

Supporting Information

Urinary Metabolites of Polycyclic Aromatic Hydrocarbons and their Association with Lipid Peroxidation: A Biomarker-Based Study between Los Angeles and Beijing

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Table S1. Information of OH-PAHs standards, the monitored ions of methylated OH-PAHs in GC-MS (EI) and the method detection limits of OH-PAHs

Compound	Abbreviation	Ions monitored	Detection Limits (pg mL ⁻¹)	Purity	Source
1-hydroxynaphthalene	1-OH-NAP	158, 159	14.9	98%	Dr. Ehrenstrofer ^a
2-hydroxynaphthalene	2-OH-NAP	158, 159	13.6	99%	Dr. Ehrenstrofer
2-hydroxybiphenyl	2-OH-BP	184, 185	13.0	97.7%	AccuStandard ^b
3-hydroxybiphenyl	3-OH-BP	184, 185	8.6	100%	AccuStandard
4-hydroxybiphenyl	4-OH-BP	184, 185	18.2	100%	AccuStandard
2,2'-dihydroxybiphenyl	2,2'-DOH-BP	214, 215	11.9	100%	AccuStandard
3,4-dihydroxybiphenyl	3,4-DOH-BP	214, 215	12.3	100%	AccuStandard
4,4'-dihydroxybiphenyl	4,4'-DOH-BP	214, 215	9.7	100%	AccuStandard
2-hydroxydibenzofuran	2-OH-DBF	198, 199	10.8	98.3%	Chiron AS ^c
2-hydroxyfluorene	2-OH-FLU	196, 197	7.5	98%	Aldrich ^d
3-hydroxyfluorene	3-OH-FLU	196, 197	12.3	N.A. ^f	Aldrich
1-hydroxyphenanthrene	1-OH-PHE	208, 209	8.8	99%	Dr. Ehrenstrofer
2-hydroxyphenanthrene	2-OH-PHE	208, 209	17.0	99%	Dr. Ehrenstrofer
3-hydroxyphenanthrene	3-OH-PHE	208, 209	12.8	99%	Dr. Ehrenstrofer
4-hydroxyphenanthrene	4-OH-PHE	208, 209	9.9	99.6%	Dr. Ehrenstrofer
1-hoxypyrene	1-OH-PYR	217, 218	14.3	100%	AccuStandard
(SS) ¹³ C ₆ -3-hydroxyphenanthrene	¹³ C ₆ -3-OH-PHE	214, 215		≥95%	CIL ^e
(IS) d ₁₀ -anthracene	d ₁₀ -ANT	188, 189		99.2%	AccuStandard
(IS) d ₁₀ -pyrene	d ₁₀ -PYR	212, 213		99.1%	AccuStandard

^a. Augsburg, Germany; ^b. New Haven, CT, USA; ^c. Trondheim, Norway; ^d. St. Louis, MO, USA;

^e. Andover, MA, USA^f. Notavailable.

Table S2. The influence of individual characteristic and physical activities on urinary OH-PAHs based on multivariate regression model with step-wise approach

