

Supplemental Information of

**Spatiotemporal Variability in the Oxidative Potential of Ambient Fine
Particulate Matter in Midwestern United States**

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Content	
Number of pages	12
Number of tables	5

Table S1. Dates of samples collection at five sampling sites.

Season	Week count	Sampling period	CHI	STL	IND	CMP	BON
Summer 2018	1	2018/5/22 – 2018/5/25	✓	✓	✓	✓	✘
	2	2018/5/29 – 2018/6/1	✓	✓	✓	✓	✘
	3	2018/6/5 – 2018/6/8	✓	✓	✓	✓	✓
	4	2018/6/12 – 2018/6/15	✓	✓	✓	✓	✓
	5	2018/6/19 – 2018/6/22	✓	✓	✓	✓	✘
	6	2018/6/26 – 2018/6/29	✓	✓	✓	✓	✓
	7	2018/7/3 – 2018/7/6	✓	✓	✓	✓	✓
	8	2018/7/10 – 2018/7/13	✓	✓	✓	✓	✘
	9	2018/7/17 – 2018/7/20	✓	✓	✓	✓	✘
	10	2018/7/24 – 2018/7/27	✘	✓	✓	✓	✓
	11	2018/7/31 – 2018/8/3	✓	✓	✓	✓	✓
	12	2018/8/7 – 2018/8/10	✓	✓	✓	✓	✓
	13	2018/8/14 – 2018/8/17	✓	✓	✓	✓	✓
	14	2018/8/21 – 2018/8/24	✓	✓	✓	✓	✓
	15	2018/8/28 – 2018/8/31	✓	✓	✓	✓	✓
Fall 2018	16	2018/9/4 – 2018/9/7	✓	✓	✓	✓	✓
	17	2018/9/11 – 2018/9/14	✓	✓	✓	✓	✓
	18	2018/9/18 – 2018/9/21	✓	✓	✓	✓	✓
	19	2018/9/25 – 2018/9/28	✘	✓	✓	✓	✘
	20	2018/10/2 – 2018/10/5	✘	✓	✓	✓	✘
	21	2018/10/9 – 2018/10/12	✓	✘	✓	✓	✓
	22	2018/10/16 – 2018/10/19	✓	✓	✓	✓	✓
	23	2018/10/23 – 2018/10/26	✓	✓	✓	✓	✓
	24	2018/10/30 – 2018/11/2	✓	✓	✓	✘	✓
	25	2018/11/6 – 2018/11/9	✓	✘	✓	✓	✓
	26	2018/11/13 – 2018/11/16	✓	✘	✓	✓	✓
	27	2018/11/20 – 2018/11/23	✓	✓	✓	✘	✓
	28	2018/11/27 – 2018/11/30	✓	✓	✓	✓	✓
Winter 2018	29	2018/12/4 – 2018/12/7	✓	✓	✓	✓	✓
	30	2018/12/11 – 2018/12/14	✘	✓	✓	✓	✓
	31	2018/12/18 – 2018/12/21	✘	✓	✓	✓	✓
	32	2018/12/25 – 2018/12/28	✘	✓	✓	✓	✓
	33	2019/1/1 – 2019/1/4	✘	✓	✓	✓	✓
	34	2019/1/8 – 2019/1/11	✘	✓	✓	✓	✓
	35	2019/1/15 – 2019/1/18	✘	✓	✓	✓	✘
	36	2019/1/22 – 2019/1/25	✓	✓	✓	✓	✘
	37	2019/1/29 – 2019/2/1	✓	✓	✓	✓	✓
	38	2019/2/5 – 2019/2/8	✓	✓	✓	✓	✓
	39	2019/2/12 – 2019/2/15	✓	✓	✓	✓	✓
	40	2019/2/19 – 2019/2/22	✓	✓	✓	✓	✓
	41	2019/2/26 – 2019/3/1	✓	✓	✓	✓	✓
Spring 2019	42	2019/3/5 – 2019/3/8	✓	✓	✓	✓	✓
	43	2019/3/12 – 2019/3/15	✘	✓	✓	✓	✓
	44	2019/3/19 – 2019/3/22	✓	✓	✓	✓	✓
	45	2019/3/26 – 2019/3/29	✓	✓	✓	✓	✓
	46	2019/4/2 – 2019/4/5	✓	✓	✓	✓	✓
	47	2019/4/9 – 2019/4/12	✓	✓	✓	✓	✓
	48	2019/4/16 – 2019/4/19	✓	✓	✓	✘	✓
	49	2019/4/23 – 2019/4/26	✓	✓	✓	✓	✓
	50	2019/4/30 – 2019/5/3	✓	✘	✓	✓	✓
	51	2019/5/7 – 2019/5/10	✓	✓	✓	✓	✓
	52	2019/5/14 – 2019/5/17	✓	✘	✓	✓	✓
	53	2019/5/21 – 2019/5/24	✓	✘	✓	✓	✓
	54	2019/5/28 – 2019/5/31	✓	✘	✓	✓	✓

The symbol ✓ denotes the collection of a sample, and the symbol ✘ denotes no collection of the sample in that week (due to several reasons such as unfavorable weather conditions, broken sampler, etc.).

