Supplementary Material

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

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Text S1: Organic carbon (OC) and elemental carbon (EC) analysis

OC and EC were analysed on a punch (0.526 cm^2) from quartz filter by thermal optical reflectance (TOR) technique following the IMPROVE_A protocol on a thermal/optical carbon analyser (DRI Model 2001, Atmoslytic Inc., Calabasas, CA). The detection limit of EC and OC were below 1.0 μg m^{-3}. Details of the chemical analysis can be referred to Pathak et al. (2011).

Text S2: Analysis of inorganic elements and water soluble ions

Teflon-membrane filter samples were sent to the Institute of Earth Environment, Chinese Academy Sciences (IEECAS, Xi'an, China) in a temperature controlled package (< 4 °C) for elemental analysis by an Energy Dispersive X-Ray Fluorescence analyzer (ED-XRF, Epsilon 5, PANalytical Company, Almelo, The Netherlands) (Watson et al.,1999; Chow and Watson, 2012). Twenty inorganic elements (Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Sb, Ba and Pb) returned concentrations exceeding method detection limits (MDL) for >50% of the samples, and these elements are included in data analysis. Field blanks were analyzed following the same procedure. MDLs were within the range of 0.5 to 33 ng/m^3. The concentrations of ions were determined in the aqueous extracts of the filter samples. One-fourth of each quartz filter were placed into a separate 15 mL vials containing 10 mL distilled–

deionized water (18.2 MΩ resistivity). The vails were placed in ultrasonic water bath for 60 min and then shaken by mechanical shaker for complete extraction. The extracts were then filtered with a 0.45 μm pore size microporous membrane, and the filtrates were stored at 4°C in a clean tube until analysis. A Dionex-600 Ion Chromatograph (Dionex Inc., Sunnyvale, CA, USA) was used for the determinations of cations and anions in the aqueous extracts. IonPacCS12A column (20 mM methanesulfonic acid as the eluent) was used for cation analyses, and IonPac AS14A column (8 mM Na_2CO_3/1 mM $NaHCO_3$ as the eluent) was used for anions analysis. The method detection limits were: 4.6 mg L^{-1} for Na^+, 4.0 mg L^{-1} for NH_4^+, 0.5 mg L^{-1} for Cl^-,10.0 mg L^{-1} for K^+, Mg^{2+} and Ca^{2+} and NO_3^-, and 20 mg L^{-1} for SO_4^{2-}. The blank values were subtracted from sample concentrations. One sample in each group of ten samples was analyzed twice for quality control.

Text S3: Determination of polycyclic aromatic compounds (PACs)

Each quartz filter (with the sampled $PM_{2.5}$) was cut into smaller pieces and transferred into a 33 mL accelerated solvent extractor (ASE) extraction cell and spiked with 40 μL of a mixture seven deuterated-PAHs (naphthalene-D8, acenaphthene-D10, phenanthrene-D10, pyrene-D10, chrysene-D12, perylene-D12 and benzo[ghi]perylene-D12 each at a concentration of 10 μg/mL), and 40 μL of mixture of three deuterated compounds [benzophenone-D5 (20 μg/mL), 9,10-anthraquinone-D8 (20 μg/mL) and carbazole-D8 (5μg/mL)]. The seven deuterated PAHs,

the two oxygenated compounds (benzophenone-D5 & 9,10-anthraquinone-D8) and carbazoleD8 served as the internal standards for the quantification of the PAHs, OPAHs and AZAs, respectively. Then inert bulk sorbent (Isolute HMN, Biotage, Uppsala, Sweden) were used to fill up the extra space of the extraction cells. Blanks samples (n =2) made of bulk sorbent alone were also transferred into ASE extraction cells and spiked with same amount of deuterated internal standards as with the samples. Each sample was and extracted twice by pressurized liquid extraction (ASE 200, Dionex, Sunnyvale, CA, USA) with the same ASE instrument parameters as previously outlined (Bandowe and Wilcke, 2010; Bandowe et al., 2014; Bandowe et al., 2016). The organic solvent used in the first extraction of each sample was dichloromethane. Each sample was extracted a second time with acetone:dichloromethane mixture (2:1 v/v). Extracts from each sample were combined and transferred into a turbo-vap extraction tube, 10 ml of hexane and 3 drops of toluene was then added. The extracts were then concentrated on a Turbo Vap evaporating system (at 35°C) until a volume < 1 mL. Each sample was then transferred into a 2 ml GC-vial. Floranthene-D10 (25 µL 22 µg/mL) was spiked into some of the extracts in the GC-vials to serve as recovery standards. The target compounds (27 PAHs, 18 OPAHs and 4 azaarenes, Table S2) in each extract were measured with an Agilent 7890N gas chromatograph coupled to an Agilent 5975C mass selective detector (GC-MS) operating in selected ion monitoring mode. The quantification of the target compounds were performed with an internal standard methods with the deuterated compounds spiked into each

86 sample before extraction. Further information about the analysis method can be referred to in
87 previous articles (Bandowe and Wilcke, 2010; Bandowe et al. 2014a,b; Bandowe et al., 2016;
88 Lui et al., 2016).
89 High-purity solvents (pesticide residue grade) were used for all extractions, rinsings, and
90 preparation of standards. Glassware and metallic parts of ASE cells, and other labware were
91 machine-washed and baked at 250 °C. Prior to usage, glassware was further rinsed with high-
92 purity solvents. Target compounds were determined in blanks which were analyzed with the
93 analytical procedure as the samples. The average amount of the target compounds that were
94 found in the blanks were deducted from that of the same compound found in the sample to
95 correct for laboratory contamination and field contamination. Limit of detection was defined as
96 a mass of target compound three times greater than the baseline noise (S/N = 3). The recovery
97 of the deuterated internal standards spiked to the samples before extracted were determined as
98 indicator of the accuracy of the analytical measurement. The recoveries (mean ± standard
99 deviation) of the deuterated internal standards were: naphthalene-D8 (72±8%), acenaphthene-
100 D10 (73±8%), phenanthrene-D10 (73±8%), pyrene-D10 (76±7%), chrysene-D12 (78±6%),
101 perylene-D12 (81±12%), benzo(ghi)perylene-D12 (88±14%), benzophenone-D5 (42±4%),
102 9,10-anthraquinone-D8 (55±5%) and carbazole-D8 (46±4%).
103 The accuracy and precision of the analytical method for target PAHs, alkyl PAHs, OPAHs and
104 and azaarenes were checked by a spike and recovery experiments (n = 3) and reported in