Pollutants ^a	Gender	Age	BMI	City	CD ^b	BD ^c	BI ^d	BFI ^e	DD ^f	HD ^g	PD ^h	ED ⁱ
1-OH-NAP	0.6	0.61	0.15	<0.001	0.89	0.29	0.21	0.39	0.24	0.61	0.07	0.43
2-OH-NAP	<0.001	0.14	0.76	0.89	0.36	0.05	0.23	0.75	0.98	0.67	<0.05	<0.01
2-OH-BP	<0.05	<0.05	<0.05	0.11	0.51	0.93	0.34	0.91	<0.05	0.34	0.28	<0.05
4-OH-BP	0.24	0.8	0.11	<0.001	0.84	0.1	0.34	0.18	0.82	0.98	0.48	0.46
4,4'-DOH-BP	0.83	0.77	<0.05	<0.001	0.84	0.25	0.3	0.24	0.85	0.59	0.09	<0.05
2-OH-DBF	0.24	0.62	<0.001	<0.001	0.34	0.15	0.48	0.22	0.92	0.44	0.76	0.08
2-OH-FLU	0.37	0.86	<0.001	<0.001	0.54	0.22	0.39	0.34	0.95	0.74	0.68	0.11
3-OH-FLU	0.84	1	<0.001	<0.001	0.48	0.26	0.25	0.31	0.51	0.37	0.14	<0.05
1-OH-PHE	0.99	<0.05	<0.001	<0.001	0.73	0.05	0.15	0.8	0.94	0.78	0.6	0.23
2-OH-PHE	0.09	0.78	<0.05	<0.001	0.82	0.32	0.66	0.53	0.69	0.81	0.43	0.38
4-OH-PHE	0.36	0.7	0.99	<0.001	0.21	0.38	0.42	0.69	0.3	0.79	0.69	0.23
1-OH-PYR	0.48	0.33	0.26	<0.001	0.58	0.27	0.51	<0.05	0.55	0.46	0.95	0.23
ΣOH-NAPs	<0.05	0.34	0.4	0.11	0.86	0.22	0.24	0.85	0.34	0.07	0.41	<0.05
ΣOH-BPs	0.54	0.09	<0.01	<0.001	0.9	0.27	0.7	0.41	0.09	0.11	0.64	<0.01
ΣOH-FLUs	0.85	0.14	<0.001	<0.001	0.13	<0.05	0.07	0.28	0.66	0.41	0.32	0.09
ΣOH-PHEs	0.84	<0.05	<0.01	<0.001	0.18	<0.05	<0.05	0.43	0.64	0.56	0.75	0.32
Σ ₈ OH-PAHs	<0.05	0.45	0.44	<0.01	0.68	0.12	0.5	0.38	0.39	0.61	0.32	0.15
Σ ₁₂ OH-PAHs	<0.05	0.25	0.21	<0.001	0.73	0.14	0.59	0.29	0.28	0.35	0.38	<0.05

^a Data was log-transformed before the analysis; ^b CD – Cooking duration (h); ^c BT – Barbecue duration (h); ^d BI – Barbecue intake (pound); ^e BFI – Baked food intake (pound); ^f DD – Driving duration (h); ^g HT – Duration near heavy traffic(h); ^h PD – Duration in public traffic transportation (h); ⁱ ED – Duration in environmental tobacco smoke (h).

Table S3. Comparison between the physical activities in Los Angeles and Beijing

	In Los Angeles (n=57)	In Beijing (n=47)	<i>p</i> -value ^a
Cooking Duration(h)	0.86±0.99 ^b	0.00±0.02	<0.01
Barbecue Duration (h)	0.16±0.52	0.00±0.00	<0.05
Barbecue intake (pound)	0.23±0.48	0.26±0.59	0.73
Baked food intake (pound)	0.54±0.78	0.66±0.56	<0.05
Driving Duration (h)	0.67±1.14	0.16±0.67	<0.01
Duration near heavy traffic	0.62±0.99	4.03±3.49	<0.01
Duration in public transportation (h)	0.58±1.21	3.00±4.18	<0.01
Duration in environmental tobacco smoke (h)	0.02±0.09	1.49±2.39	<0.01

a. Mann-Whitney U-test, two-tailed

b. Mean±standard deviation

Table S4 Descriptive statistics of biomarkers urine samples in Beijing (BJ) and Los Angeles before (LA₁) and after the trip(LA₂)