Table S2. Comparison of ambient PM_{2.5} OP measured in our current study with those reported in the literatures. Asterisk - * indicates that the reported results are methanol-soluble OP, while all the other results (without the asterisk) are water-soluble OP.

(a) OP^{AA}

Reference	PM size fraction (μm)	Levels	Location	Location type	Sample size
Fang et al. (2016) ^a	≤ 2.5	0.2 - 5.2 nmol·min ⁻¹ ·m ⁻³	Southeast US	Urban and rural	483
Mudway et al. (2005) ^b	≤ 2.5	0.012 ± 0.0001 nmol·min ⁻¹ ·μg ⁻¹	Eksaal, India	Biomass burning	3
Künzli et al. (2006) ^b	≤ 2.5	0.0096 ± 0.0025 nmol·min ⁻¹ ·μg ⁻¹	19 European cities	Urban	716
Szigeti et al. (2016) ^{b,c}	≤ 2.5	0.0017 – 0.04 nmol·min ⁻¹ ·μg ⁻¹	8 European cities	Urban	22
Godri et al. (2011) ^b	1.0 – 1.9	0.0058 ± 0.0025 nmol·min ⁻¹ ·μg ⁻¹	London, United Kingdom	Urban	14
Yu et al. (2020)	≤ 2.5	0.004 – 0.077 nmol·min ⁻¹ ·μg ⁻¹ median: 0.012 nmol·min ⁻¹ ·μg ⁻¹ 0.044 – 0.745 nmol·min ⁻¹ ·m ⁻³ median: 0.160 nmol·min ⁻¹ ·m ⁻³	Midwest US (5 sites)	Urban (4), rural (1)	54
This study	≤ 2.5	0.002 – 0.077 nmol·min ⁻¹ ·μg ⁻¹ median: 0.007 nmol·min ⁻¹ ·μg ⁻¹ 0.012 – 0.908 nmol·min ⁻¹ ·m ⁻³ median: 0.078 nmol·min ⁻¹ ·m ⁻³	Midwest US (5 sites)	Urban (4), rural (1)	189

^a The study assessed OP^{AA} of ambient PM samples in an AA-only model (no other antioxidants involved).

^b The composition of lung lining fluid (200 μM AA, 200 μM GSH and 200 μM UA) was different in these studies than the SLF used in our study. Moreover, total consumption of AA in 4 hours was reported, and we have estimated the rates assuming linear pattern of AA consumption with time.

^c The author compared the OP activities between indoor air PM and outdoor air PM. Only the results of outdoor air PM were included in this table.

(b) OP^{GSH}

Reference	PM size fraction (μm)	Levels	Location	Location type	Sample size
Mudway et al. (2005) ^a	≤ 2.5	$0.0083 \pm 0.0002 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	Eksaal, India	Biomass burning	3
Künzli et al. (2006) ^a	≤ 2.5	$0.0041 \pm 0.0017 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	19 European cities	Urban	716
Szigeti et al. (2016) ^{a,b}	≤ 2.5	$0 - 0.0275 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	8 European cities	Urban	22
Godri et al. (2011) ^a	1.0 – 1.9	$0.0042 \pm 0.0033 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	London, United Kingdom	Urban	14
Yu et al. (2020)	≤ 2.5	$0.001 - 0.040 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: $0.010 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ $0.008 - 0.463 \text{ nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: $0.100 \text{ nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54
This study	≤ 2.5	$0.002 - 0.035 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: $0.007 \text{ nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ $0.013 - 0.419 \text{ nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: $0.074 \text{ nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	208

^a The composition of lung lining fluid (200 μM AA, 200 μM GSH and 200 μM UA) was different in these studies than the SLF used in our study. Moreover, total consumption of GSH in 4 hours was reported, and we have estimated the rates assuming linear pattern of GSH consumption with time.

^b The author compared the OP activities between indoor air PM and outdoor air PM. Only the results of outdoor air PM were included in this table.

(c) OP^{OH-SLF}

Reference	PM size fraction (μm)	Levels	Location	Location type	Sample size
Vidrio et al. (2009) ^a	≤ 2.5	$0.253 \pm 0.135 \text{ pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	Davis, CA	Urban	~90
Ma et al. (2015) ^a	≤ 2.5	$0.092 \pm 0.019 \text{ pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	Guangzhou, China	Urban	72
Yu et al. (2020)	≤ 2.5	$0.085 - 0.967 \text{ pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: $0.307 \text{ pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ $0.857 - 7.884 \text{ pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: $3.559 \text{ pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54
This study	≤ 2.5	$0.040 - 1.217 \text{ pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: $0.142 \text{ pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ $0.269 - 12.13 \text{ pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: $1.449 \text{ pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	194

^a The SLF used in these studies had the same composition as ours (200 μM AA, 100 μM GSH, 100 μM UA and 300 μM CA). However, total $\bullet\text{OH}$ generated in 24 hours was reported, and we have estimated the rates assuming linear pattern of $\bullet\text{OH}$ generation with time.