previous articles (Bandowe et al., 2014; Bandowe et al., 2016). Average recoveries of targeted PAHs/alkyl PAHs and OPAHs were 102% (range: 67 to 154%) and 96% (64 to 152 %), respectively (Bandowe et al., 2014). The relative standard deviation (RSD) for the PAHs were 5% (range: 0.7 to 10%) and 9% (range: 2 to 31%) for PAHs/alkyl-PAHs and OPAHs respectively. (Bandowe et al., 2014). The recoveries (mean ±standard deviation) of azaarenes in spike and recovery experiment were 75±5%, 87±6% and 54±5 %, for quinoline, benzo[h]quinoline and acridine, respectively (Bandowe et al., 2016). The method we applied in the current study is only a slight modification of the method applied in the previous works. The solvent used for the extraction in the current study was DCM (first extraction) followed by acetone: DCM (2:1 v/v). This extraction solvent was already applied to extract same target PACs from $PM_{2.5}$ on filters (Lui et al., 2016). Since the method applied in the current study is very similar to the previous study (Bandowe et al., 2014; Bandowe et al., 2016), the accuracy and precision can be gauged from the results of the previous spike and recovery study (Bandowe et al., 2014; Bandowe et al., 2016).

Text S4: Determination of carbonyls

Each sample ($PM_{2.5}$ on the filters) were transferred into separate in 50 mL Falcon tubes and extracted with 20 mL ultrapure methanol (HPLC grade, Sigma-Aldrich Corporation, USA) on an ultrasonic bath (Branson 5510E-DTH, 40 kHz) containing water at 25 °C for 20 minutes.

The extract was transferred to a round-bottom flask and evaporated by rotary evaporator (RV10 Basic Rotary Evaporators, IKA Works, VWR, USA) at 30 °C until 5 ml remained. The concentrated extract from each sample was transferred to Eppendorf vials and purged with nitrogen at room temperature until dryness. The dried extract was stored at -20 °C until the analysis of carbonyls. In summary, the dried sample extracts (containing the carbonyl compounds) were re-dissolved in in water to a concentration of 1 mg/L. An excess amount of O-(2,3,4,5,6-pentafluorobenzyl)hydroxylamine hydrochloride (PFBHA) in aqueous solution (e.g., 0.5 ml of 5 mg/ml solution) was added to 5 ml of the extracts. The target carbonyls in the extracts react with the PFBHA to form derivatives. The solution containing the PFBHA-derivatives was then acidified to pH 2 and allowed to stand for 24 hours at room temperature. This solution was then extracted with 2 ml hexane, separated from the aqueous layer and dried with 50 mg anhydrous Na_2SO_4. Finally, 1 µl of analyte dissolved in hexane was transferred into a vial for gas chromatography–mass spectrometry (GC/MS) analysis. The analysed carbonyls are shown in Table S2 (Supplementary Material). Detailed description of the method for the derivatization of carbonyls with PFBHA and the subsequent GC/MS analytical procedure was described previously (Yu et al., 1995). Further experimental details were described previously (Dai et al., 2012).

Text S5: Cell culture and cell viability

PM sample were prepared using two-stage sonication of the Teflon filter in methanol and followed by drying under nitrogen air (Totlandsdal et al., 2012). The PM samples were re-suspended in dimethyl sulfoxide (DMSO) (<0.01 vol/vol %) and mixed with Roswell Park Memorial Institute (RPMI) cell culture medium before being used for exposure of cells. Control sample was prepared by a blank filter as the sample preparation described above.

A549 cells were obtained from the American Type Culture Collection (ATCC) and cultured using RPMI cell culture medium (Thermo Fisher Scientific Inc., MA, USA) supplemented with 10% heat-inactivated fetal bovine serum (Biowest, MO, USA) and 1% antibiotics penicillin/streptomycin (100 U mL^{-1}), in a humidified incubator supplied with 5% carbon dioxide (CO_2) at 37 °C. A549 cells were seeded onto inserts in 24-well transwells at a density of 1×10^5 cells ml^{-1} and incubated for 24 h. The cell medium was removed and replaced with 300 µl of the prepared samples for the next 24 h. Each experiment was conducted in quadruplicate.

Cell viability were determined by MTT (3-[4, 5-dimethylthiazol-2-yl]-2, 5 diphenyl tetrazolium bromide) assay. After $PM_{2.5}$ exposure for 24 hrs, the supernatant was collected. Then, 100 ml of 10% MTT solution (Sigma Aldrich, St. Louis, MO, USA) were added at 37 °C for 4 h for color development. Optical density was measured at 540 nm by absorbance microplate reader (ELx800, BioTek, VT, USA). The results were presented as a percentage of the absorbance of control.

Text S6: Reactive Oxygen Species (ROS) and TNF-α analysis

Cellular ROS was determined by the fluorogenic cell-based method using 2′,7′-dichlorodihydrofluorescein diacetate (DCFH-DA) as the indicator, which has been commonly used for environmental toxicology (Eruslanov and Kusmartsev, 2010; Montesinos et al., 2015). After 24 hours exposure to $PM_{2.5}$, DCFH-DA was added to the A549 cells, and cultured for 30 min. Each well was washed with PBS to remove the DCFH-DA that did not combined with cells. The fluorescence intensity (IF) was determined by a Light Luminescence Plate Reader (VICTOR™ X; PerkinElmer, Waltham, USA) at an excitation wavelength of 485 nm and an emission wavelength of 530 nm.