Biomarker	Median (IQR ^a)			p-value ^b			Beijing/LA Ratio(95%CI;p-value) ^c
	In LA ₁ (n=30)	In BJ(n=47)	In LA ₂ (n=27)	LA ₁ vs. BJ	LA ₂ vs. BJ	LA ₁ vs. LA ₂	
Exposure biomarker (ng/mL)							
1-hydroxynaphthalene	0.86 (0.15, 1.81)	2.37 (0.95, 4.40)	0.51 (0.16, 1.09)	<0.001	<0.001	0.51	4.1 (2.3-7.3; <0.001)
2-hydroxynaphthalene	1.92 (0.78, 5.69)	5.20 (2.08, 6.46)	2.57 (0.80, 5.52)	<0.05	<0.05	0.79	1.7 (1.1-2.6; <0.05)
∑hydroxynaphthalenes	3.47 (1.08, 8.53)	6.95 (3.78, 13.1)	2.95 (0.89, 7.30)	<0.01	<0.01	1.00	2.1 (1.3-3.2; <0.01)
2-hydroxybiphenyl	0.44 (0.28, 0.66)	0.53 (0.39, 1.22)	0.64 (0.46, 0.94)	<0.05	0.84	<0.05	1.7 (1.1-2.5; <0.05)
4-hydroxybiphenyl	0.24 (0.13, 0.39)	1.65 (0.88, 3.32)	0.25 (0.09, 0.67)	<0.001	<0.001	0.73	5.8 (3.8-8.7; <0.001)
4,4'-dihydroxybiphenyl	0.27 (0.12, 0.56)	1.44 (0.65, 2.43)	0.41 (0.07, 0.70)	<0.001	<0.001	0.61	4.2 (2.7-6.7; <0.001)
∑hydroxybiphenyls	1.04 (0.55, 1.72)	3.77 (2.17, 6.75)	1.34 (0.74, 2.98)	<0.001	<0.001	0.14	3.6 (2.4-5.2; <0.001)
2-hydroxydibenzofuran	0.18 (0.10, 0.37)	2.19 (1.33, 4.53)	0.30 (0.12, 0.58)	<0.001	<0.001	0.31	9.5 (6.5-13.7; <0.001)
2-hydroxyfluorene	0.14 (0.09, 0.29)	1.75 (0.80, 2.97)	0.26 (0.09, 0.43)	<0.001	<0.001	0.39	9.0 (6.1-13.3; <0.001)
3-hydroxyfluorene	0.06 (0.04, 0.09)	0.57 (0.33, 1.08)	0.07 (0.02, 0.15)	<0.001	<0.001	0.64	8.6 (5.6-13.2; <0.001)
∑hydroxyfluorenes	0.21 (0.13, 0.40)	2.24 (1.12, 4.02)	0.28 (0.11, 0.58)	<0.001	<0.001	0.41	8.8 (6.0-13.0; <0.001)
1-hydroxyphenanthrene	0.08 (0.05, 0.24)	0.70 (0.30, 1.09)	0.11 (0.05, 0.23)	<0.001	<0.001	0.97	6.1 (4.0-9.3; <0.001)
2-hydroxyphenanthrene	0.05 (0.03, 0.11)	0.35 (0.21, 0.56)	0.07 (0.03, 0.10)	<0.001	<0.001	0.74	5.5(3.9-7.7; <0.001)
4-hydroxyphenanthrene	0.04 (0.01, 0.08)	0.18 (0.07, 0.29)	0.05 (0.02, 0.10)	<0.001	<0.001	0.53	4.2 (2.7-6.6; <0.001)
∑hydroxyphenanthrenes	0.19 (0.10, 0.47)	1.26 (0.49, 2.04)	0.28 (0.09, 0.49)	<0.001	<0.001	0.82	5.4 (3.7-7.9; <0.001)
1-hydroxypyrene	0.06 (0.03, 0.13)	0.51 (0.16, 0.94)	0.09 (0.04, 0.18)	<0.001	<0.001	0.52	5.2 (3.3-8.2; <0.001)
∑ ₈ hydroxylated PAHs ^d	4.05 (1.27, 8.76)	11.6 (5.89, 20.9)	3.43 (1.20, 8.27)	<0.001	<0.001	0.89	2.8(1.8-4.2; <0.001)
∑ ₁₂ hydroxylated PAHs ^e	5.51 (2.17, 10.4)	18.1 (10.3, 33.7)	6.58 (2.04, 11.1)	<0.001	<0.001	0.51	3.2 (2.2-4.7; <0.001)
Effect biomarker (ng/mL)							
malondialdehyde	27.9 (14.3, 52.3)	49.4 (20.2, 92.4)	36.1 (16.1, 62.8)	<0.05	0.25	0.24	1.5 (1.0-2.0; <0.05)

^a. IQR: interquartile range; ^b. Mann-Whitney test; ^c. Calculated by multivariate linear regression models with enter approach, adjusted by age, gender and BMI;

^d. Sum of OH-NAPs, OH-FLUs, OH-PHEs, and 1-OH-PYR; ^e.Sum of OH-PAHs, OH-BPs, and 2-OH-DBF

Table S5. Pearson correlation among different OH-PAHs and metabolite ratios (data was log-transformed)

