plans, information of exhaust system and
137 sampling safety were carefully considered prior to the sampling campaign. Each of the selected
138 restaurants had to possess an independent exhaust system. This was to ensure that the collected
139 samples were only generated from the target restaurant.
140 The simulated cooking by two common Chinese-style techniques (namely as stir-frying and
141 deep-frying) were conducted in stainless steel environmental chamber of 19.1 m^3 (3.05 m ×
142 3.05 m × 2.05 m) that was designed for measuring emissions from indoor sources.

143

144 ### 2.2 Experimental procedures

145 #### 2.2.1 Sampling from restaurant exhaust system

146 Sampling duration was synchronised to the peak hours of the restaurants. The peak hours in
147 each restaurant were based on information provided by the owners. A particle sampler (DRI
148 MEDVOL) was used for sampling in this study. The device consisted of a $PM_{2.5}$ cyclone, an
149 inlet stilling chamber, a conical plenum, open-faced filter packs and differential pressure flow

150 control together with a pump. The $PM_{2.5}$ cyclone (Bendix 240) was operated at 113 L/min,
151 which removed particles (aerodynamic diameter > 2.5 μm) in the air stream. The air was purged
152 through the cyclone and further diffused inside to the plenum. The plenum was coated with
153 perfluoro alkoxyalkane (PFA) teflon. The PFA teflon open faced filter holder (Savillex 47-mm
154 injection-moulded) was used throughout the sampling section. A Teflon-membrane (47mm),
155 Nuclepore polycarbonate membrane and quartz-fiber filter (47 mm) were positioned in the
156 filter holders separately.
157 Sampling was not performed in the kitchen area but at the rooftop, where sufficient space and
158 access to exhaust for sampling were assured. The $PM_{2.5}$ samples were collected simultaneously
159 during the cooking process. The sampling duration was set as 1.5 hours. Background samples
160 from the kitchens were also collected when the kitchens were not in operation. The background
161 samples were typically collected 1.5-3 hours after the restaurants have closed their operations
162 for the day. Four cooking emission samples, in addition to background and field blank samples
163 were collected at each restaurant for four consecutive sampling days.
164
165 2.2.2 Sampling from simulated cooking in an environmental test chamber
166 Induction hot plates were used for the cooking. Each cooking experiment was completed in 1.5
167 hours. The dishes were representative of typical Hong Kong cuisine including fried “Choy
168 Sum” (a leafy vegetable commonly used in Chinese cuisine) and deep fried chicken breast. The
169 portion of all dishes was enough for 4-5 members of a family. The same chef was responsible
170 for the cooking experiments in order to ensure sample harmony. $PM_{2.5}$ emitted from these
171 simulated cooking experiments (sample D*-E* from test chamber and sample A-C from
172 commercial restaurant) were sampled with an identical sampling system as described in section
173 2.2.1. After each sampling, the filters were sealed in Petri dishes and frozen (-20 °C) until
174 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

323 is that it cooks and serves Cantonese and Hong Kong local cuisine that are prepared with
324 methods such as deep frying, stir frying, pan frying and steaming (Table 1). Since the cooking
325 methods and size of restaurant C is comparable to restaurant B, the main reason for the high
326 concentration of the chemical species in C can be explained by higher emissions during
327 cooking of Cantonese dishes than during the cooking of mixed cuisines and Western dishes
328 prepared in restaurants B and A, respectively (Table 1). Such influence of various cultural
329 cooking styles on the amount of emitted PAHs, as well as the fact that Asian cooking style
330 emits higher PAHs than Western cooking has been reported in previous studies (See and
331 Balasubramanian, 2008; Abdullahi et al., 2013). Our study reveals that different cultural
332 cooking styles will also result in different concentrations of the PAH derivatives (AZAs and
333 OPAHs) in cooking fumes. Specific studies have shown that cooking can result in the formation
334 of AZAs and OPAHs (Blaszczyk and Janoszka, 2008; Li et al., 2016; Szterk, 2015). A
335 combination of factors such as food components, relatively high usage of oil, type of oils, fat
336 content of meat, spices and cooking methods applied (frying at high temperatures) during the
337 cooking of Cantonese and local dishes results in the highest emitted concentrations of PAHs,
338 OPAHs and AZAs (Li et al., 2018; See and Balasubramanian, 2008). Oil-based cooking
339 methods could generally release more PAHs and their derivatives due to direct evaporation,
340 oxidation, pyrolysis, and/or degradation of organic compounds from oils at higher temperature
341 (Abdullahi et al., 2013; Moret and Conte, 2000). The higher concentration of Σ30PAHs,
342 Σ17OPAHs and Σ4AZAs in sample E* compared to sample D* demonstrates that meat cooking
343 by pan frying not only generates higher concentrations of PAHs (which has also been
344 previously reported) but also OPAHs and AZAs than cooking of plant based food by stir frying
345 (Abdullahi et al., 2013; Schauer et al., 1999a). The pyrolysis of animal fats and oils during the
346 cooking of meat could be an explanation for the formation of PAHs and OPAHs. Azaarenes
347 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 ≥ 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.