Enzyme-linked immunosorbent assay (ELISA) (R&D Systems, Inc., MN, USA) was used to determine tumor necrosis factor-α (TNF-α) levels in supernatant according to the manufacturer's instructions (Chuang et al., 2018).

For PSA, gels (0.6% Agarose; Bioline, UK) were prepared using Tris/Borate/EDTA (TBE) buffer solution (Thermo Scientific, UK) diluted 10 times with agarose and the solution was heated by microwave (EMS-820; Electron Microscopy Services, USA) to clarity and transparency. The solidified gel was placed in an electrophoresis cell (DYCP-34A type; NANBEI, China) containing 10 times diluted TBE buffer.

Bromophenol blue stain (14 μL; Sigma-Aldrich, UK) was added to the DNA-$PM_{2.5}$ samples

and placed on a rocking platform (Bio-Rad, UK) for 4 hours. Post-mixing, 20μL of the DNA-$PM_{2.5}$ mixtures were aliquoted into each gel well. Three parallel samples were made for each sample. Ethidium bromide (EB; 20 μL; Sigma-Aldrich, China) was added to both sides of the electrophoresis tank (NANBEI, China). After the EB was fully dissolved in the buffer, the laboratory electrophoresis power supply (DYY-6C; NANBEI, China) was turned on and operated at 30 Volts for 16 hours.

Post-electrophoresis, the optical densities of three different DNA morphologies (i.e. super-coiled, relaxed and linear) in the gel were captured using a gel documentation system (ChemiDoc, Bio-Rad, UK) and the GeneTools (Version 4.3.10; Syngene, USA) image analysis software program was utilized to calculate the toxic dose of $PM_{2.5}$ causing DNA damage (%). Additional information about the PSA procedure can be found in Chuang et al., (2011).

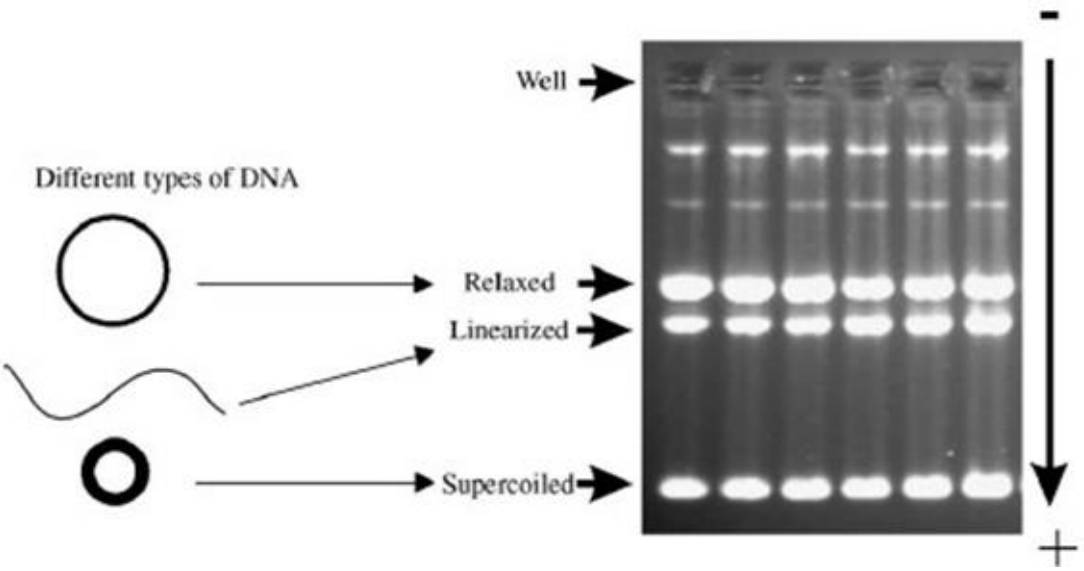


Figure S1: Identification of different types of plasmid ΦX174-RF DNA (Promega, London, UK) in gel electrophoresis.

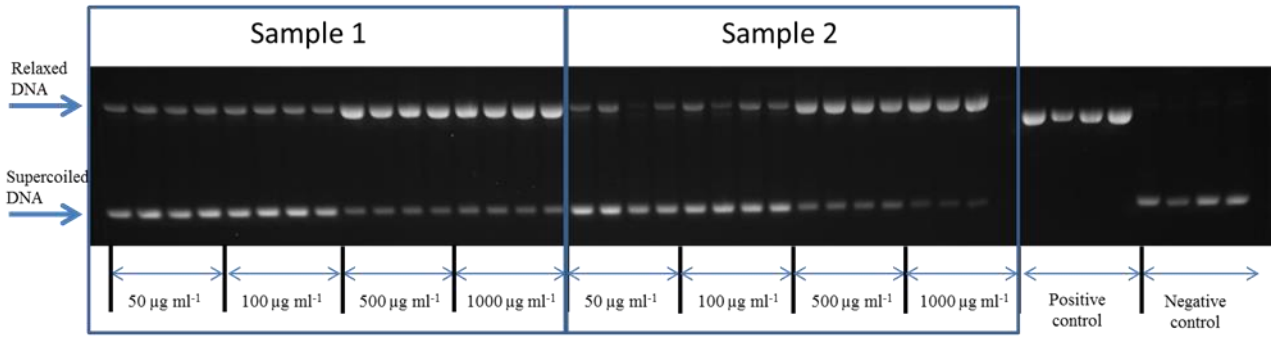


Figure S2: Gel images demonstrate oxidative damage to supercoiled DNA induced by $PM_{2.5}$ sample.