	ΣOH-BPs	2-OH-DBF	ΣOH-FLUs	ΣOH-PHEs	1-OH-PYR	1-/2-OH-NAP	1+2-/4-OHPHE
ΣOH-NAPs	0.56**	0.52**	0.57**	0.58**	0.58**	-0.13	-0.01
ΣOH-BPs		0.77**	0.76**	0.77**	0.68**	0.20*	0.11
2-OH-DBF			0.97**	0.91**	0.83**	0.33**	0.19*
ΣOH-FLUs				0.93**	0.86**	0.34**	0.22*
ΣOH-PHEs					0.86**	0.30**	0.15
1-OH-PYR						0.26**	0.14
1-/2-OH-NAP							0.11

**: $p < 0.01$; *: $p < 0.05$

Table S6. Summary of the studies cited to investigate the relationship between naphthalene exposure and 1-/2-OH-NAP ratio

No.	Exposure characteristic	Population description	Sample size	Male/Female	Smokers/No n-smokers	ΣOH-NAPs (µg/g creatinine)	1-/2-OH -NAP	Ref.
Study1	Biomass burning	Cooking women (pre-intervention)	47	0/47	0/47	34.2	1.30	15
		Cooking women (post-intervention)	47	0/47	0/47	20.3	0.97	15
Study2	Biomass burning	Cooking women (pre-intervention)	57	0/57	0/57	39.0	1.04	16
		Cooking women (post-intervention)	57	0/57	0/57	19.6	0.96	16
Study3	Coking processes	Tar distillation operators	69	N/A ^a	56/13	1220	2.62	17
		Naphthalene oil distillation workers	33	N/A	33/0	1079	1.84	17
		Unexposed workers	36	25/11	28/8	15.8	1.70	17
Study4	Bitumen vapor	bitumen-exposed workers(pre+post shift)	112	112/0	0/112	4.38	0.77	18
		Roadside construction workers(pre+post shift)	240	240/0	0/240	5.82	0.60	18
Study5	Ambient environment	Healthy people visiting a certain hospital	273	273/0	140/133	5.60	0.55	1
Study6	Coking processes	Works in distillation department	29	29/0	N/A	2204	1.75	19
		Sorting department gas fitting operators	11	11/0	N/A	677	1.89	19
Study7	Aluminum plant	Resident within 1km from the plant (Sep. 14, 2005)	65	N/A	0/65	4.65	0.92	20
		Resident within 1km from the plant (Sep. 21, 2005)	68	N/A	0/68	4.05	0.62	20
		Resident within 1km from the plant (May 23+24, 2006)	109	N/A	0/109	4.22	0.40	20
		Resident within 1km from the plant (May 30+31, 2006)	111	N/A	0/111	3.81	0.31	20

Table S6. Summary of the studies cited to investigate the relationship between naphthalene exposure and 1-/2-OH-NAP ratio (continued)

		Resident living more than 11km from the plant (Sep. 14, 2005)	62	N/A	0/62	3.14	0.80	20
		Resident living more than 11km from the plant (Sep. 21, 2005)	66	N/A	0/66	4.31	0.57	20
		Resident living more than 11km from the plant (May 23+24, 2006)	106	N/A	0/106	4.20	0.46	20
		Resident living more than 11km from the plant (May 30+31, 2006)	112	N/A	0/112	4.38	0.50	20
Study8	Coking processes	Workers at adjacement workplaces of coken ovens	118	118/0	0/118	12.8	1.23	21
		Workers at bottom of coken ovens	169	169/0	0/169	16.9	1.41	21
		Workers at side of coken ovens	82	82/0	0/82	25.1	1.15	21
		Workers at top of coken ovens	20	20/0	0/20	45.6	1.32	21
Study9	Creosote impregnation plant	Residence living at 50-360 m downwind from the plant(morning+evening)	57	N/A	0/57	3.15	1.08	22
		Residence living at 0.75-1.1 km upwind from the plant(morning+evening)	51	N/A	0/51	5.23	1.28	22
Study10	Traffic emission	Schoolchildren near a road	64	38/26	0/64	6.81	0.41	23
Study11	Brick kiln	Brick kiln workers	46	46/0	0/46	6.60	2.39	24
		Non-exposed group	34	34/0	0/46	1.66	0.97	24
Study12	Jet fuel	Workers with low exposure(pre+post shift)	172	N/A	0/172	2.95	0.41	25
		Workers with high exposure(pre+post shift)	141	N/A	0/141	4.98	0.57	25

