

Online Research @ Cardiff

This is an Open Access document downloaded from ORCA, Cardiff University's institutional repository: <https://orca.cardiff.ac.uk/id/eprint/145106/>

This is the author's version of a work that was submitted to / accepted for publication.

Citation for final published version:

Bandowe, Benjamin A. Musa, Lui, K.H., Jones, Timothy ORCID: <https://orcid.org/0000-0002-4466-1260>, BeruBe, Kelly ORCID: <https://orcid.org/0000-0002-7471-7229>, Adams, Rachel, Niu, Xinyi, Wei, Chong, Cao, Jun-Ji, Lee, S.C., Chuang, Hsiao-Chi and Ho, K.F. 2021. The chemical composition and toxicological effects of fine particulate matter (PM2.5) emitted from different cooking styles. *Environmental Pollution* 288, 117754. 10.1016/j.envpol.2021.117754 filefile

Publishers page: <http://dx.doi.org/10.1016/j.envpol.2021.117754>
<<http://dx.doi.org/10.1016/j.envpol.2021.117754>>

Please note:

Changes made as a result of publishing processes such as copy-editing, formatting and page numbers may not be reflected in this version. For the definitive version of this publication, please refer to the published source. You are advised to consult the publisher's version if you wish to cite this paper.

This version is being made available in accordance with publisher policies.

See

<http://orca.cf.ac.uk/policies.html> for usage policies. Copyright and moral rights for publications made available in ORCA are retained by the copyright holders.



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

3 Benjamin A. Musa Bandowe^{1,2#}, K. H. Lui^{3#}, Timothy Jones⁴, Kelly BéruBé⁵, Rachel
4 Adams⁶, Xinyi Niu³, Chong Wei⁷, Jun-Ji Cao^{8,9}, S. C. Lee¹⁰, Hsiao-Chi Chuang^{11,12}, K. F.
5 Ho^{3*}

6
7 ¹*Institute of Geography, University of Bern, Hallerstrasse 12, 3012 Bern, Switzerland*

8 ²*Max Planck Institute for Chemistry, Multiphase Chemistry Department, Hahn-Meitner-Weg 1, 55128*
9 *Mainz, Germany*

10 ³*The Jockey Club School of Public Health and Primary Care, The Chinese University of Hong Kong,*
11 *Hong Kong, China*

12 ⁴*School of Earth and Environmental Sciences, Cardiff University, Park Place, Cardiff, U.K.*

13 ⁵*School of Biosciences, Cardiff University, Museum Avenue, Cardiff, U.K.*

14 ⁶*Cardiff School of Health Sciences, Cardiff Metropolitan University, Western Avenue, Cardiff, U.K.*

15 ⁷*Shanghai Carbon Data Research Center (SCDRC) CAS Key Laboratory of Low-Carbon Conversion*
16 *Science and Engineering, Shanghai Advance Research Institute, Chinese Academy of Sciences,*
17 *Shanghai 2010210, China*

18 ⁸*Key Laboratory of Aerosol Chemistry and Physics, SKLLQG, Institute of Earth Environment, Chinese*
19 *Academy of Sciences, Xi'an 710075, China*

20 ⁹*Institute of Global Environmental Change, Xi'an Jiaotong University, Xi'an, China*

21 ¹⁰*Department of Civil and Structural Engineering, Research Center of Urban Environmental*
22 *Technology and Management, The Hong Kong Polytechnic University, Hong Kong, China*

23 ¹¹*School of Respiratory Therapy, College of Medicine, Taipei Medical University, Taipei, Taiwan*

24 ¹²*Division of Pulmonary Medicine, Department of Internal Medicine, Shuang Ho Hospital, Taipei*
25 *Medical University, Taipei, Taiwan*

26 [#]These authors contributed equally to this work.

27 ^{*}Corresponding author. Tel.: +852 22528763; fax: +852 26063500

28 E-mail address: kfho@cuhk.edu.hk

29

30 **Abstract**

31 The mass, chemical composition and toxicological properties of fine particulates (PM_{2.5})
32 emitted from cooking activities in three Hong Kong based restaurants and two simulated
33 cooking experiments were characterized. Extracts from the cooking PM_{2.5} elicited significant
34 biological activities [cell viability, generation of reactive oxygen species (ROS), DNA damage
35 and inflammation effect (TNF- α)] in a dose-dependent manner. The composition of PAHs,
36 oxygenated PAHs (OPAHs) and azaarenes (AZAs) mixtures differed between samples. The
37 concentration ranges of the Σ 30PAHs, Σ 17OPAHs and Σ 4AZAs and Σ 7 carbonyls in the
38 samples were 9627 – 23452 pg m⁻³, 503 – 3700 pg m⁻³, 33 – 263 pg m⁻³ and 158 – 5328 ng m⁻³,
39 respectively. Cell viability caused by extracts from the samples was positively correlated to
40 the concentration of benzo[a]anthracene, indeno[1,2,3-cd]pyrene and 1,4-naphthoquinone in
41 the PM_{2.5} extracts. Cellular ROS production (upon exposure to extracts) was positively
42 correlated with the concentrations of PM_{2.5}, decaldehyde, acridine, Σ 17OPAHs and 7
43 individual OPAHs. TNF- α showed significant positive correlations with the concentrations of
44 most chemical species (elemental carbon, 16 individual PAHs including benzo[a]pyrene,
45 Σ 30PAHs, SO₄²⁻, Ca²⁺, Ca, Na, K, Ti, Cr, Mn, Fe, Cu and Zn). The concentrations of Al, Ti,
46 Mn, Σ 30 PAHs and 8 individual PAHs including benzo[a]pyrene in the samples were positively
47 correlated with DNA damage caused by extracts from the samples. This study demonstrates
48 that inhalation of PM_{2.5} emitted from cooking could result in adverse human health effects.

49

50 **Keywords:**

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

52

53

54 1. Introduction

55 Emissions from cooking constitute an important source of particulate matter (PM) in the indoor
56 and outdoor environment (Abdullahi et al., 2013; Cheng et al., 2016). Emissions from cooking
57 has been identified as an important source of fine particulate matter (aerodynamic diameter <
58 2.5 μm : $\text{PM}_{2.5}$) in populated urban areas such as Hong Kong (Allan et al., 2010; Huang et al.,
59 2011; Mohr et al., 2012). A previous study showed that commercial cooking restaurants in the
60 South Coast Air Basin, USA emitted ~10.4 tons/day of $\text{PM}_{2.5}$ (Gysel et al., 2018). Such
61 emissions represent a major source of exposure to $\text{PM}_{2.5}$, which can adversely affect human
62 health (Chiang et al., 1997; See and Balasubramanian, 2006; Zhong et al., 1999a).

63 Previous review articles have documented the emission of PM of various size ranges and >
64 300 chemicals species [e.g. organic carbon (OC), elemental/black carbon (EC/BC),
65 metals/metalloids, water soluble ions (Cl^- , SO_4^{2-} , NO_3^- , K^+ , Ca^{2+} , Mg^{2+}), volatile organic
66 compounds (VOCs), carbonyls, polycyclic aromatic hydrocarbons (PAHs)] from cooking
67 activities (Abdullahi et al., 2013; Wang et al., 2017; Zhao and Zhao, 2018). The emitted PM,
68 chemical species in gaseous phase and bound to PM are formed from a range of reactions
69 (hydrolysis, thermal oxidation, Maillard reaction, recombination) between chemical
70 components of oils, fats, solid food components, water etc. under high temperature (Abdullahi
71 et al., 2013).

72 The amount, size distribution and composition profiles of PM and their bound chemicals
73 emitted from cooking are driven by factors such as cooking styles, cooking methods, types of
74 oil, ingredients, additives, indoor and ambient conditions (Buonanno et al., 2009; He et al.,
75 2004; McDonald et al., 2003; See et al., 2006; Torkmahalleh et al., 2013; Ho et al., 2006;
76 Huang et al., 2011; Nolte et al., 1999; Saito et al., 2014; Zhao et al., 2007). Cooking
77 (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).

102 Epidemiological studies have demonstrated associations between exposure to cooking fumes
103 and lung cancer risk (Seow et al., 2000; Zhong et al., 1999a). Several studies found that the
104 risk of lung cancer in non-smoking women increases with increasing exposure to cooking
105 fumes (Zhong et al., 1999a; Seow et al., 2000; Metayer et al., 2002). Some of the studies linked
106 in lung cancer risk to exposure to fumes from cooking (frying) with oil at high temperature
107 such as in Chinese-style cooking (Metayer et al., 2002; Shields et al., 1995; Zhong et al.,
108 1999b). PM emitted from cooking was shown to have mutagenic activity (Chiang et al., 1997;
109 Qu et al., 1992; Wu et al., 1998). The International Agency for Research on Cancer (IARC)
110 has classified fumes originating from high temperature frying as a probable human carcinogen
111 (Straif et al., 2006).

112 Upon inhalation, PM_{2.5} and the chemicals bound to it such as quinones and transition metals it
113 can trigger the overproduction of reactive oxygen species (ROS), which counteracts anti-
114 oxidative defences (Gao et al., 2020; Charrier et al., 2014). The physiological effects of ROS
115 imbalance can cause oxidative stress, inflammatory response, DNA and cell damage, which is
116 the basis for several diseases (Kelly, 2003; Bitterle et al., 2006). Transition metals, secondary
117 organic aerosols and OPAHs (quinones) were shown to induce ROS formation (Charrier et al.,
118 2014; Bates et al., 2019). Polycyclic aromatic compounds (PAHs, OPAHs, AZAs) can cause
119 oxidative DNA damage and DNA adduct formation (Clergé et al., 2019; Xue and
120 Warshawsky., 2005; Yamada et al., 2004; Bolton et al., 2000) that can result in
121 carcinogenic/mutagenic effects and cancers.