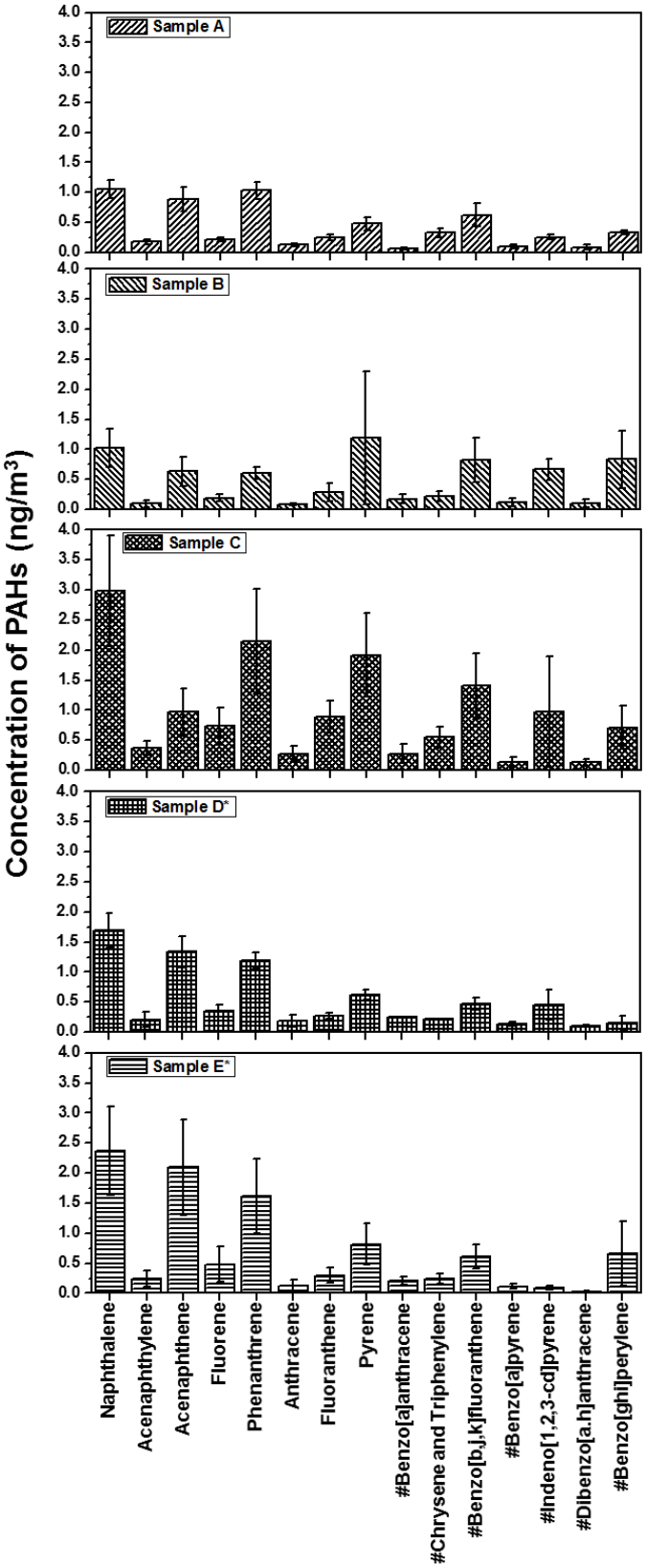
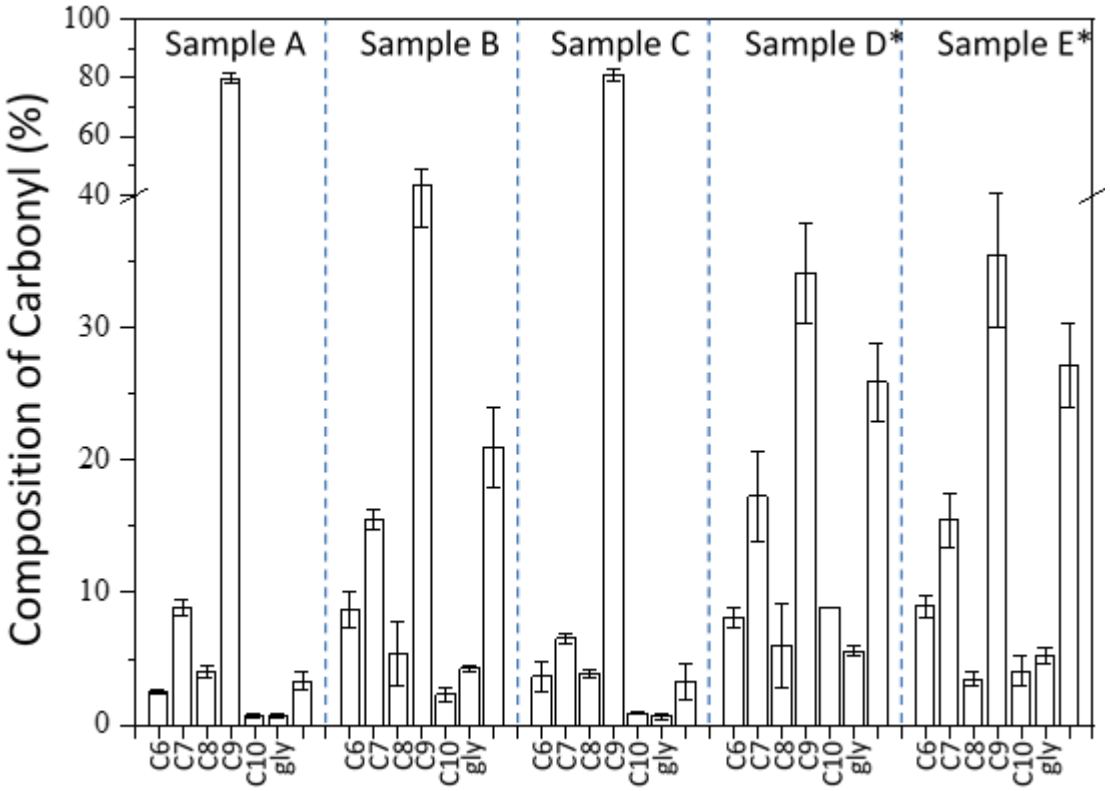


Figure S3: Descriptive analysis and relative abundances of U.S. EPA Priority PAHs. Hash (#) indicated by U.S. EPA as probable human carcinogen.