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

452 Median lethal dose (LD_{50}) of the samples (Figure S5) were determined by dosage response
453 analysis in Figure S6 (Supplementary Material). Sample A showed highest LD_{50} and the lowest
454 LD_{50} was in sample E*. The results indicate that lower $PM_{2.5}$ concentration was required to
455 cause 50% DNA damage for sample E* compared to the other samples. Both sample B and C
456 show comparable DNA damage in this study. This observation could be due to similar cooking
457 characteristics.
458 Chemical species and elements some of which have been quantified (e.g. heterocyclic amines,
459 metals, and PACs) in emissions from cooking could cause oxidative stress, cell injury, DNA
460 damage and mutations, which could result in the above observed effects and risk of diseases
461 such as lung cancer, cardiovascular and pulmonary diseases (Pardo et al., 2015; Gerlofs-
462 Nijland et al., 2009; Wei et al., 2009; Xue and Warshawsky, 2005; Bolton et al., 2000; Seow
463 et al., 2000).
464

465 3.7 Correlation between chemical components and bioreactivities
466 Cell viability showed significant negative correlations with the concentrations of
467 benzo[a]anthracene, indeno [1,2,3-cd]pyrene and 1,4-naphthoquinone in the samples (Table
468 4). These relationships suggest that these compounds might contribute to the toxicity of extracts
469 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

494 positive correlation with ROS generation in cellular assays (Shafer et al., 2016; Daher et al.,
495 2014; Verma et al., 2009; Hu et al., 2008).

496 Strong positive correlations were also found between TNF-α and the concentration of several
497 chemical species (EC, Ca^{2+}, SO_4^{2-}, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Ni, 14 individual PAHs and
498 Σ30PAHs; Table 4 and Figure S7). The TNF-α was however significantly negatively correlated
499 with the concentration of more polar PAH derivatives (Σ4AZAs and Σ30OPAHs). This is
500 despite the fact that the concentrations of PAH derivatives were positively correlated with the
501 concentrations of PAHs. This suggests that the impact of PAHs extracted from the $PM_{2.5}$ on
502 the generation of pro-inflammatory cytokines was opposite to the impact of PAH derivatives
503 (OPAHs and AZAs). Most of the PAHs showing strongest positive correlations with TNF-α
504 are those with 4-6 ring sizes (HMW-PAHs) with several of them classified as probable human
505 carcinogens. Our study suggests that inflammation is a strong pathway for the toxicity of $PM_{2.5}$
506 emitted from cooking emissions and that several chemical species including PAHs, transition
507 metals might be the triggers for the inflammatory response. Relationships between
508 inflammation and metal/PAH contents of PM has been reported in other studies (Pardo et al.,
509 2015; Gerlofs-Nijland et al., 2009).

510 Significant positive correlation was observed between DNA damage and concentrations of
511 several elements (Al, Ti, Cr and Mn), Σ30PAHs and individual PAHs (1,2,3,4-
512 tetrahydronaphthalene, naphthalene, acenaphthene, dibenzo[a,h]anthracene, perylene,
513 benzo[a]pyrene, benzo[b+j+k]fluoranthene and benzo[ghi]perylene) (Table 4). PAHs are
514 known mutagens and carcinogens and damage to DNA is a known mechanism for their
515 carcinogenic effect (Wei et al., 2009; IARC 2005; Bolton et al., 2000). A previous study
516 showed cytotoxicity of Cr in Chinese commercial cooking $PM_{2.5}$ emissions (Sun et al., 2020).
517