122 Knowledge of the amounts, chemical composition and toxicity of PM_{2.5} emitted from cooking
123 activities remain limited. The aims of this study are to: 1) characterize the chemical
124 composition (including the particularly understudied OPAHs and AZAs) of PM_{2.5} samples
125 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 m²), 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 m³ (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.

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

180

181 2.3 Analytical methods

182 2.3.1 Chemical components analysis

183 Detailed description of the analytical methods applied for the determination of the chemical
184 components [OC, EC, inorganic elements, water-soluble ions (Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, Ca²⁺ and
185 NH₄⁺), PAHs, OPAHs, AZAs and carbonyls] in the sampled PM_{2.5} can be found in
186 Supplementary Material (Text S1-S4). In summary, the OC and EC were analysed by the
187 thermal/optical carbon analyser. The inorganic elements and water soluble ions were analysed
188 by the Energy Dispersive X-ray Fluorescence Analyser (ED-XRF) and ion chromatography
189 system, respectively. The polycyclic aromatic compounds (PACs) in the filters were extracted
190 with organic solvents and determined by gas chromatography–mass spectrometry (GC-MS)
191 (Bandowe et al., 2016; Bandowe et al., 2014). The carbonyls were extracted from filters with
192 methanol, derivatized using pentafluorobenzylhydroxyl amine (PFBHA) and analyzed by GC-
193 MS. A list of the chemical species determined in the samples are shown in Table S1.

194

195 2.3.2 Cell culture and bioreactivity analysis

196 The human alveolar epithelial cells (A549) were obtained from American Type Culture
197 Collection (ATCC, Rockville, MD, USA) and were cultured in RPMI medium containing 10%
198 fetal bovine serum, penicillin, and streptomycin at 37 °C with 95% humidity and 5% CO₂.
199 PM_{2.5} samples were from Teflon filters removed according to previous reports (Wang et al.,

200 2021), followed by resuspended in 0.01% dimethyl sulfoxide (DMSO) in serum-free minimum
201 essential RPMI medium. Cells were exposed to the PM_{2.5} samples at 0 (from blank filter), 100
202 and 200 µg/mL for 24 hrs. Cells were examined for cell viability and ROS activity, whereas
203 the supernatant was collected for cytokine analyses.

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

210

211

212 2.3.3 Oxidative DNA damage

213 The plasmid scission assay (PSA) was used to determine the capability of each sample to
214 induce oxidative DNA damage. The level of particle–DNA interaction and subsequent damage
215 were measured by the three conformations of plasmid DNA topological states namely:
216 supercoiled (no damage), relaxed (minor damage), and linear (severe damage) as shown in
217 Figure S1-2 (Supplementary Material). Due to the amount of sample required for the analysis,
218 PM_{2.5} samples were pooled together for PSA analysis. Additional information about the
219 procedure can be referred to in previous studies (Chuang et al., 2013; Shao et al., 2006). The
220 PM_{2.5} samples were run in suspension using molecular grade water over a range of
221 concentrations. Twenty nanograms (20 ng) of Φ X174 RF DNA was added to the liquid and
222 incubated for the analysis. The samples were conducted in triplicate. The final gel results were
223 captured in images and determined by densitometric analysis (Genetools; Syngene system,

224 UK). Molecular grade water and restriction enzyme PstI were used as control and positive
225 control in this study, which caused 4.1% and 95.9% DNA damage, respectively.

226

227 2.4 Calculations and statistical analysis

228 The sum of the concentration of all analysed PAHs, parent-PAHs, 16 US EPA PAH, OPAHs,
229 AZAs, carbonyls are referred to as $\Sigma 30$ PAHs, $\Sigma 21$ parent-PAHs, Σ US-EPA PAHs, $\Sigma 17$ OPAHs,
230 $\Sigma 4$ AZAs and Σ Carbonyls, respectively. The sum of the concentration of parent-PAHs with 2-
231 3 and 4-7 benzene rings are referred to as Σ LMW-PAHs and Σ HMW-PAHs, respectively.
232 Σ Carci-PAHs refers to the sum of the concentration of eight carcinogenic-PAHs (benzo
233 [a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene,
234 indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, benzo[ghi]perylene). Statistical analysis was
235 performed using SPSS 21.0 software. The significance level was set at $p < 0.05$. Due to the small
236 sample size and non-parametric nature of the dataset, comparisons of the means of all samples types
237 were done with Games-Howell test. Spearman's rank correlation was applied to identify relationships
238 between chemical species and also between chemical species and biological end points [cell viability,
239 ROS formation, TNF- α , oxidative DNA damage]. The DNA damage value at 1000 $\mu\text{g ml}^{-1}$ dosage and
240 ROS production and TNF- α values at extract concentration of 200 $\mu\text{g/ml}$ were chosen for the correlation
241 analysis.

242

243 3. Results and discussion

244 3.1 Concentration of PM_{2.5}, OC and EC

245 Mass concentrations of PM_{2.5} in all samples are shown in Table 2. The highest and lowest mass
246 concentrations of PM_{2.5} were in samples C ($711.5 \pm 257.2 \mu\text{g m}^{-3}$) and B ($177.4 \pm 58.4 \mu\text{g m}^{-3}$).
247 The concentration of PM_{2.5} in sample C was ~ 1.45 – 4.01 times higher than in other samples.
248 The high concentration of PM_{2.5} in sample C could be attributed to cooking methods (stir-frying

249 and deep-frying) in this restaurant which is supported by similar findings from other studies
250 (Abdullahi et al., 2013; See and Balasubramanian, 2006). The concentrations of PM_{2.5} in all
251 samples are comparable to a recent study focused on outdoor char broiling and conventional
252 Chinese cooking (Li et al., 2018). The concentrations of PM_{2.5} in sample D* and E* (chamber)
253 are also comparable to other Chinese cooking styles (Shandong and Hunan) as reported in a
254 previous study (Wang et al., 2015).

255 The OC was the most abundant chemical component in the PM_{2.5} (samples A-E*). The average
256 OC concentration is in a range of 99.9–338.8 µg/m³. The OC concentration in sample C
257 (highest concentration) is ~3.39 times higher than in sample B (lowest concentration). The
258 composition of the PM_{2.5} mass was predominantly comprised of carbonaceous particles,
259 particularly in OC at a proportion, which are higher than or comparable to previous studies (Li
260 et al., 2018; Wang et al., 2015). The results are consistent with other findings that PM emissions
261 from cooking operations were primarily organic in nature (Gysel et al., 2017; Li et al., 2015;
262 Wang et al., 2015; Zhang et al., 2017). The lower OC composition (< 90%) compared to another
263 study on emissions from charbroiling/grilling of chicken and beef could be due to relatively
264 higher use of vegetables in Chinese cuisine (McDonald et al., 2003). A previous study also
265 reported that OC constituted the highest fraction of the mass PM emissions from different
266 cooking processes (See and Balasubramanian, 2008). The highest carbon fractions (> 60%) in
267 sample D* and E* could possibly be due to high-fat content of the cooking materials and high
268 amount of oils used in the cooking processes (Zhang et al., 2017). The EC fractions contribute
269 < 5% of the total PM_{2.5} mass in each of the five samples and is consistent with a previous study
270 (Wang et al., 2015). The variations in the average concentrations of PM_{2.5}, OC and EC in the
271 five samples could be due to the differences in cooking ingredients, cooking conditions and
272 methods (Gysel et al., 2018) some of which are outlined in Table 1.

273

274 3.2 Concentration of inorganic elements and water extractable ions

275 The concentrations of elements and ions can be found in Table 2. The two most abundant
276 elements in each sample were S and Cl. Other major elements found in the samples were Na,
277 K, Fe, Mg, Al, Ca, Zn and Ba. Elements with medium range concentrations in the samples
278 were Mn, Sb, Pb and Cu while four other elements (i.e. Ti, V, Cr, Co and Ni) were in low
279 abundance ($< 10 \text{ ng m}^{-3}$) in all samples. The highest concentration of 16 of the 20 elements
280 studied was in sample C. Most of the elements are components of food ingredients (vegetables,
281 meat, cooking oils, salt, spices, water (Butnariu and Butu; Cobos and Diaz, 2014; Epstein, 1999)
282 and can therefore be emitted as part of the PM in cooking fumes. Previous studies have also
283 detected these elements in fumes from restaurants with different cooking styles and cooking of
284 meat in simulated facilities (Gysel et al., 2018; Abdullahi et al., 2013; See and Balasubramian,
285 2008; McDonald et al., 2003). The presence of Fe, Ni, Cr and Cu in the samples could also be
286 due to the release from cooking utensils (See et al., 2006; Taner et al., 2013; Gysel et al., 2018).
287 A previous study showed that the Cr composition of stainless steel materials could vary from
288 11 to 30% (Kuligowski and Halperin, 1992). Some of the elements could have contaminated
289 the food materials during their growing or processing because these elements are present in
290 environmental compartments (air, soil) from anthropogenic (traffic) and natural sources crustal
291 materials, dust, rocks, soils (Kebata-Pendias and Mukherjee, 2007; Louie et al., 2005). Some
292 of the elements (e.g. Na, Mg, Ca, Fe, Zn, Cu, Co, Mn) that have been detected in the PM
293 samples are essential for humans and are therefore harmless at the required concentrations, but
294 others (e.g. Pb) have no known biological functions in humans and are only harmful to human
295 health (Kebata-Pendias and Mukherjee, 2007). Upon inhalation, some of the transition metals
296 (e.g. Fe, Cu, Mn) can catalyse ROS production leading to oxidative stress, inflammatory effects
297 and oxidative DNA damage, which results in diseases such as cancers, respiratory and

298 cardiovascular diseases (Gao et al., 2020; Pardo et al., 2015; Danielson et al., 2011; Gerlofs-
299 Nijland et al., 2009).