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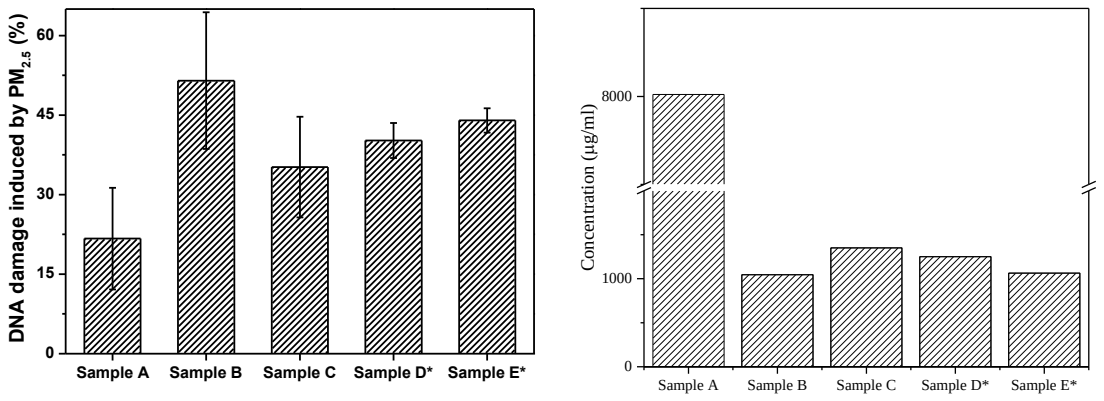
206 **Figure S4:** Mean contributions of individual carbonyl compounds to the total concentration of
207 carbonyls in samples from different sampling locations. The y-axis was broken at 40% to
208 enlarge the scale before the break. Error bars indicate standard deviations for each sample.

209



210

211 **Figure S5:** The average DNA damage induced by extracts of $PM_{2.5}$ (1000 μg/ml) and LD_{50} of
212 $PM_{2.5}$ collected from five sampling locations (n = 4 for Sample A, B, C and E* and n = 3 for
213 sample D*).

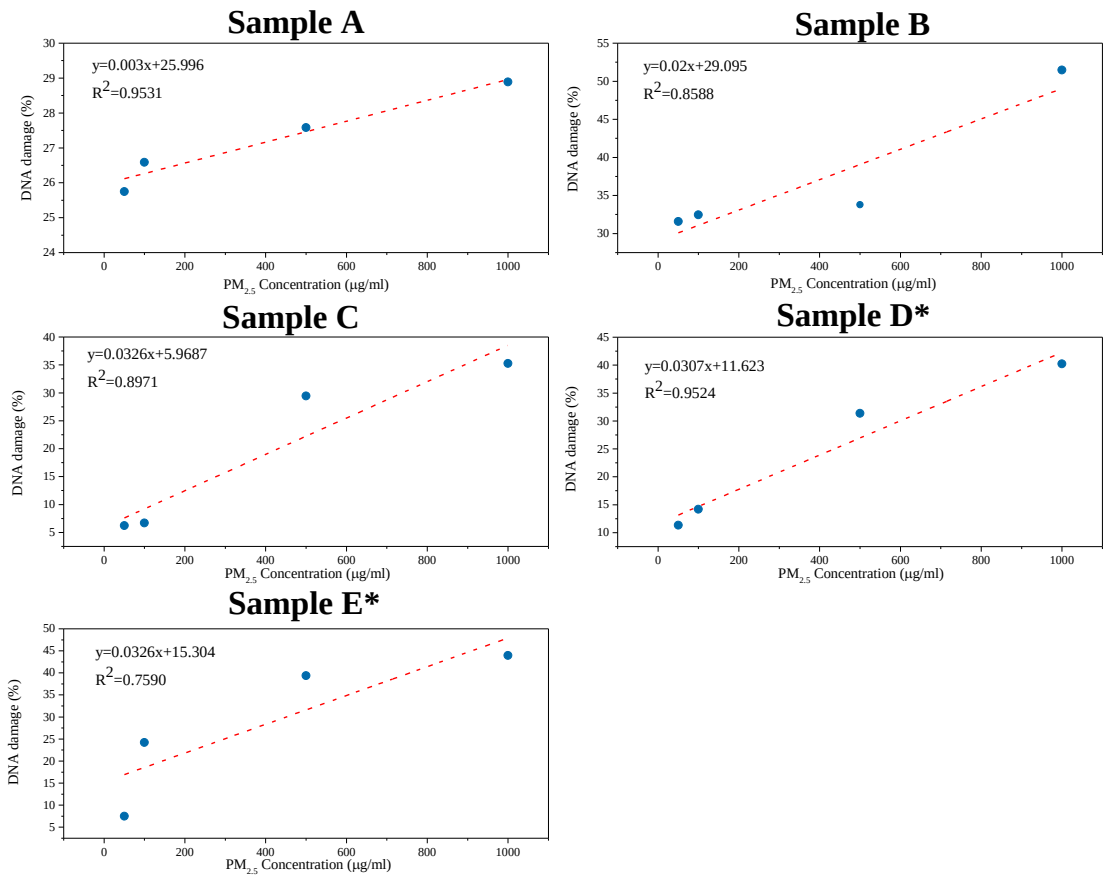
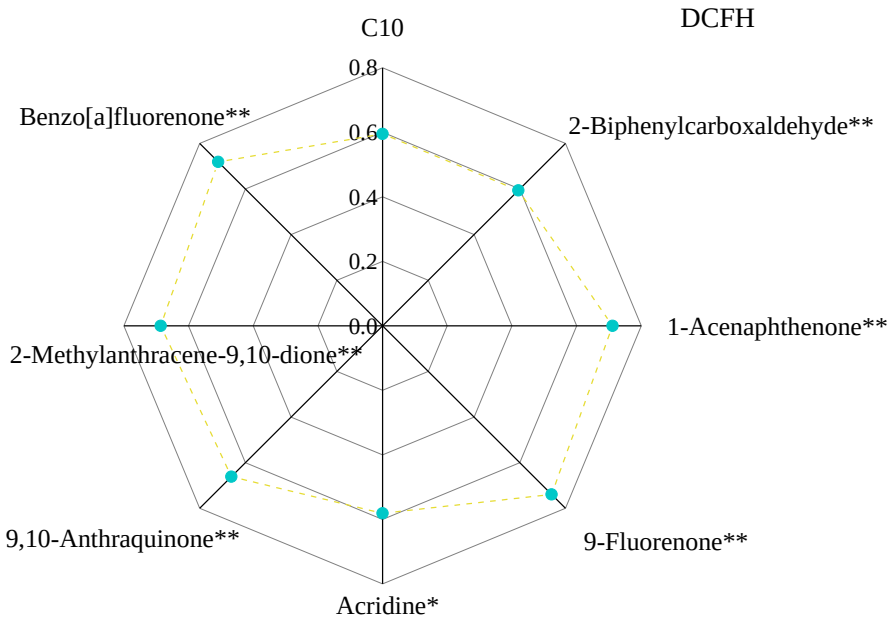
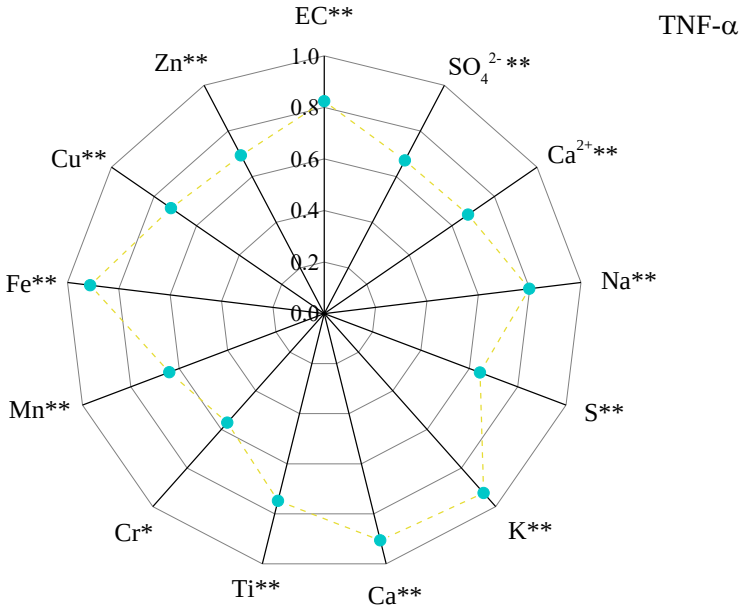