^{a.} not available

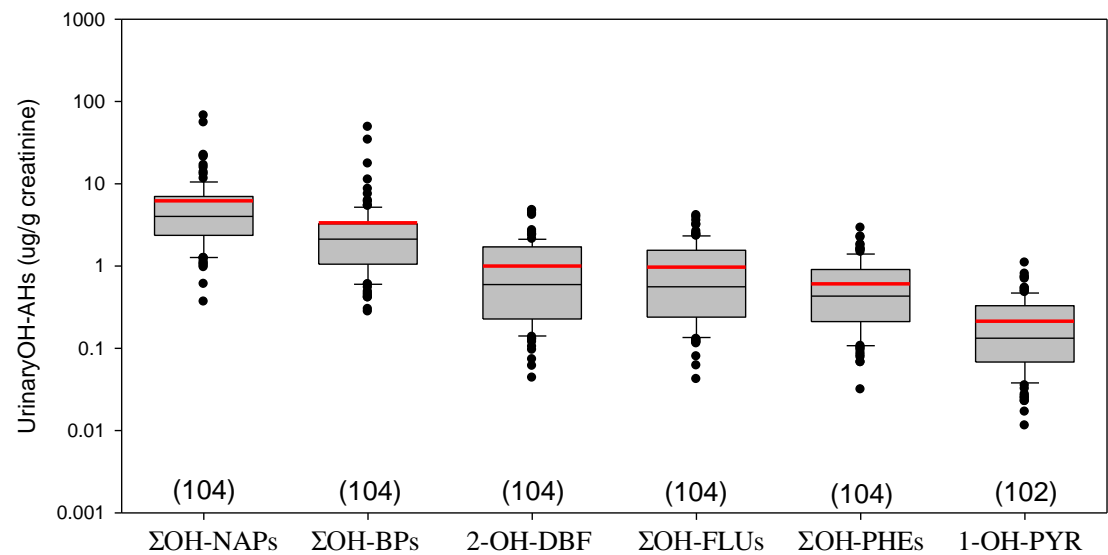


Figure S1. Concentration of grouped urinary OH-PAHs. The *solid horizontal line* represents the median, and the *red horizontal line* represents the mean. The *box* represents the 25th–75th percentiles, and the *whiskers* represent the 10th–90th percentiles. *Numbers in parentheses* are the number of detections.