300 Water extractable ions were also found in all sample extracts (Table 2), which is consistent
301 with previous studies that also detected these substances in cooking fumes both from cooking
302 test chambers and restaurants (Abdullahi et al., 2013; Schauer et al., 2002; See and
303 Balasubramian, 2008). The highest concentrations of each of the ions can be found in Sample
304 C. The concentration of the water soluble ions in sample C showed the following trend: NO_3^-
305 $> \text{SO}_4^{2-} > \text{NH}_4^+ > \text{Cl}^- > \text{Na}^+ > \text{Ca}^{2+}$. The trends in other samples are slightly different. NO_3^- ,
306 SO_4^{2-} and NH_4^+ could be components of water and other cooking ingredients but could also be
307 secondary products formed from SO_2 , NO_x and NH_3 emitted from heating of cooking
308 ingredients (See and Balasubramian, 2008; Schauer et al., 2002).

309

310 3.3 PAHs, OPAHs and AZAs in samples

311 The concentrations of PACs measured in the samples are shown in Table 3 and Figure S3.
312 Samples C shows the highest concentrations of $\Sigma 30\text{PAHs}$, $\Sigma 17\text{OPAHs}$ and $\Sigma 4\text{AZAs}$. Sample
313 C also showed the highest concentrations of 27 of 30 PAHs measured in the samples and the
314 sums of PAHs sub-groups ($\Sigma 16$ US-EPA PAHs, ΣCarci PAHs, $\Sigma \text{LMW-PAHs}$ and $\Sigma \text{HMW-}$
315 PAHs). Out of the 17 individual OPAHs measured, 13 had the highest average concentrations
316 in Sample C, while the highest concentration of each of the individual AZAs was also in
317 Sample C (Table 3). Benzo[a]pyrene is often used as a main indicator or marker of carcinogenic
318 PAHs (Boström et al., 2002). The concentration (mean \pm standard deviation in pg/m^3) of
319 benzo[a]pyrene in the samples increased in the order: D (135 ± 32) $>$ C (134.5 ± 82.4) $>$ B
320 (124 ± 67) $>$ E (114 ± 35) $>$ A (94 ± 34). The comparatively large amount of food processed and
321 cooked in the kitchen of this commercial restaurant (Sample C) could be a reason for $\text{PM}_{2.5}$
322 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; Sztark, 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 $\Sigma 30$ PAHs,
342 $\Sigma 17$ OPAHs and $\Sigma 4$ AZAs 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

348 compound components (e.g. aromatic amino acids) of food ingredients (Blaszczyk and
349 Janoszka, 2008). The formed PACs could then subsequently be carried with the cooking fumes
350 (Rogge et al., 1991). Food ingredients may also be contaminated with PAHs, which can
351 subsequently be released with the cooking fumes (Martorell et al., 2010). The composition
352 pattern of the PAHs were slightly different for the different samples (Figure 1). The most
353 abundant PAHs in samples A were naphthalene (10.9%), phenanthrene (10.7%), acenaphthene
354 (9.2%) with Σ LMW-PAHs/ Σ HMW-PAHs of 1.4. Sample B was dominated by pyrene (10.6%),
355 naphthalene (10.2%) and benzo[b+k+k]fluoranthene (8.1%) with Σ LMW-PAHs/ Σ HMW-
356 PAHs of 0.65. The most abundant PAHs in sample C were naphthalene (13%),
357 cyclopenta[def]phenanthrene (11%) and pyrene (8%) with Σ LMW-PAHs/ Σ HMW-PAHs of
358 1.08. Sample D* were dominated by acenaphthene (11.2%), phenanthrene (9.9%) and retene
359 (9.8%) while sample E* was dominated by naphthalene (14.4%), retene (12.6%) and
360 phenanthrene (9.6%). The Σ LMW-PAHs/ Σ HMW-PAHs were 1.85 and 2.30 for samples D and
361 E, respectively (Table S3). A previous study showed that the concentration of pyrene emitted
362 from Chinese style cooking was higher than emitted from other cooking styles (e.g. Japanese)
363 (He et al., 2004).

364 The composition pattern of OPAHs mixtures differed in the various samples (Figure 1). Sample
365 A was dominated by 1,4-chysenequinone (21%), 1-indanone (11.4%) and 9-fluorenone (9.5%),
366 Sample B on the other hand was dominated by 1-indanone (14.4%), 1,4-chysenequinone
367 (13.8%) and 6H-benzo(cd)pyren-6-one (8.3%). 9-fluorenone (14%), 9,10-anthraquinone
368 (12%) and 2-methylanthracene-9,10-dione (9.9%) were the dominant OPAHs in sample C.
369 Sample D* was dominated by 1-indanone (19%), 1,4-naphthoquinone (9.9%), 2-methyl-
370 anthracene-9,10-dione (14%) while sample E* was dominated by 1-indanone (20.2%), 9-
371 fluorenone (13.2%) and 1,4-anthraquinone (12.2%). Many of these found can also be found in
372 other environmental compartments such as ambient air and soil (Clergé et al., 2019). The

373 individual OPAH/parent-PAH ratio was in most cases highest in sample C. The 9,10-
374 anthraquinone/anthracene ratio was >1 for some samples (Table S3). The concentrations of
375 $\Sigma 17$ OPAHs was significantly correlated with $\Sigma 30$ PAHs ($r = 0.91$, $p < 0.05$). Many individual
376 OPAHs were also significantly correlated with their related parent-PAHs. This can be
377 explained by their similar sources and fate.

378 The four individual AZAs targeted in this study were identified and quantified in each of the
379 five samples (Table 3). Sample C had the highest average concentration of the $\Sigma 4$ AZAs, which
380 is similar to all the other PAC groups (Table 3). The contribution of individual AZAs to the
381 $\Sigma 4$ AZAs concentration was slightly different for each of the samples (Figure 1). For sample A,
382 the highest contributions were by carbazole (48%) and quinoline (32%). Quinoline (45%) and
383 benzo[h]quinoline (24%) were the dominant AZAs in sample B. Sample C was dominated by
384 quinoline (38.4%) and carbazole (32.4%). This same trend was in sample D* and E* in which
385 the highest contribution was from quinoline, followed by carbazole (Figure 1). The substances
386 are toxicologically relevant with carbazole being classified as possible human carcinogen
387 (IARC, 2011a; Yamada et al., 2004). None of the samples had individual AZA/parent-PAH
388 concentration ratio >1. The concentration ratios of individual AZA/parent-PAH were mostly <
389 10%, except in a few samples in which the acridine/anthracene and carbazole/fluorene
390 concentration ratio were $\geq 10\%$ (Table S3). These ratios were thus generally lower than the
391 individual OPAH/parent-PAH and individual alkylated PAH/parent-PAH concentration ratios
392 (Table S3). The $\Sigma 4$ AZAs were also significantly correlated with $\Sigma 30$ PAHs ($r=0.83$, $p < 0.05$).
393 This can be explained by the sources and fate of AZAs are similar to those of the PAHs and
394 OPAHs (Bandowe et al., 2016).

395

396 3.4 Concentration of carbonyls

397 The average concentrations of high-molecular-weight (HMW) mono-carbonyl ($C \geq 6$) and di-
398 carbonyl compounds (glyoxal and methylglyoxal) in $PM_{2.5}$ are shown in Table S4. Sample C
399 ($5327.6 \pm 1974.1 \text{ ng/m}^3$) showed the highest total concentration of HMW mono-carbonyl and
400 di-carbonyl compounds, whereas sample B recorded the lowest ($159.2 \pm 48.7 \text{ ng/m}^3$).
401 Nonanaldehyde was the most abundant component in all samples, accounting for ~ 31 - 81% of
402 total carbonyl compounds (Figure S4). The contribution of nonanaldehyde to the carbonyl
403 mixtures was $> 80\%$ in samples A and C. The contribution of each of the other carbonyl
404 compounds to the total mixture was $< 10\%$ in sample A and C. Nonanaldehyde was also the most
405 dominant contributor to the total carbonyl compounds in the other three sampling locations (B,
406 D*, E*) but at these sites methylglyoxal also made a high contribution ($> 10\%$). Nonanaldehyde
407 was typically identified as a dominant carbonyl component in cooking that involves usage of
408 edible oils (Ho et al., 2006). The presence of nonanaldehyde could be due to the decomposition
409 of 9-octadecenoic acid (oleic acid), a known fatty acid produced from cooking oil thermal
410 decomposition (Schauer et al., 2002). A previous study showed that kitchens involved with
411 frequent frying activities (e.g. western fast-food chain shops and Korean barbecue restaurant)
412 could be more abundant in nonanaldehyde (Ho et al., 2006). The two dicarbonyl compounds
413 were detected in all sampling locations, accounting for ~ 0.6 - 26.1% of total carbonyl
414 compounds. Sample A and C showed similar glyoxal (0.7% and 0.6%) and methylglyoxal
415 (3.3% and 2.9%) in their compositions. The highest contribution of decaldehyde to the carbonyl
416 mixtures was observed in sample D* (10.5%) with much lower contributions (0.7 - 4.1%) in the
417 other samples (Figure S4). This observation could be attributed to the types of oils usage in the
418 cooking processes. A previous study showed that different types of seed oils (e.g. soybean and
419 canola oil) could generate ~ 4.77 times difference of decaldehyde in emissions (Schauer et al.,
420 2002), although further study is necessary. The results show that cooking activities are
421 significant anthropogenic source of semi-volatile aldehydes.