Figure S6: Dose response analysis between DNA damage and $PM_{2.5}$ concentration using plasmid scission assay (PSA).



221



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223 **Figure S7**: Correlations between the concentrations of chemical species extracted from $PM_{2.5}$
224 and biological effects [ROS generation (DCFH) and TNF-α].
225

226

227 **Table S1:** List of chemical species analyzed in samples their abbreviations

Group				
Carbonaceous Species	Organic carbon (OC)	Elemental carbon (EC)		
Ions	Sodium ion (Na^+) Nitrate (NO_3^-)	Calcium ion (Ca^{2+}) Sulfate (SO_4^{2-})	Ammonium (NH_4^+)	Chloride (Cl^-)
& Inorganic Elements	Sodium (Na) Sulfur (S) Zine (Zn) Vanadium (V) Aluminum (Al) Barium (Ba)	Potassium (K) Chromium (Cr) Nickel (Ni) Stibium (Sb) Silicon (Si) Calcium (Ca)	Calcium (Ca) Titanium (Ti) Lead (Pb) Cobalt (Co) Manganese (Mn)	Chlorine (Cl) Iron (Fe) Magnesium (Mg) Rubidium (Rb) Copper (Cu)
Polycyclic Aromatic Hydrocarbons (PAHs) and Alkyl-PAHs	1,2,3,4-Tetrahydronaphthalene (THNAPH)	Naphthalene (NAPH)	2-Methylnaphthalene (2-MNAPH)	1-Methylnaphthalene (1-MNAPH)
	Biphenyl (BP)	1,3-Dimethylnaphthalene (1,3-DMNAPH)	Acenaphthylene (ACENY)	Acenaphthene (ACEN)
	Fluorene (FLUO)	Phenanthrene (PHEN)	Anthracene (ANTH)	4H-Cyclopenta(d,e,f)phenanthrene (CPHEN)
	1-Methylphenanthrene (1-MPHEN)	3,6-Dimethylphenanthrene (3,6-DMPHEN)	Fluoranthene (FLUA)	Pyrene (PYR)
	Retene (RET)	Benz[a]anthracene (B(A)A)	Chrysene and Triphenylene (CHRY)	Benzo[b+j+k]fluoranthene (B(BJK)
	Benzo[e]pyrene (B(E)P)	Benzo[a]pyrene (B(A)P)	Perylene (PERY)	Indeno[1,2,3-cd]pyrene (IND)
	Dibenz[a,h]anthracene (DIBE)	Benzo[ghi]perylene (B(GHI)	Coronene (COR)	
Oxygenated-Polycyclic Aromatic Hydrocarbons (OPAHs)	1-Indanone (1-INDA)	1,4-Naphthoquinone (1,4-NQ)	1-Naphthaldehyde (1-NLD)	2-Biphenylcarboxaldehyde (2-BIP)
	1-Acenaphthenone (1-ACENONE)	9-Fluorenone (9-FLO)	9,10-Anthraquinone (9,10-ANQ)	1,8-Naphthalic anhydride (1,8-ANA)
	1,4-Anthraquinone (1,4-ANQ)	4H-Cyclopenta[d,e,f]phenanthrene-4-one (CPHENONE)	2-Methylanthracene-9,10-dione (2-METH)	Benzo[a]fluorenone (B(A)FLUONE)
	7H-Benz[de]anthracene-7-one (BANTONE)	Benz[a]anthracene-7,12-dione (7,12-B(A)A)	1,4-Chrysenequinone (1,4-CHQ)	Naphthacene-5,12-dione (NAP-5,12)
	6H-benzo[c,d]pyrene-6-one (BPYRONE)			
Azaarenes	Quinoline (QUI)	Benzo[h]quinolone (BQI)	Acridine (ACR)	Carbazole (CBZ)
Carbonyls	Hexaldehyde (C6)	Heptaldehyde (C7)	Octaldehyde (C8)	Nonaldehyde (C9)
	Decaldehyde (C10)	Glyoxal (gly)	Methylglyoxal (mgly)	