518 **4. Conclusions**

519 This study investigated $PM_{2.5}$ emissions generated from different cooking conditions. The
520 concentrations of nearly all chemical species were highest in one restaurant that served
521 Cantonese dishes and applied oil frying methods, suggesting the cooking ingredients and
522 conditions could be the determining factors for higher emissions. The extracts from the samples
523 elicited toxicologically relevant responses (cell death, ROS generation, inflammation activity
524 and DNA damage). The responses were in dose dependent manner and demonstrated higher
525 responses under higher $PM_{2.5}$ concentrations. These responses were correlated to
526 concentrations of specific chemical species in the $PM_{2.5}$ suggesting that some of the determined
527 chemical species might play roles in the toxicity of $PM_{2.5}$ emitted from cooking activities.
528
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532
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890 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

891 [a] represents samples collected from stainless steel environmental constant temperature and humidity test chamber that mimic residential kitchen hood condition.
892 [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.
893 [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.
894 [d] represents the sampling condition of exhaust system in restaurant and the test chamber.
895

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).

909 Table 3: The average concentrations ± standard deviations (pg/m^3) of PAHs, OPAHs and AZAs
910 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

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

916 Table 4: Spearman's rank correlation coefficients (R) of the biological responses [cell viability,
917 ROS generated (DCFH), inflammatory activity (TNF-α), and oxidative DNA damage] elicited
918 by extracts with the concentrations of chemical species.
919

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).