422

423 3.5 Relationships between chemical species

424 There were significant correlations between the concentrations of many individual and sums
425 of chemical species (Table S5). For example, the significant correlations between the $\Sigma 30$ PAHs,
426 $\Sigma 17$ OPAHs and $\Sigma 4$ AZAs and the $PM_{2.5}$ mass, TC, EC, OC and Σ carbonyls can be attributed to
427 their similar sources, the fact that they are co-sorbed with each other and hence have similar
428 fates. Some ions like Cl^- and several metals were also strongly correlated with PAHs, OPAHs
429 and AZAs, which might also be strong indication of their similarity in sources but especially
430 similarity in their fates. Unlike Cl^- the correlations between the PACs and SO_4^{2-} , NO_3^- , NH_4^+ ,
431 and Ca^{2+} were not statistically significant indicating that the sources and fate of these ions
432 might be much more different to that of the PACs.

433

434 3.6 Bioreactivity of $PM_{2.5}$

435 The cell viability, oxidative potential (ROS generation), inflammatory reactions (TNF- α) and
436 oxidative DNA damage elicited by $PM_{2.5}$ collected from different cooking sites are shown in
437 Figures 2 and Table S2 (i.e. oxidative DNA damage under particle concentrations of 50, 100,
438 500 and 1000 $\mu g/ml$ dosage). The cell viability of A549 cells demonstrates negative dose-
439 response from all samples (Figure 2). Under the particle concentration of 200 $\mu g/ml$, it can be
440 observed that sample C and D* further showed lowest cell viabilities. Positive dose-response
441 was nevertheless identified in ROS generation and TNF- α (Figure 2) suggesting that oxidative
442 and inflammatory reactions could be enhanced by the increase of particle concentration.
443 Sample C shows the highest ROS generation followed by sample B, whereas sample D* and
444 E* demonstrated higher oxidative potential (under 200 $\mu g/ml$ dosage) in comparison with other
445 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 PM₁₀ derived from coal burning (0-55% under 500
451 µg/ml) (Shao et al., 2016).

452 Median lethal dose (LD₅₀) of the samples (Figure S5) were determined by dosage response
453 analysis in Figure S6 (Supplementary Material). Sample A showed highest LD₅₀ and the lowest
454 LD₅₀ 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.

470 Significant positive correlations were found between the ROS levels and the concentrations
471 Σ 17OPAHs in addition to several individual OPAHs (2-biphenylcarboxaldehyde, 1-
472 acenaphthenone, 9-fluorenone, 9,10-anthraquinone, 2-methylanthraquinone,
473 benzo[a]fluorenone), acridine and decaldehyde (Table 4 and Figure S7). The results of our
474 study are consistent with other studies that show that OPAHs (and other quinones) are redox
475 active chemical species in air PM that are associated with particle induced oxidative potential
476 (Gao et al., 2020; Tuet et al., 2019; Sheng and Lu, 2017; Shang et al., 2013; Bolton et al., 2000).

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

487 None of the (transition) metals which have also been identified as strong inducers of ROS
488 formation were significantly positively correlated with ROS generation in this study (Gao et
489 al., 2020; Bates et al., 2019; Daher et al., 2014; Verma et al., 2009; Hu et al., 2008). It is
490 important to note that the method for determining the concentration of metals in our study (ED-
491 XRF, see Text S2) measures the total concentration and not the water-soluble
492 (bioavailable/bioaccessible fractions). It is the water-soluble concentrations (not the total
493 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, Ca²⁺, SO₄²⁻, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Ni, 14 individual PAHs and
498 Σ 30PAHs; Table 4 and Figure S7). The TNF- α was however significantly negatively correlated
499 with the concentration of more polar PAH derivatives (Σ 4AZAs and Σ 30OPAHs). 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), Σ 30PAHs 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

529 **Acknowledgments**

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

532

533 **References**

- 534 Abdullahi, K.L., Delgado-Saborit, J.M., Harrison, R.M., 2013. Emissions and indoor concentrations of
535 particulate matter and its specific chemical components from cooking: a review. *Atmospheric*
536 *Environment* 71: 260-294. <https://doi.org/10.1016/j.atmosenv.2013.01.061>.
- 537 Allan, J.D., Williams, P.I., Morgan, W.T., Martin, C.L., Flynn, M.J., Lee, J., Nemitz, E., Phillips, G.J.,
538 Gallagher, M.W., Coe, H., 2010. Contributions from transport, solid fuel burning and cooking
539 to primary organic aerosols in two UK cities. *Atmospheric Chemistry and Physics* 10: 647-668.
540 <https://doi.org/10.5194/acp-10-647-2010>.
- 541 Bandowe, B.A.M., Meusel, H., Huang, R., Ho, K., Cao, J., Hoffman, T., Wilcke, W., 2014. PM_{2.5}-
542 bound oxygenated PAHs, 2014. PM_{2.5}-bound oxygenated PAHs, nitro-PAHs and parent-PAHs
543 from the atmosphere of a Chinese megacity: seasonal variation, sources and cancer risk
544 assessment. *Science of the Total Environment* 473-474, 77-87
- 545 Bandowe, B.A.M., Meusel, H., Huang, R., Hoffmann, T., Cao, J., Ho, K., 2016. Azaarenes in fine
546 particulate matter from the atmosphere of a Chinese megacity. *Environmental Science and*
547 *Pollution Research* 23, 16025-16036 <https://doi.org/10.1007/s11356-016-6740-z>.
- 548 Baulig, A., Poirault, J.J., Ausset, P., Schins, R., Shi, T., Baralle, D., Dorlhene, P., Meyer, M., Lefevre,
549 R., Baeza-Squiban A., Marano, F., 2004. Physicochemical characteristics and biological
550 activities of seasonal atmospheric particulate matter sampling in two locations of Paris.
551 *Environmental Science and Technology* 38: 5985-5992. <https://doi.org/10.1021/es049476z>.
- 552 Bein, K.J., Wexler, A.S., 2015. Compositional variance in extracted particulate matter using different
553 filter extraction techniques. *Atmospheric Environment* 107: 24-34.
554 <https://doi.org/10.1016/j.atmosenv.2015.02.026>.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

605 De Oliveira, L.C.C., Vieira, M.A., Ribeiro, A.S., Lisboa, M.T., Gonçalves, R.A., De Campos, R.C.,
606 2012. Determination of silicon in vegetable oil and biodiesel by high-resolution continuum
607 source flame atomic absorption spectrometry using sample dilution with xylene. *Energy &*
608 *Fuels* 26: 7041-7044. <https://doi.org/10.1021/ef3012867>.

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

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

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

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

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

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

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

632 Gysel, N., Dixit, P., Schmitz, D.A., Engling, G., Cho, A.K., Cocker, D.R., Karavalakis, G., 2018.
633 Chemical speciation, including polycyclic aromatic hydrocarbons (PAHs), and toxicity of
634 particles emitted from meat cooking operations. *Science of The Total Environment* 633: 1429-
635 1436. <https://doi.org/10.1016/j.scitotenv.2018.03.318>.

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

639 He, L-Y., Hu, M., Huang, X-F., Yu, B-D., Zhang, Y-H., Liu, D-Q., 2004. Measurement of emissions
640 of fine particulate organic matter from Chinese cooking. *Atmospheric Environment* 38: 6557-
641 6564. <https://doi.org/10.1016/j.atmosenv.2004.08.034>.

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

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

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

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

655 IARC, 2011a. Bitumens and bitumen emissions, and some N- and S-heterocyclic aromatic
656 hydrocarbons. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans 103, 1-
657 311. [https://publications.iarc.fr/Book-And-Report-Series/Iarc-Monographs-On-The-
658 Identification-Of-Carcinogenic-Hazards-To-Humans/Bitumens-And-Bitumen-Emissions-
659 And-Some-Em-N-Em---And-Em-S-Em--Heterocyclic-Polycyclic-Aromatic-Hydrocarbons-
660 2013](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)

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

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

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

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

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

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

678 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.
679 Characteristics of polycyclic aromatic hydrocarbons in PM 2.5 emitted from different cooking
680 activities in China. Environmental Science and Pollution Research 25: 4750-4760.
681 <https://doi.org/10.1007/s11356-017-0603-0>.

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

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

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

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

694 Lui, K.H., Bandowe, B.A.M et al., 2016. Characterization of the chemical components and bioreactivity
695 of fine particulate matter (PM_{2.5}) during incense burning. Environmental Pollution 213, 524-
696 533.

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

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

704 Metayer, C., Wang, Z., Kleinerman, R.A., Wang, L., Brenner, A.V., Cui, H., Cao J., Lubin, J.H., 2002.
705 Cooking oil fumes and risk of lung cancer in women in rural Gansu, China. Lung Cancer 35:
706 111-117. [https://doi.org/10.1016/S0169-5002\(01\)00412-3](https://doi.org/10.1016/S0169-5002(01)00412-3).