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Table S2: DNA damage induced by the $PM_{2.5}$ emitted from five sampling locations

	Concentration (µg/ml)	Average DNA damage (%)	Concentration (µg/ml)	Average DNA damage (%)	Concentration (µg/ml)	Average DNA damage (%)	Concentration (µg/ml)	Average DNA damage (%)
Sample A	1		2		3		4	
(sub-sample number 1-4)	50	25.3	50	21.3	50	20.1	50	36.3
	100	35.7	100	35.7	100	16.9	100	18.1
	500	24.0	500	26.8	500	24.6	500	34.9
	1000	15.5	1000	37.8	1000	13.7	1000	19.9
Sample B	5		6		7		8	
(sub-sample number 5-8)	50	29.5	50	36.0	50	25.7	50	35.1
	100	30.3	100	29.9	100	37.4	100	32.2
	500	25.9	500	43.8	500	30.7	500	34.8
	1000	33.4	1000	61.1	1000	65.8	1000	45.6
Sample C	9		10		11		12	
(sub-sample number 9-12)	50	7.1	50	11.4	50	2.3	50	4.0
	100	10.7	100	6.9	100	4.9	100	4.2
	500	44.9	500	33.6	500	18.0	500	21.3
	1000	40.6	1000	48.2	1000	26.8	1000	25.5
Sample D*[a]	13		14		15			
(sub-sample number 13-15)	50	6.4	50	7.7	50	19.8		
	100	7.8	100	16.2	100	18.6		
	500	29.8	500	30.9	500	33.5		
	1000	40.6	1000	44.1	1000	36.0		
Sample E*	16		17		18		19	
(sub-sample number 16-19)	50	4.4	50	7.7	50	4.7	50	13.2
	100	21.3	100	19.7	100	28.7	100	27.1
	500	36.6	500	35.7	500	41.9	500	43.6
	1000	44.3	1000	46.8	1000	40.4	1000	44.4

[a]* represents samples collected from stainless steel environmental chamber that mimic residential kitchen hood condition.

231 **Table S3:** Selected mean concentration ratios (±standard deviations) of PAHs, OPAHs, AZAs in different samples.

	Sample A	Sample B	Sample C	Sample D*	Sample E*
2-MNAPH/NAPH	0.20±0.05	0.19±0.01	0.18±0.02	0.20±0.01	0.17±0.02
1-MNAPH/NAPH	0.25±0.07	0.22±0.02	0.20±0.01	0.25±0.03	0.38±0.08
1,3-DMNAPH/NAPH	0.50±0.10	0.28±0.04	0.30±0.04	0.29±0.03	0.24±0.02
ΣAlkyl-NAPH/NAPH	0.45±0.12	0.41±0.03	0.38±0.02	0.45±0.04	0.55±0.07
2-MPHEN/PHEN	0.18±0.05	0.16±0.03	0.22±0.02	0.16±0.02	0.20±0.03
3,6-DMPHEN/PHEN	0.09±0.02	0.10±0.03	0.12±0.02	0.12±0.01	0.16±0.01
ΣAlkyl-PHEN/PHEN	0.27±0.07	0.27±0.06	0.34±0.03	0.28±0.02	0.36±0.03
PHEN/(PHEN+ANTH)	0.89±0.03	0.88±0.01	0.89±0.01	0.87±0.05	0.93±0.02
B(A)A/(B(A)A+CHR)	0.17±0.03	0.45±0.02	0.31±0.08	0.54±0.02	0.47±0.02
FLUA/(FLUA+PYR)	0.35±0.02	0.24±0.10	0.32±0.04	0.31±0.01	0.27±0.02
IND/(IND+B(GHI))	0.43±0.03	0.48±0.16	0.50±0.26	0.73±0.28	0.14±0.05
∑LMW-PAHs/∑HMW-PAHs	1.41±0.16	0.65±0.18	1.08±0.18	1.86±0.25	2.30±0.14
Σ6alkPAHs/Σ21PAHs	0.29±0.04	0.17±0.03	0.25±0.03	0.35±0.02	0.44±0.05
Σ6AlkylPAHs/Σ29PAHs	0.20±0.02	0.13±0.02	0.17±0.01	0.23±0.01	0.27±0.03
Σ17OPAHs/Σ29PAHs	0.06±0.02	0.05±0.01	0.15±0.05	0.10±0.01	0.16±0.04
Σ17OPAHs/Σ21PAHs	0.09±0.03	0.06±0.02	0.22±0.09	0.15±0.01	0.27±0.07
B(E)P/B(A)P	2.60±1.11	3.03±0.24	5.76±2.83	1.02±0.10	1.56±0.24
B(GHI)/B(A)P	3.76±0.85	6.63±2.54	7.38±5.99	0.99±0.66	5.18±2.74
1,4-NQ/NAPH	0.02±0.00	0.03±0.01	0.09±0.08	0.07±0.02	0.02±0.01
1-NLD/1-MNAPH	0.08±0.03	0.08±0.03	0.14±0.08	0.12±0.04	0.09±0.04
9-FLO/FLUO	0.25±0.05	0.19±0.08	0.67±0.23	0.34±0.10	0.81±0.28
9,10-ANQ/ANTH	0.34±0.14	0.45±0.19	1.51±0.18	0.58±0.14	1.88±0.64
7,12-B(A)A/B(A)A	0.23±0.05	0.06±0.02	0.43±0.09	0.05±0.01	0.12±0.03
1-ACEQ/ACEN	0.03±0.00	0.02±0.00	0.16±0.05	0.03±0.00	0.04±0.02
1-ACEQ/ACENY	0.14±0.06	0.16±0.05	0.43±0.17	0.22±0.10	0.35±0.07
1,4-CHRQ/CHR	0.49±0.46	0.36±0.33	0.50±0.27	0.55±0.17	0.52±0.47
QUI/NAPH	0.01±0.00	0.01±0.00	0.03±0.02	0.03±0.00	0.03±0.02
BQI/PHEN	0.01±0.00	0.01±0.00	0.02±0.01	0.03±0.02	0.02±0.01
ACR/ANTH	0.03±0.01	0.05±0.03	0.12±0.05	0.10±0.03	0.26±0.08
CBZ/FLUO	0.12±0.07	0.03±0.02	0.10±0.05	0.10±0.06	0.08±0.05