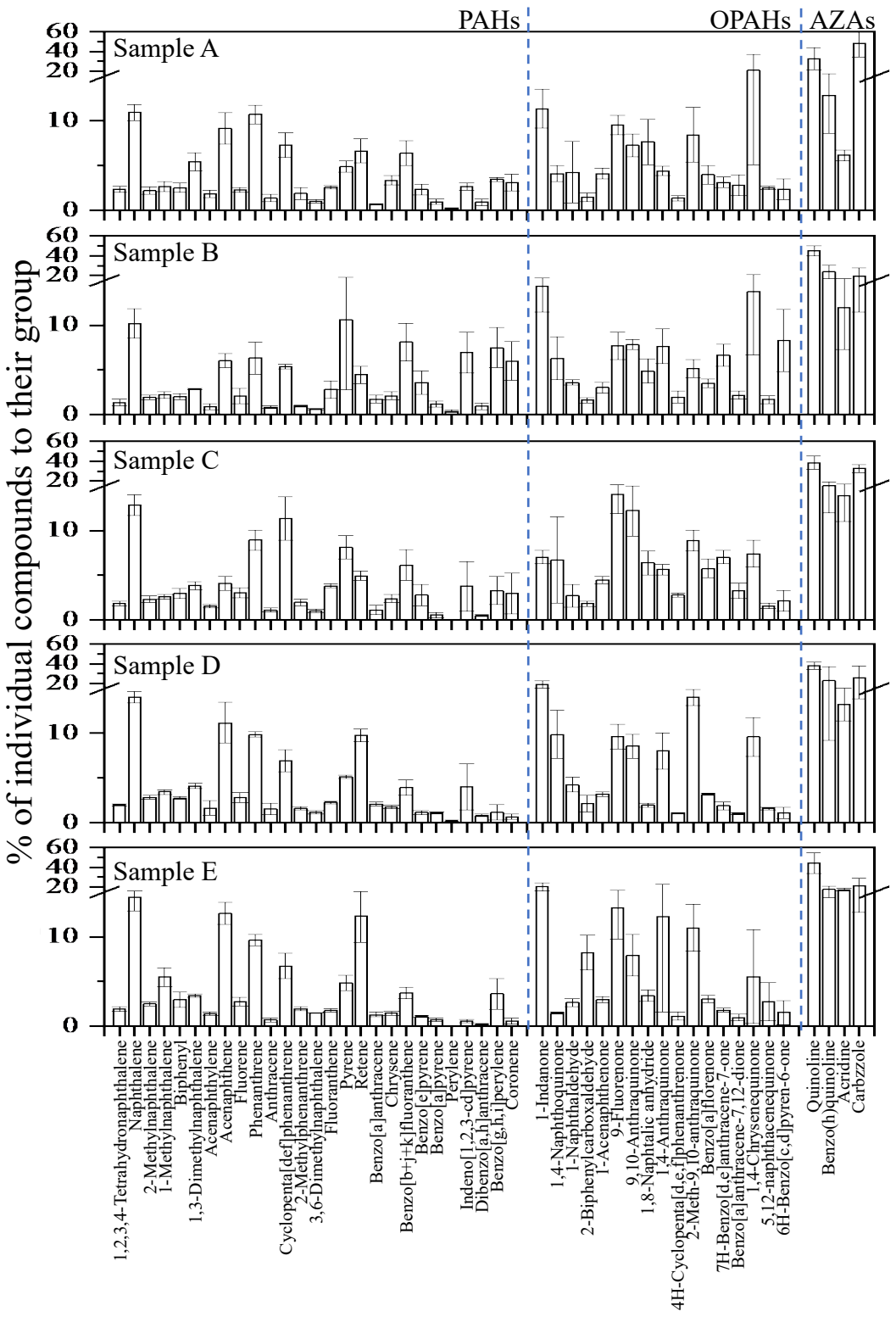
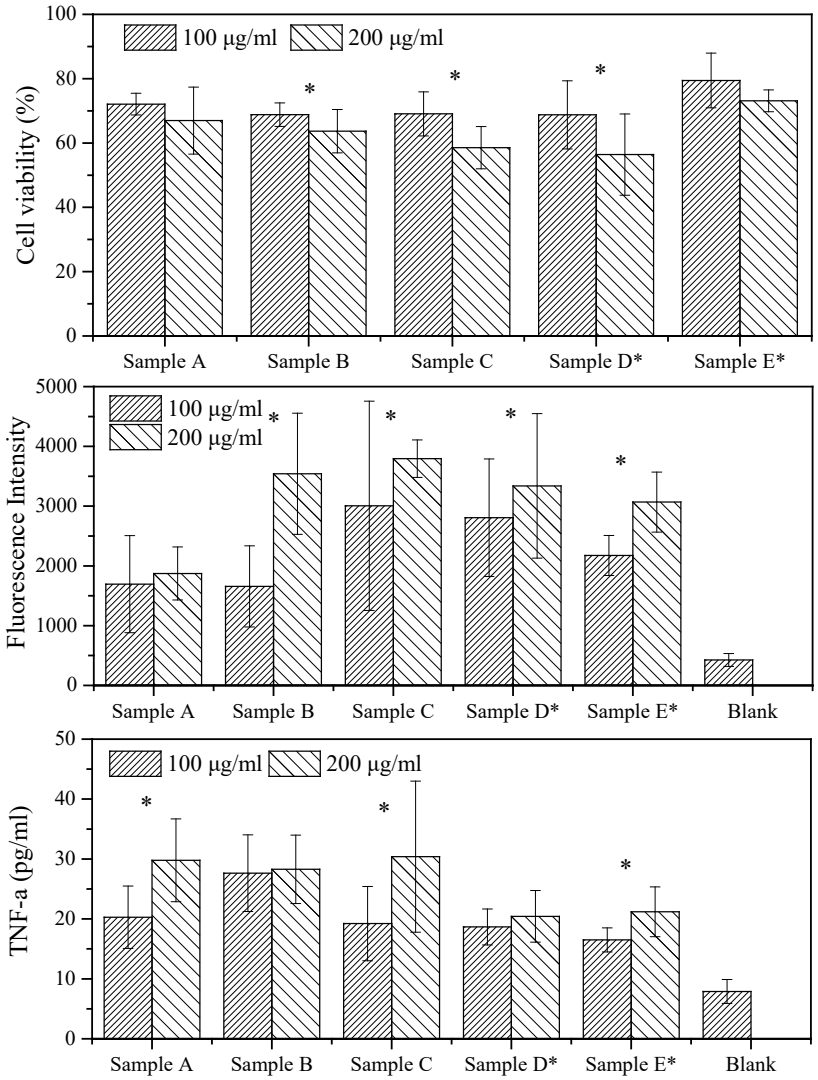


Figure 1: Mean contributions of individual PAHs, OPAHs, and AZAs to the Σ30PAHs, Σ15OPAHs, and Σ4AZAs concentrations in different sampling locations. The y-axis was broken at 15% to enlarge the scale before the break. Error bars indicate standard deviations for each sample.



929

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

933

934