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

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

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

717 Nolte, C.G., Schauer, J.J., Cass, G.R., Simoneit, B.R.T., 1999. Highly polar organic compounds present
718 in meat smoke. *Environmental Science & Technology* 33: 3313-3316.
719 <https://doi.org/10.1021/es990122v>.

720 Pardo, M., Shafer, M.M., Rudich, A., Schauer, J.J., Rudich, Y., 2015. Single exposure to near Roadway
721 particulate matter leads to confined inflammatory and defence responses: possible role of
722 metals. *Environmental Science and Technology* 49, 8777-8785.
723 <https://doi.org/10.1021/acs.est.5b01449>

724 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.
725 Genotoxicity of heated cooking oil vapors. *Mutation Research/Genetic Toxicology* 298: 105-
726 111. [https://doi.org/10.1016/0165-1218\(92\)90035-X](https://doi.org/10.1016/0165-1218(92)90035-X).

727 Robberecht, H., Van Dyck, K., Bosscher, D., Van Cauwenbergh, R., 2008. Silicon in Foods: content and
728 bioavailability. *International Journal of Food Properties* 11, 638-645.
729 <https://www.tandfonline.com/doi/full/10.1080/10942910701584252>.

730 Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T., 1991. Sources of fine
731 organic aerosol. 1. Charbroilers and meat cooking operations. *Environmental Science &*
732 *Technology* 25: 1112-1125. <https://doi.org/10.1021/es00018a015>.

733 Roper, C., Chubb, L.G., Cambal, L., Tunno, B., Clougherty, J.E., Fattman, C., Mischler, S.E., 2017.
734 Association of IL-6 with PM 2.5 Components: Importance of Characterizing Filter-Based PM
735 2.5 Following Extraction. *Water, Air, & Soil Pollution* 228: 43. [https://doi.org/10.1007/s11270-](https://doi.org/10.1007/s11270-016-3219-y)
736 [016-3219-y](https://doi.org/10.1007/s11270-016-3219-y).

737 Roper, C., Delgado, L.S., Barrett, D., Massey Simonich, S.L., Tanguay, R.L., 2018. PM2. 5 Filter
738 Extraction Methods: Implications for Chemical and Toxicological Analyses. *Environmental*
739 *science & technology* 53: 434-442. <https://doi.org/10.1021/acs.est.8b04308>.

740 Saito, E., Tanaka, N., Miyazaki, A., Tsuzaki, M., 2014. Concentration and particle size distribution of
741 polycyclic aromatic hydrocarbons formed by thermal cooking. *Food chemistry* 153: 285-291.
742 <https://doi.org/10.1016/j.foodchem.2013.12.055>.

743 Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 1999a. Measurement of emissions from air
744 pollution sources. 1. C1 through C29 organic compounds from meat charbroiling.
745 *Environmental Science & Technology* 33: 1566-1577. <https://doi.org/10.1021/es980076j>

746 Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 2002. Measurement of emissions from air
747 pollution sources. 4. C1– C27 organic compounds from cooking with seed oils. *Environmental*
748 *Science & Technology* 2002; 36: 567-575. <https://doi.org/10.1021/es002053m>.

749 See, S.W., Balasubramanian, R., 2006. Risk assessment of exposure to indoor aerosols associated with
750 Chinese cooking. *Environmental research* 102: 197-204.
751 <https://doi.org/10.1016/j.envres.2005.12.013>.

752 See, S.W., Balasubramanian, R., 2008. Chemical characteristics of fine particles emitted from different
753 gas cooking methods. *Atmospheric Environment* 42: 8852-8862.
754 <https://doi.org/10.1016/j.atmosenv.2008.09.011>.

755 See, S.W., Karthikeyan, S., Balasubramanian, R., 2006. Health risk assessment of occupational
756 exposure to particulate-phase polycyclic aromatic hydrocarbons associated with Chinese,
757 Malay and Indian cooking. *Journal of Environmental Monitoring* 8: 369-376.
758 <https://doi.org/10.1039/B516173H>

759 Seow, A., Poh, W.T., Teh, M., Eng, P., Wang, Y.T., Tan, W.C., Yu, M.C. Lee, H.P., 2000. Fumes from
760 meat cooking and lung cancer risk in Chinese women. *Cancer Epidemiology and Prevention*
761 *Biomarkers* 2000; 9: 1215-1221. <https://cebp.aacrjournals.org/content/9/11/1215>.

762 Shafer, M.M., Hemming, J.D.C., Antkiewicz, D.S., Schauer, J.J., 2016. Oxidative potential of size-
763 fractionated atmospheric aerosol in urban and rural sites across Europe. *Faraday Discussions*
764 189: 381-405. <https://doi.org/10.1039/C5FD00196J>.

765 Shang, Y., Fan, L., Feng, J., Lv, S., Wu, M., Li, B., Zang, Y.S., 2013. Genotoxic and inflammatory
766 effects of organic extracts from traffic-related particulate matter in human lung epithelial A549
767 cells. The role of quinones. *Toxicology in Vitro* 27: 922-931.
768 <https://doi.org/10.1016/j.tiv.2013.01.008>.

769 Shao, L., Hou, C., Geng, C., Liu, J., Hu, Y., Wang, J., Jones, T., Zhao, C., Bérubé, K., 2016. The
770 oxidative potential of PM10 from coal, briquettes and wood charcoal burnt in an experimental

771 domestic stove. *Atmospheric Environment* 127: 372-381.
772 <https://doi.org/10.1016/j.atmosenv.2015.12.007>.

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

777 Shao, L., Shi, Z., Jones, T.P., Li, J., Whittaker, A.G., Berube, K.A., 2006. Bioreactivity of particulate
778 matter in Beijing air: results from plasmid DNA assay. *Science of the Total Environment* 367:
779 261-272. <https://doi.org/10.1016/j.scitotenv.2005.10.009>.

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

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

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

790 Sun, J., Shen, Z., Niu, X., Yu, J., Zhang, Y., Liu, S., Niu, X., Zhang, Y., Xu, H., Li, X.J.A.E., 2020.
791 PM_{2.5} source profiles from typical Chinese commercial cooking activities in northern China
792 and its influences on bioreactivity of vascular smooth muscle cells (VSMCs). *Environmental*
793 *Pollution* 239: 117750.

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

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

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

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

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

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

812 Van Winkle, L.S., Bein, K., Anderson, D., Pinkerton, K.E., Tablin, F., Wilson, D., Wilson, D., Wexler,
813 A.S., 2014. Biological dose response to PM_{2.5}: effect of particle extraction method on platelet
814 and lung responses. *Toxicological Sciences* 143: 349-359.
815 <https://doi.org/10.1093/toxsci/kfu230>.

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

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

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

- 826 Wei, Y., Han, I.-K., Shao, M., Hu, M., Zhang, J., Tang, X., 2009. PM2.5 constituents and oxidative
827 DNA damage in Humans. *Environmental Science and Technology* 43, 4757-4762.
828 <https://doi.org/10.1021/es803337c>.
- 829 Wu, P.F., Chiang, T.A., Wang, L.F., Chang, C.S., Ko, Y.C., 1998. Nitro-polycyclic aromatic
830 hydrocarbon contents of fumes from heated cooking oils and prevention of mutagenicity by
831 catechin. *Mutation Research/Fundamental and Molecular Mechanisms of Mutagenesis* 403: 29-
832 34. [https://doi.org/10.1016/S0027-5107\(98\)00015-3](https://doi.org/10.1016/S0027-5107(98)00015-3).
- 833 Xue, W., Warshawsky, D., 2005. Metabolic activation of polycyclic and heterocyclic aromatic
834 hydrocarbons and DNA damage: A review. *Toxicology and Applied Pharmacology* 206, 73-
835 93. <https://doi.org/10.1016/j.taap.2004.11.006>.
- 836 Yamada, K., Suzuki, T., Kohara, A., Hayashi, M., Mizutani, T., Saeki, K-I., 2004. In vivo mutagenicity
837 of benzo[f]quinoline, benzo[h]quinoline, and 1,7-phenanthroline using *lacZ* transgenic mice.
838 *Mutation Research* 559, 83-95 <https://doi.org/10.1016/j.mrgentox.2003.12.012>.
- 839 Yang, A., Jedynska, A., Hellack, B., Kooter, I., Hoek, G., Brunekreef, B., Kuhlbusch, T.A.J., Cassee,
840 F.R., Janssen, N.A.H., 2014. Measurement of the oxidative potential of PM2. 5 and its
841 constituents: The effect of extraction solvent and filter type. *Atmospheric environment* 83: 35-
842 42. <https://doi.org/10.1016/j.atmosenv.2013.10.049>.
- 843 Zhang, J., Smith, K.R., 1999. Emissions of carbonyl compounds from various cookstoves in China.
844 *Environmental Science & Technology* 33: 2311-2320. <https://doi.org/10.1021/es9812406>.
- 845 Zhang, N., Han, B., He, F., Xu, J., Zhao, R., Zhang, Y., Bai, Z., 2017. Chemical characteristic of PM2.
846 5 emission and inhalational carcinogenic risk of domestic Chinese cooking. *Environmental*
847 *pollution* 227: 24-30. <https://doi.org/10.1016/j.envpol.2017.04.033>
- 848 Zhao, Y., Hu, M., Slanina, S., Zhang, Y., 2007. Chemical compositions of fine particulate organic
849 matter emitted from Chinese cooking. *Environmental Science & Technology* 41: 99-105.
850 <https://doi.org/10.1021/es0614518>.
- 851 Zhong, L., Goldberg, M.S., Gao, Y-T., Jin, F., 1999a. Lung cancer and indoor air pollution arising from
852 Chinese-style cooking among nonsmoking women living in Shanghai, China. *Epidemiology*
853 488-494. <https://pubmed.ncbi.nlm.nih.gov/10468420/>.
- 854 Zhong, L., Goldberg, M.S., Parent, M-É., Hanley, J.A., 1999b. Risk of developing lung cancer in
855 relation to exposure to fumes from Chinese-style cooking. *Scandinavian journal of work,*
856 *environment & health* 309-316. https://www.sjweh.fi/show_abstract.php?abstract_id=440.
- 857 Zhu, L., Wang, J., 2003. Sources and patterns of polycyclic aromatic hydrocarbons pollution in kitchen
858 air, China. *Chemosphere* 50: 611-618. [https://doi.org/10.1016/S0045-6535\(02\)00668-9](https://doi.org/10.1016/S0045-6535(02)00668-9).