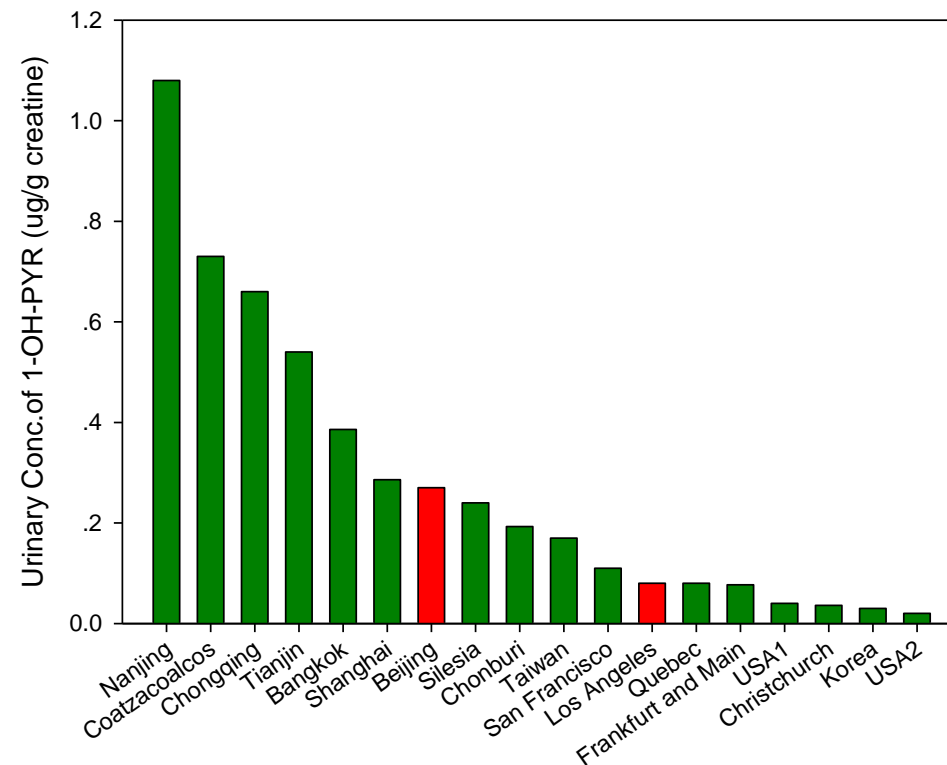


Figure S2. Concentration of urinary 1-OH-PYR in the population around world. Red bars are results from this study. Nanjing: 273 male adults;¹Coatzacoalcos: 82 school children;²Chongqing: 343 male adult;³Tianjin: 116 adults in rural sites;⁴ Bangkok: 105 schoolchildren;⁵Shanghai: 343 female adults;⁶Silesia: 108 non-smokers;⁷Chonburi: 61 schoolchildren;⁵Taiwan: 93 indoor officers;⁸ San Francisco: 76 non-smokers;⁷Quebec:140 adults;⁹Frankfurt and Main: 289 non-smokers with age>20;¹⁰ USA1: 1309 people with age>20;¹¹ Korea: 129 male university students;¹² Christchurch: 89 male students;¹³ USA2: 1625 people with age>20.¹⁴