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Table S4: The average concentrations ± standard deviation (ng/m^3) of carbonyls in five sampling locations

Compound[a]	Sample A	Sample B	Sample C	Sample D*	Sample E*
Hexaldehyde	23.8±5.5	13.4±2.6	199.8±88.3	21.6±7.1	37.4±19.1
Heptaldehyde	83.2±15.1	24.9±8.5	355.7±143.7	44.0±3.7	62.2±25.0
Octaldehyde	38.2±8.1	8.3±4.7	203.4±64.8	16.9±12.9	13.7±4.6
Nonaldehyde	757.1±168.2	69.8±25.0	4333.9±1681.7	88.3±14.1	153.2±98.7
Decaldehyde	6.6±2.4	3.3±1.5	48.1±16.9	29.5±2.1	16.8±9.7
Glyoxal	6.3±1.0	6.7±2.1	32.9±8.6	14.4±2.5	21.1±7.7
Methylglyoxal	30.8±5.3	32.8±10.5	153.8±26.6	66.6±7.2	107.6±39.0
ΣCarbonyls	946.0±170.4	158.4±42.2	5327.6±1709.6	261.7±49.3	412.1±164.2

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

Table S5: Correlation coefficients between the concentrations of PACs and carbon fractions, ion, metal and carbonyl in all samples.

	∑30PAHs	∑16PAHs	∑21PAHs	ΣLMW-PAHs	ΣHMW-PAHs	ΣAlkyl-NAP	ΣAlkyl-PHEN	Σ6alkPAHs	∑4AZAs	∑17OPAHs
$PM_{2.5}$	0.92**	0.89**	0.85**	0.95**	0.60**	0.91**	0.93**	0.93**	0.92**	0.94**
TC	0.90**	0.87**	0.83**	0.95**	0.56*	0.93**	0.92**	0.95**	0.92**	0.95**
OC	0.90**	0.87**	0.82**	0.95**	0.55*	0.93**	0.91**	0.95**	0.92**	0.95**
EC	0.55*	0.55*	0.60**	0.34	0.70**	0.26	0.44	0.26	0.31	0.38
Cl^-	0.80**	0.77**	0.75**	0.77**	0.57*	0.82**	0.77**	0.76**	0.70**	0.83**
NO_3^-	0.32	0.33	0.39	0.10	0.57*	-0.003	0.20	-0.003	0.06	0.13
SO_4^{2-}	0.14	0.16	0.21	-0.04	0.41	-0.17	0.02	-0.15	0.004	-0.02
Na^+	0.55*	0.57*	0.60*	0.38	0.64**	0.26	0.35	0.28	0.44	0.38
NH_4^+	0.42	0.42	0.46	0.27	0.57*	0.22	0.37	0.19	0.19	0.30
Ca^{2+}	0.13	0.12	0.16	0.02	0.27	-0.04	0.13	0.03	-0.03	0.03
Al	0.70**	0.70**	0.71**	0.64*	0.60*	0.54*	0.61*	0.61*	0.62*	0.59*
Si	0.71**	0.66**	0.64**	0.77**	0.37	0.79**	0.70**	0.79**	0.69**	0.76**
Cl	0.79**	0.76**	0.73**	0.79**	0.51*	0.87**	0.78**	0.81**	0.73**	0.84**
Cr	0.84**	0.83**	0.81**	0.79**	0.65**	0.73**	0.73**	0.75**	0.80**	0.82**
Ni	0.63**	0.61**	0.61**	0.56*	0.52*	0.50*	0.52*	0.48	0.64**	0.60*
Sb	0.82**	0.81**	0.78**	0.80**	0.57*	0.83**	0.69**	0.79**	0.78**	0.82**
Pb	0.63**	0.58*	0.56*	0.71**	0.29	0.66**	0.64**	0.67**	0.61**	0.69**
ΣCarbonyls	0.67**	0.63**	0.64**	0.62**	0.55*	0.49*	0.71**	0.55*	0.55*	0.62**

** indicate significant correlations at the p = 0.01 level (2-tailed).

* indicate significant correlations at the p = 0.05 level (2-tailed).

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