859

860

861

862

863

864

865

866

867

868

869 **List of Tables**

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

871 Table 2: Average concentrations \pm standard deviations of PM_{2.5} ($\mu\text{g}/\text{m}^3$), TC ($\mu\text{g}/\text{m}^3$), OC
872 ($\mu\text{g}/\text{m}^3$) and EC ($\mu\text{g}/\text{m}^3$) of inorganic elements (ng/m^3), water extractable ions ($\mu\text{g}/\text{m}^3$) in
873 cooking emission samples.

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

876

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

880

881 **List of Figures and Figure Legends**

882 Figure 1: Mean contributions of individual PAHs, OPAHs, and AZAs to the $\Sigma 30$ PAHs,
883 $\Sigma 17$ OPAHs, and $\Sigma 4$ AZAs concentrations in different sampling locations. The y-axis was
884 broken at 15% to enlarge the scale before the break. Error bars indicate standard deviations for
885 each sample.

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

889

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

896 Table 2: Average concentrations \pm standard deviations of PM_{2.5} ($\mu\text{g}/\text{m}^3$), TC ($\mu\text{g}/\text{m}^3$), OC
 897 ($\mu\text{g}/\text{m}^3$) and EC ($\mu\text{g}/\text{m}^3$) of inorganic elements (ng/m^3), water extractable ions ($\mu\text{g}/\text{m}^3$) in
 898 cooking emission samples.

Component ^a	Sample A	Sample B	Sample C	Sample D*	Sample E*
PM _{2.5}	234.5 \pm 22.1	177.4 \pm 50.6	711.5 \pm 222.6	354.1 \pm 60.8	492.0 \pm 257.9
TC	113.8 \pm 4.2	105.2 \pm 32.6	353.1 \pm 124.1	217.9 \pm 25.5	319.3 \pm 164.9
OC	108.3 \pm 4.2	99.9 \pm 31.5	338.8 \pm 121.1	215.0 \pm 25.1	315.5 \pm 164.4
EC	5.5 \pm 0.7	5.3 \pm 1.5	14.4 \pm 4.0	2.9 \pm 0.5	3.7 \pm 0.9
Elements ^a					
Na	181.9 \pm 21.8	349.8 \pm 106.2	1046.7 \pm 315.1	189.0 \pm 14.4	191.7 \pm 33.5
Mg	93.7 \pm 76.0	44.6 \pm 78.1	156.8 \pm 147.5	64.2 \pm 111.2	49.0 \pm 84.9
Al	73.5 \pm 49.5	89.7 \pm 65.1	171.7 \pm 80.3	84.4 \pm 73.5	45.2 \pm 48.2
Si	169.9 \pm 65.2	163.1 \pm 94.3	279.9 \pm 60.5	267.4 \pm 89.4	312.2 \pm 123.1
S	1276.1 \pm 676.8	1100.5 \pm 262.5	3459.0 \pm 796.3	1575.7 \pm 915.0	476.5 \pm 293.6
Cl	329.9 \pm 55.6	616.3 \pm 210.4	2882.9 \pm 855.0	1078.6 \pm 401.9	3497.1 \pm 2024.4
K	224.5 \pm 99.5	258.7 \pm 131.4	467.1 \pm 139.1	107.4 \pm 42.9	47.9 \pm 22.4
Ca	88.9 \pm 19.9	114.0 \pm 74.6	129.0 \pm 50.4	67.5 \pm 40.0	31.7 \pm 13.6
Ti	8.0 \pm 1.6	7.2 \pm 5.8	5.3 \pm 4.6	4.8 \pm 3.0	2.8 \pm 2.8
V	2.9 \pm 2.9	1.7 \pm 0.7	10.3 \pm 10.5	3.8 \pm 3.1	5.3 \pm 7.0
Cr	4.0 \pm 0.3	4.1 \pm 1.1	7.6 \pm 1.0	5.4 \pm 0.6	6.9 \pm 4.0
Mn	14.8 \pm 3.6	22.2 \pm 9.6	27.4 \pm 14.6	13.7 \pm 3.5	21.1 \pm 5.2
Fe	147.2 \pm 20.5	203.4 \pm 127.4	330.0 \pm 191.5	94.6 \pm 27.1	91.4 \pm 22.1
Co	1.6 \pm 1.8	1.5 \pm 1.8	0.3 \pm 0.6	1.4 \pm 1.4	0.4 \pm 0.7
Ni	1.6 \pm 1.1	3.0 \pm 2.1	6.6 \pm 5.1	3.6 \pm 0.7	3.2 \pm 2.2
Cu	14.7 \pm 4.6	12.3 \pm 5.0	28.1 \pm 11.4	11.3 \pm 3.4	11.4 \pm 3.1
Zn	87.1 \pm 21.7	99.4 \pm 68.9	132.8 \pm 60.6	45.0 \pm 6.3	74.3 \pm 16.7
Sb	16.6 \pm 6.1	22.9 \pm 7.8	39.4 \pm 12.1	33.4 \pm 12.5	48.7 \pm 30.3
Ba	48.5 \pm 8.9	38.8 \pm 16.2	144.3 \pm 37.3	72.6 \pm 18.0	107.9 \pm 62.0
Pb	28.5 \pm 14.3	20.3 \pm 10.5	38.6 \pm 4.8	30.3 \pm 11.1	36.5 \pm 5.2
Ions ^a					
Cl ⁻	0.2 \pm 0.0	0.4 \pm 0.1	3.0 \pm 0.8	0.5 \pm 0.3	2.7 \pm 1.4
NO ₃ ⁻	4.8 \pm 0.9	2.4 \pm 0.7	23.9 \pm 8.5	0.3 \pm 0.0	0.4 \pm 0.1
SO ₄ ²⁻	3.7 \pm 1.6	2.2 \pm 0.6	10.0 \pm 2.6	2.5 \pm 1.1	0.5 \pm 0.5
Na ⁺	0.3 \pm 0.3	0.4 \pm 0.1	1.1 \pm 0.7	0.6 \pm 0.5	0.2 \pm 0.2
NH ₄ ⁺	1.9 \pm 0.5	1.1 \pm 0.3	8.0 \pm 2.4	0.7 \pm 0.4	0.5 \pm 0.1
Ca ²⁺	0.3 \pm 0.0	0.1 \pm 0.0	0.3 \pm 0.1	0.1 \pm 0.1	0.1 \pm 0.1

899 ^aName of the individual component can be referred to Table S1 (Supplementary Material).