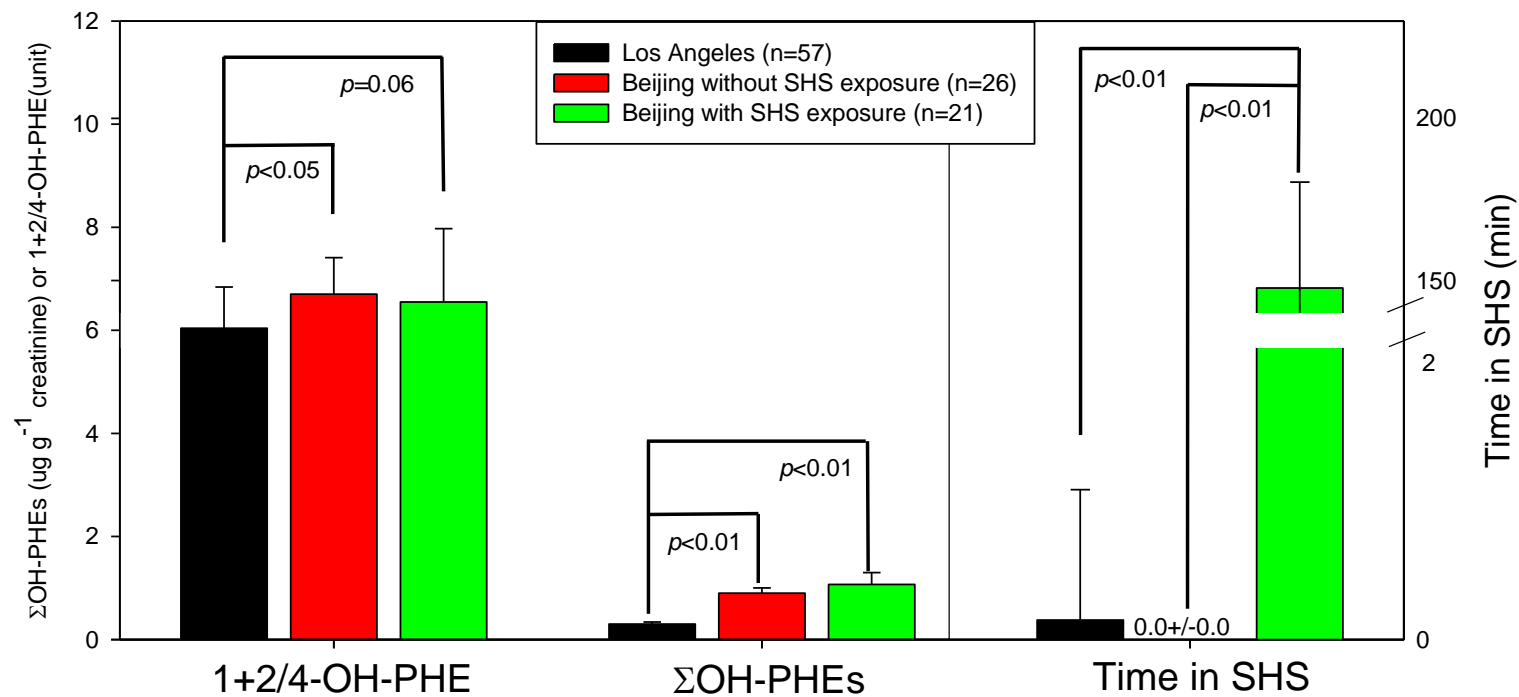


Figure S3. Comparison in 1+2/4-PHEs, $\Sigma\text{OH-PHEs}$ and time in secondhand smokes (SHS) among population in Los Angeles, in Beijing without SHS exposure and in Beijing with SHS. Error bar indicates the standard error. p -value is calculated by Mann-Whitney U-test.

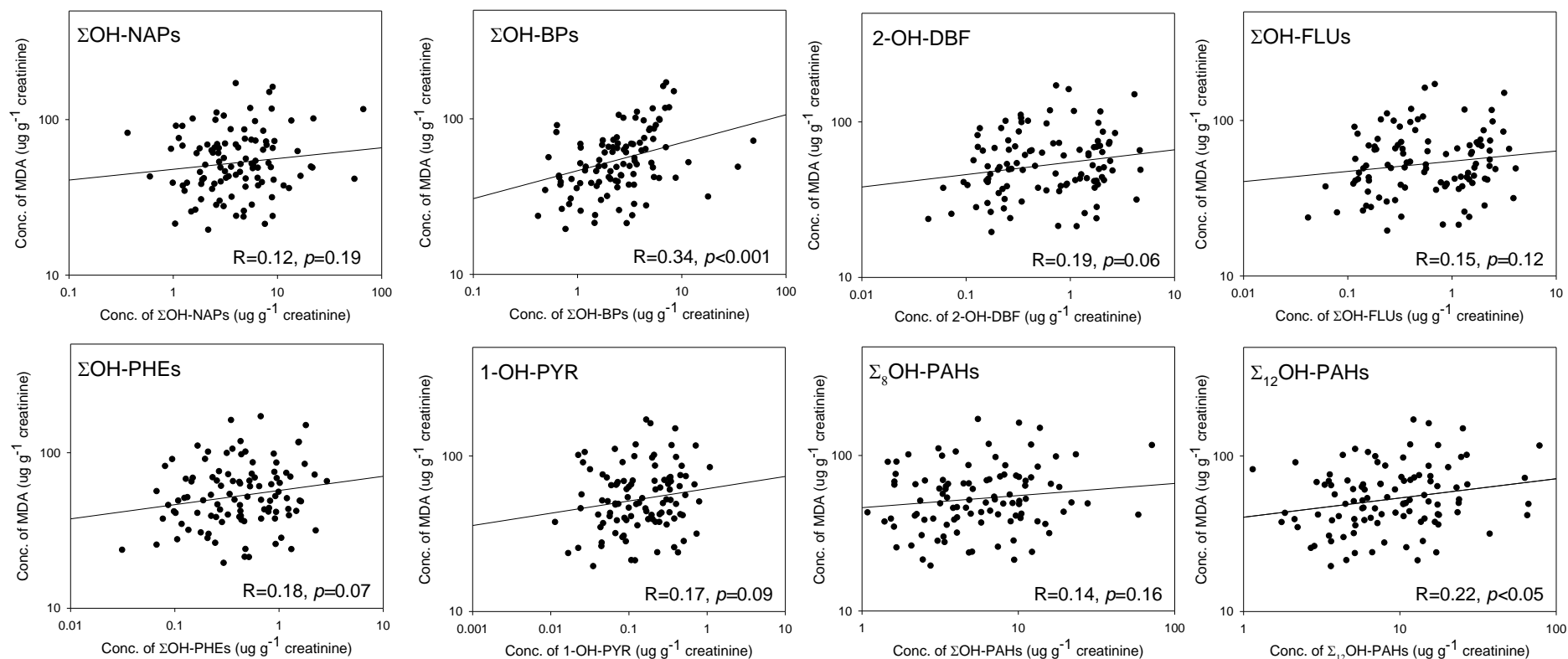


Figure S4. Correlation between MDA and OH-PAHs in both cities. $\Sigma_8\text{OH-PAHs}$ sum of $\Sigma\text{OH-NAPs}$, $\Sigma\text{OH-FLUs}$, $\Sigma\text{OH-PHEs}$, 1-OH-PYR; $\Sigma_{12}\text{OH-PAHs}$ sum of $\Sigma_8\text{OH-PAHs}$, $\Sigma\text{OH-BPs}$ and 2-OH-DBF.