900
 901
 902
 903
 904
 905
 906
 907
 908

909 Table 3: The average concentrations \pm 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 ^{a,b}	Sample E [*]
PAHs (pg/m^3)					
1,2,3,4-Tetrahydronaphthalene	224.4 \pm 43.8	135.8 \pm 50.6	435.4 \pm 158.6	242.9 \pm 38.9	330.8 \pm 149.7
Naphthalene	1049.9 \pm 147.9	1021.7 \pm 317.1	2983.5 \pm 923.7	1694.9 \pm 289.8	2363.3 \pm 736.2
2-Methylnaphthalene	212.6 \pm 48.5	193.8 \pm 52.7	542.7 \pm 194.0	342.8 \pm 55.1	409.1 \pm 135.8
1-Methylnaphthalene	256.5 \pm 62.5	218.1 \pm 53.7	617.6 \pm 216.5	412.3 \pm 24.1	921.2 \pm 432.8
Biphenyl	242.3 \pm 55.1	201.0 \pm 58.4	681.1 \pm 208.9	333.3 \pm 39.6	477.8 \pm 146.0
1,3-Dimethylnaphthalene	527.1 \pm 129.1	294.2 \pm 102.9	912.8 \pm 348.4	490.0 \pm 30.1	571.4 \pm 210.2
Acenaphthylene	178.5 \pm 42.1	92.5 \pm 53.5	364.2 \pm 119.2	205.4 \pm 128.8	243.1 \pm 139.2
Acenaphthene	883.1 \pm 204.0	630.5 \pm 237.4	966.0 \pm 398.4	1335.8 \pm 264.2	2096.3 \pm 795.8
Fluorene	217.8 \pm 29.3	192.8 \pm 55.1	734.5 \pm 311.9	346.3 \pm 101.4	481.7 \pm 290.0
Phenanthrene	1028.6 \pm 144.0	603.4 \pm 102.9	2150.5 \pm 871.9	1187.9 \pm 131.1	1615.2 \pm 625.7
Anthracene	127.9 \pm 23.6	82.0 \pm 20.7	273.8 \pm 123.6	190.6 \pm 99.0	129.8 \pm 93.8
4H-Cyclopenta(d,e,f)phenanthrene	692.6 \pm 56.7	550.8 \pm 189.7	2662.8 \pm 1126.2	847.5 \pm 258.8	1125.7 \pm 437.3
1-Methylphenanthrene	179.1 \pm 56.4	99.0 \pm 32.1	481.3 \pm 206.6	192.1 \pm 32.9	315.9 \pm 89.2
3,6-Dimethylnaphthalene	94.3 \pm 19.3	63.7 \pm 21.5	246.5 \pm 100.5	136.9 \pm 4.4	251.6 \pm 105.8
Fluoranthene	249.4 \pm 49.2	285.3 \pm 155.6	888.7 \pm 271.1	273.9 \pm 47.4	296.7 \pm 126.8
Pyrene	475.1 \pm 114.0	1190.9 \pm 1105.9	1917.8 \pm 696.3	617.4 \pm 86.8	814.9 \pm 341.6
Retene	638.3 \pm 148.9	439.5 \pm 132.3	1185.8 \pm 445.5	1175.2 \pm 118.9	2210.6 \pm 1465.4
Benzo[a]anthracene	66.3 \pm 17.7	174.8 \pm 68.6	275.2 \pm 162.1	248.7 \pm 6.8	210.8 \pm 70.0
Chrysene ^a	324.9 \pm 73.2	212.6 \pm 81.3	551.1 \pm 176.5	210.1 \pm 11.9	238.9 \pm 80.5
Benzo[b+j+k]fluoranthenes ^b	625.7 \pm 199.0	823.7 \pm 363.5	1404.4 \pm 541.3	472.1 \pm 104.8	611.9 \pm 208.3
Benzo[e]pyrene	230.1 \pm 78.0	387.0 \pm 243.0	645.6 \pm 346.5	139.9 \pm 46.6	177.4 \pm 60.1
Benzo[a]pyrene	94.1 \pm 33.8	123.8 \pm 67.2	134.5 \pm 82.4	135.0 \pm 32.0	114.1 \pm 35.3
Perylene	23.7 \pm 11.8	42.1 \pm 29.0	1.8 \pm 0.7	19.4 \pm 12.0	6.7 \pm 2.1
Indeno[1,2,3-cd]pyrene	255.2 \pm 43.0	664.0 \pm 171.7	972.4 \pm 920.1	456.6 \pm 253.2	89.8 \pm 36.4
Dibenzo[a,h]anthracene	88.4 \pm 45.1	98.8 \pm 67.8	127.2 \pm 62.6	97.7 \pm 18.3	31.2 \pm 10.3
Benzo[g,h,i]perylene	332.0 \pm 26.0	832.1 \pm 477.7	712.3 \pm 355.7	145.5 \pm 127.4	655.1 \pm 538.9
Coronene	309.0 \pm 114.1	675.5 \pm 419.6	582.8 \pm 373.9	81.7 \pm 44.1	81.2 \pm 22.3
Σ LMW-PAHs ^c	3485.8 \pm 341.3	2622.8 \pm 632.7	7472.6 \pm 2320.5	4960.8 \pm 653.4	6929.3 \pm 2174.9
Σ HMW-PAHs ^d	2511.0 \pm 435.2	4406.1 \pm 1580.9	6983.6 \pm 2056.0	2657.1 \pm 76.4	3063.4 \pm 1079.1
Σ Carci-PAHs ^e	1786.5 \pm 297.0	2929.9 \pm 930.3	4177.1 \pm 1245.3	1765.8 \pm 37.1	1951.8 \pm 724.5
Σ US-EPA PAHs	5996.8 \pm 752.3	7029.0 \pm 2100.8	14456.2 \pm 4183.6	7617.9 \pm 713.6	9992.7 \pm 3253.2
Σ 21 Parent-PAHs	6559.6 \pm 888.1	8133.6 \pm 2608.7	15686.4 \pm 4242.0	7858.9 \pm 775.0	10258.1 \pm 3283.9
Σ 30PAHs	9626.7 \pm 1078.7	10329.7 \pm 3166.4	23452.4 \pm 6498.8	12031.9 \pm 1219.4	16872.1 \pm 5753.7
OPAHs (pg/m^3)					
1-Indanone	65.3 \pm 12.9	68.1 \pm 32.8	264.5 \pm 136.8	226.9 \pm 51.3	642.4 \pm 428.9
1,4-Naphthoquinone	23.7 \pm 5.9	28.2 \pm 14.0	305.9 \pm 257.3	116.2 \pm 27.4	45.6 \pm 26.1
1-Naphthaldehyde	22.1 \pm 10.6	17.5 \pm 8.0	95.8 \pm 68.8	51.8 \pm 16.6	78.9 \pm 40.0
2-Biphenylcarboxaldehyde	8.1 \pm 1.6	7.8 \pm 3.4	71.7 \pm 39.1	23.9 \pm 5.0	236.2 \pm 126.3
1-Acenaphthenone	23.5 \pm 4.1	14.0 \pm 5.7	168.1 \pm 84.4	38.2 \pm 5.9	91.7 \pm 55.2
9-Fluorenone	54.6 \pm 7.8	35.7 \pm 14.9	524.7 \pm 243.3	112.6 \pm 5.1	423.1 \pm 264.4
9,10-Anthraquinone	41.5 \pm 7.0	38.4 \pm 17.8	413.1 \pm 165.9	103.6 \pm 29.7	261.6 \pm 185.6
1,8-Naphthalic anhydride	41.9 \pm 2.3	25.9 \pm 14.9	216.8 \pm 82.6	23.8 \pm 6.2	105.0 \pm 66.9
1,4-Anthraquinone	25.5 \pm 5.3	42.2 \pm 24.6	217.4 \pm 111.4	96.3 \pm 28.4	359.6 \pm 116.3
4H-Cyclopenta[d,e,f]phenanthrenone	7.8 \pm 1.5	10.2 \pm 5.9	101.3 \pm 46.9	12.6 \pm 2.1	38.5 \pm 29.5
2-Meth-9,10-anthraquinone	47.7 \pm 13.5	26.4 \pm 16.1	317.1 \pm 133.1	165.7 \pm 18.6	355.8 \pm 243.3
Benzo[a]fluorenone	22.5 \pm 4.2	16.8 \pm 7.8	216.1 \pm 98.8	38.0 \pm 7.1	84.9 \pm 36.2
7H-Benzo[d,e]anthracene-7-one	17.8 \pm 2.3	32.1 \pm 14.2	259.9 \pm 121.6	22.8 \pm 7.0	52.4 \pm 27.3
Benzo[a]anthracene-7,12-dione	15.2 \pm 2.6	10.1 \pm 4.2	112.4 \pm 45.5	12.2 \pm 2.1	25.2 \pm 8.2
1,4-Chrysenequinone	140.7 \pm 106.6	80.1 \pm 60.9	291.1 \pm 152.6	114.9 \pm 28.0	149.3 \pm 6.3
5,12-naphthacenequinone	14.7 \pm 3.2	8.4 \pm 5.3	57.1 \pm 24.9	18.6 \pm 1.7	105.3 \pm 108.8
6H-Benzo[c,d]pyran-6-one	12.5 \pm 3.1	40.7 \pm 21.7	66.8 \pm 42.1	13.2 \pm 6.2	58.6 \pm 65.1
Σ 17OPAHs	585.2 \pm 135.6	502.7 \pm 249.4	3700.1 \pm 1735.3	1191.1 \pm 190.8	2944.5 \pm 1494.4
AZAs (pg/m^3)					
Quinoline	15.3 \pm 3.5	14.1 \pm 5.8	97.9 \pm 62.9	51.9 \pm 14.2	83.0 \pm 44.8
Benzo(h)quinoline	5.9 \pm 0.7	7.0 \pm 2.4	41.9 \pm 28.0	28.8 \pm 14.5	33.6 \pm 21.7
Acridine	3.3 \pm 1.5	4.4 \pm 3.2	36.0 \pm 20.6	18.2 \pm 6.0	35.3 \pm 24.5
Carbazole	26.8 \pm 17.4	7.2 \pm 5.8	87.5 \pm 52.3	38.9 \pm 24.7	49.0 \pm 45.5
Σ 4AZAs	51.3 \pm 19.3	32.7 \pm 15.7	263.3 \pm 160.0	137.8 \pm 39.3	200.8 \pm 133.0

911 ^a represents chrysene + triphenylene; ^b represents benzo[b]fluoranthene + benzo[j]fluoranthene

912 + benzo[k]fluoranthene; ^c Σ LMW-PAHs: low molecular weight PAHs with 2-3 aromatic rings;

913 ^d Σ HMW-PAHs: high molecular weight PAHs with 4-7 aromatic rings; ^e Σ Carci-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