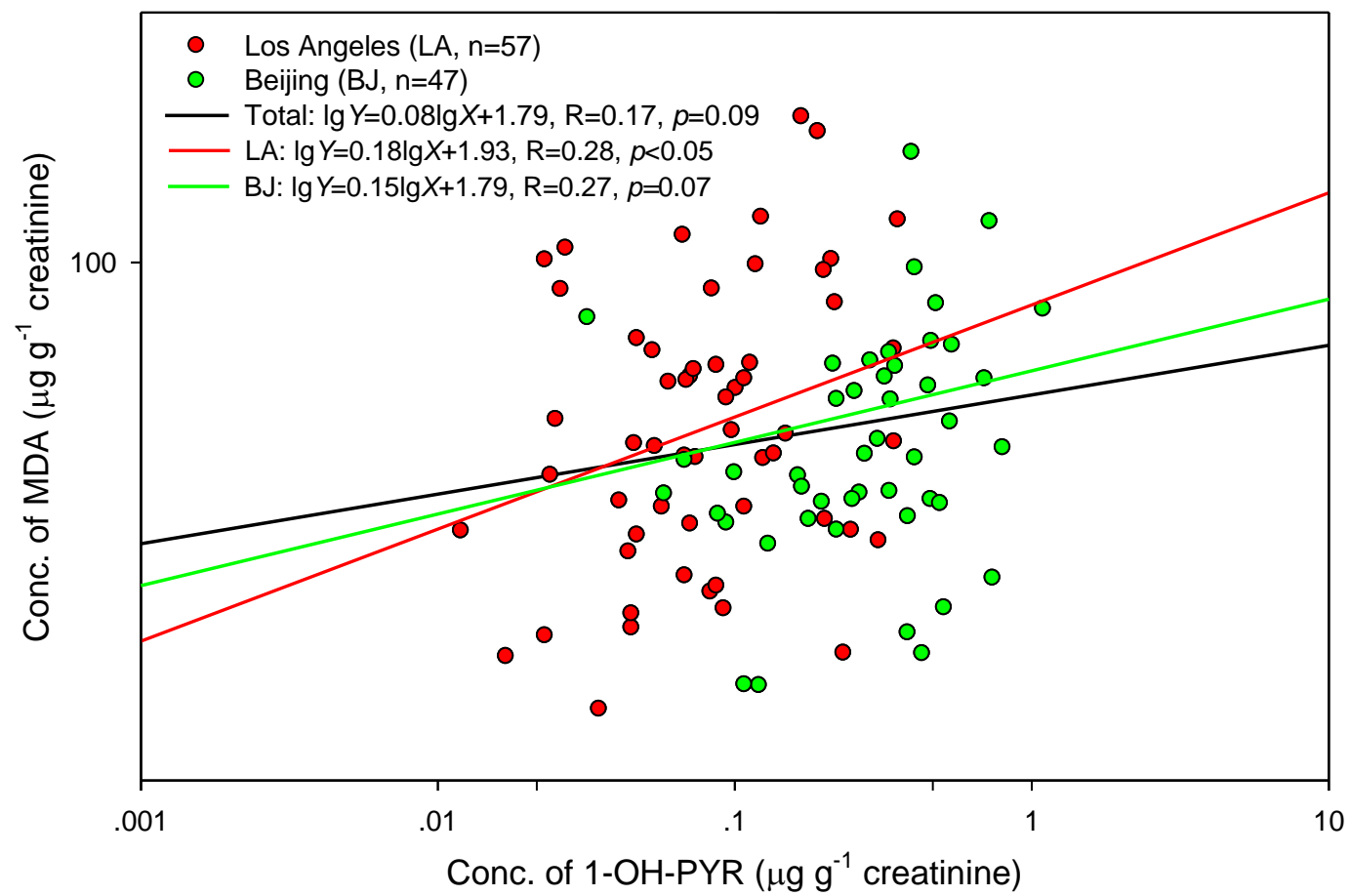


Figure S5. Correlation between MDA and 1-OH-PYR in Los Angeles and Beijing

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