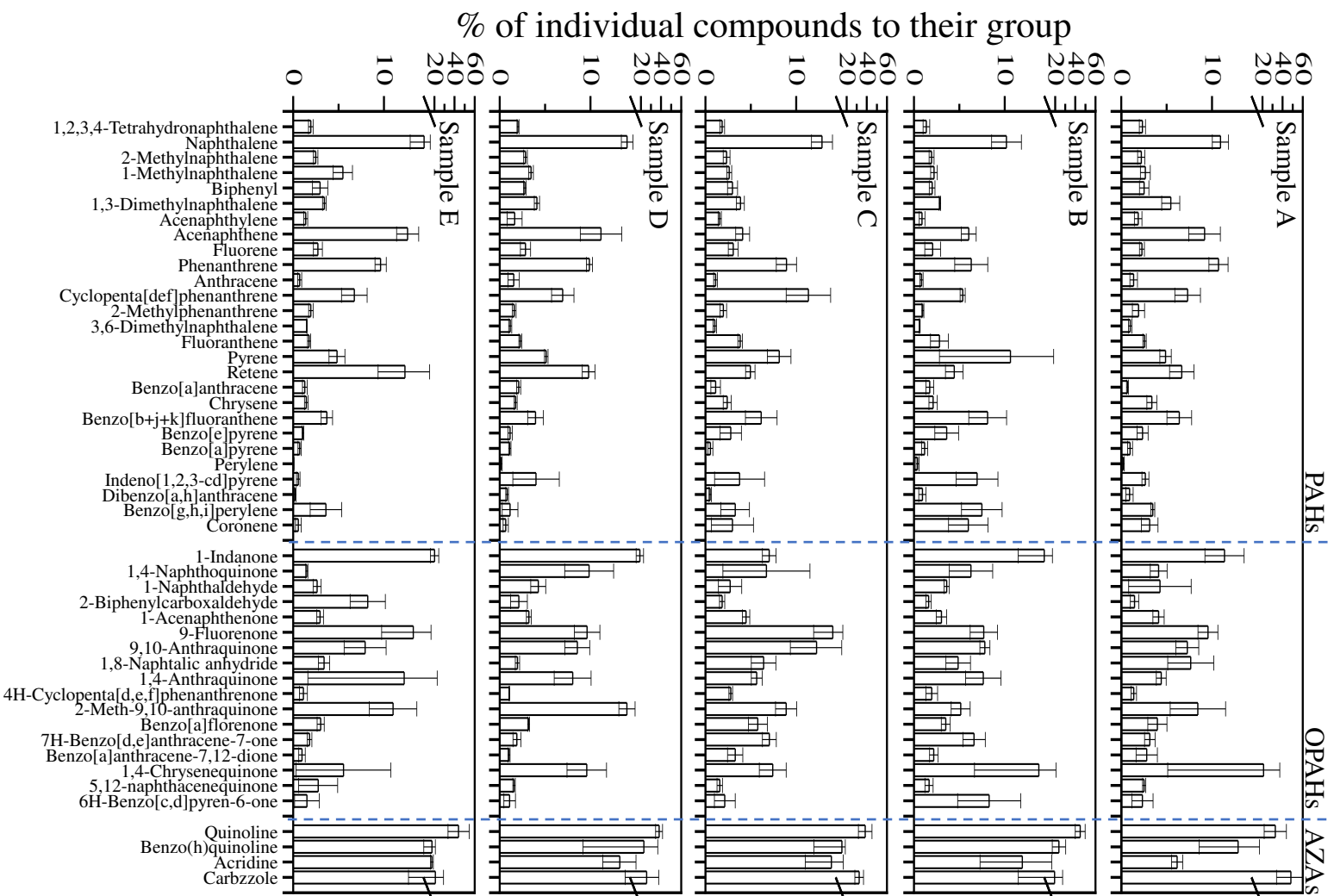
Chemical Species	Cell viability	ROS generated (DCFH)	TNF- α	DNA damage
TC	-0.102	-0.239	-0.336	0.307
OC	-0.101	-0.158	-0.423	0.233
EC	-0.028	-0.502 ^{ab}	0.824**	0.325
Cl ⁻	0.327	0.367	-0.386	-0.292
NO ₃ ⁻	0.216	0.151	0.437	-0.239
SO ₄ ²⁻	-0.148	-0.176	0.671**	-0.046
Na ⁺	-0.346	-0.238	0.389	0.393
NH 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	<i>-0.593**</i>	0.822**	0.447
Benzo [b+j+k] fluoranthene	-0.153	<i>-0.660**</i>	0.873**	0.474*
Benzo[e]pyrene	-0.210	<i>-0.549*</i>	0.896**	0.346
Benzo[a]pyrene	-0.336	<i>-0.789**</i>	0.580*	0.763**
Perylene	-0.308	<i>-0.888**</i>	0.688**	0.740**
Indeno [1,2,3-cd] pyrene	-0.615**	<i>-0.478*</i>	0.743**	0.248
Dibenzo[ah]anthracene	-0.456	<i>-0.677**</i>	0.818**	0.599**
Benzo[ghi]perylene	0.081	<i>-0.696**</i>	0.660**	0.690**
Coronene	-0.152	<i>-0.587*</i>	0.853**	0.465
Quinoline	0.005	0.378	<i>-0.603**</i>	-0.247
1-Indanone	0.104	0.178	<i>-0.631**</i>	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**	<i>-0.796**</i>	-0.339
1-Acenaphthenone	0.435	0.711**	<i>-0.637**</i>	<i>-0.516*</i>
9-Fluorenone	0.409	0.739**	<i>-0.756**</i>	<i>-0.495*</i>
Benzo[h]quinoline	-0.144	0.266	-0.445	-0.219
Acridine	0.044	0.581*	<i>-0.733**</i>	-0.358
Carbazole	0.174	0.424	-0.283	-0.196
9,10-Anthraquinone	0.234	0.661**	<i>-0.577*</i>	<i>-0.471*</i>
1,8-Naphthalic anhydride	<i>0.524*</i>	0.446	-0.072	-0.382
1,4-Anthraquinone	0.173	0.335	<i>-0.621**</i>	-0.114
4H-Cyclopenta[def]phenanthrenone	0.046	0.444	-0.133	-0.314
2-Methylanthracene-9,10-dione	0.187	0.686**	<i>-0.884**</i>	-0.418
Benzo[a]fluorenone	0.359	0.719**	<i>-0.591**</i>	-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	<i>0.665**</i>	0.415	<i>-0.541*</i>	-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	<i>-0.876**</i>	0.745**	0.763**
Σ17OPAHs	0.371	0.474*	<i>-0.617**</i>	-0.223
Σ4AZAs	0.067	0.419	<i>-0.558*</i>	-0.124

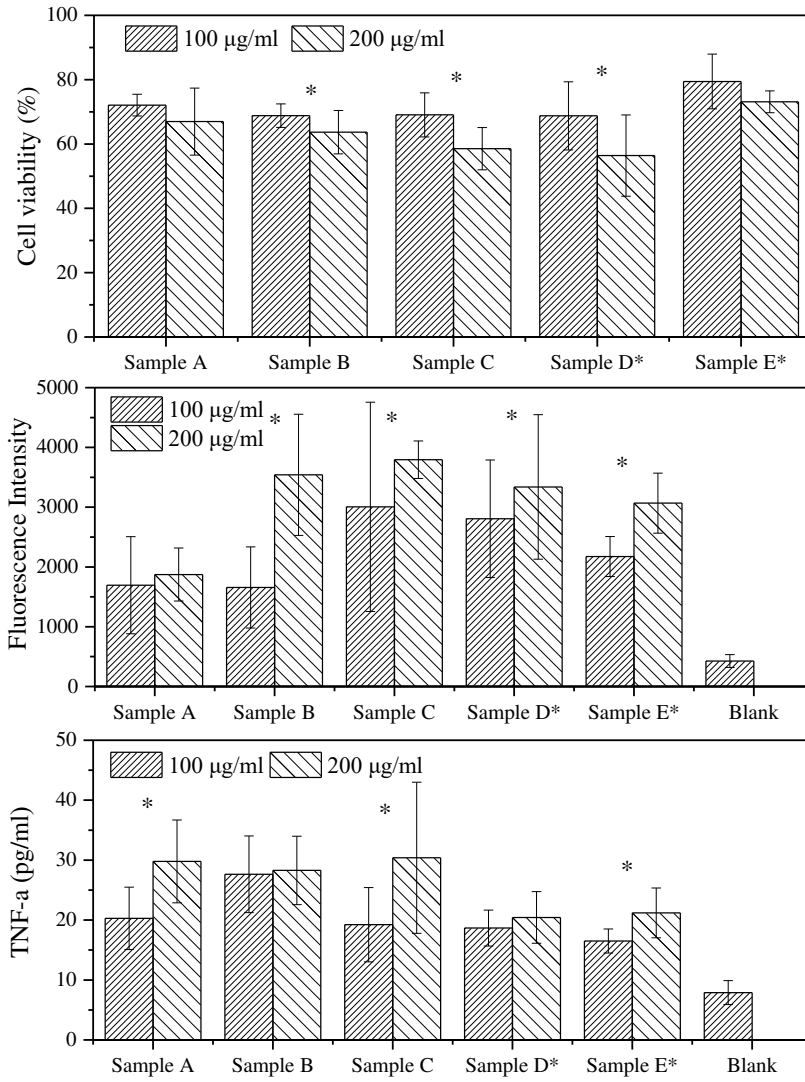
920 ^{a**}Correlation is significant at the 0.01 level (2-tailed).

921 ^{b*} Correlation is significant at the 0.05 level (2-tailed).

922



923
 924 Figure 1: Mean contributions of individual PAHs, OPAHs, and AZAs to the Σ 30PAHs,
 925 Σ 150PAHs, and Σ 4AZAs concentrations in different sampling locations. The y-axis was
 926 broken at 15% to enlarge the scale before the break. Error bars indicate standard deviations for
 927 each sample.
 928



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 \pm standard
 932 deviation

933

934