(d) OP^{DTT}

Reference	PM size fraction (μm)	Levels	Location	Location type	Sample size
Fang et al. (2014)	≤ 2.5	0.010 – 0.097 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.024 – 0.041 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.05 – 0.81 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.23 – 0.31 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Southeast US	Urban and rural	503
Xiong et al. (2017)	≤ 2.5	0.1 – 0.18 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Urbana, IL	Urban	10
Cho et al. (2005)	≤ 2.5	0.013 – 0.047 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.029 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	Los Angeles basin, CA	Urban	11
Charrier and Anastasio (2012)	≤ 2.5	0.02 – 0.061 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.029 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	San Joaquin, CA	Urban, rural	6
Gao et al. (2017)	≤ 2.5	0.09 – 0.30 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.19 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Atlanta, GA (2 sites)	Urban	66
Gao et al. (2020)	≤ 2.5	0.005 – 0.070 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ average: 0.024 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.05 – 0.48 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ average: 0.22 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Atlanta, GA	Urban	349
Hu et al. (2008)	0.25 – 2.5	0.014 – 0.024 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.019 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.10 – 0.16 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.14 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Los Angeles harbor, CA	Urban	6
Yu et al. (2020)	≤ 2.5	0.004 – 0.193 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.014 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.041 – 1.282 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.146 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54
Verma et al. (2012)*	≤ 2.5	0.020 – 0.054 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.034 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$	Atlanta, GA	Urban	8
Gao et al. (2017)*	≤ 2.5	0.14 – 0.47 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.30 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Atlanta, GA (2 sites)	Urban	66

(d) (Continued)

Reference	PM size fraction (μm)	Levels	Location	Location type	Sample size
Gao et al. (2020)*	≤ 2.5	0.012 – 0.116 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ average: 0.027 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.13 – 0.58 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ average: 0.28 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Atlanta, GA	Urban	349
This study	≤ 2.5	0.004 – 0.032 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.014 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.029 – 0.561 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.150 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	218
This study*	≤ 2.5	0.004 – 0.042 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.021 $\text{nmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.031 – 0.639 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.234 $\text{nmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	235

Asterisk - * indicates that the reported results are methanol-soluble OP^{DTT} .

(e) $\text{OP}^{\text{OH-DTT}}$

Reference	PM size fraction (μm)	Levels	Location	Location type	Sample size
Xiong et al. (2017)	≤ 2.5	0.2 – 0.6 $\text{pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Urbana, IL	Urban	10
Yu et al. (2018)	≤ 2.5	0.2 – 1.1 $\text{pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Urbana, IL	Urban	10
Yu et al. (2020)	≤ 2.5	0.034 – 0.357 $\text{pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.082 $\text{pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.360 – 4.152 $\text{pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 1.054 $\text{pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54
This study	≤ 2.5	0.004 – 0.357 $\text{pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ median: 0.065 $\text{pmol}\cdot\text{min}^{-1}\cdot\mu\text{g}^{-1}$ 0.022 – 3.565 $\text{pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$ median: 0.722 $\text{pmol}\cdot\text{min}^{-1}\cdot\text{m}^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	240

Table S3. Results of 1-way ANOVA test for assessing the temporal and spatial variability of mass-normalized and volume-normalized OP endpoints for water-soluble PM_{2.5} samples.

(a) Temporal variability

Sampling Site	Endpoint	F value	Significantly different group(s)
Chicago, IL (CHI)	OP ^{AA} _m	1.12	
	OP ^{AA} _v	0.69	
	OP ^{GSH} _m	3.19*	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	OP ^{GSH} _v	0.78	
	OP ^{OH-SLF} _m	21.84**	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	OP ^{OH-SLF} _v	17.72**	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	OP ^{DTT} _m	2.67	Summer 2018, Fall 2018, Spring 2019
	OP ^{DTT} _v	1.03	
	OP ^{OH-DTT} _m	7.26**	Summer 2018, Winter 2018, Fall 2018, Spring 2019
OP ^{OH-DTT} _v	6.68**	Summer 2018, Fall 2018, Spring 2019	
St. Louis, MO (STL)	OP ^{AA} _m	1.37	
	OP ^{AA} _v	1.48	
	OP ^{GSH} _m	1.74	Spring 2019, Fall 2018
	OP ^{GSH} _v	1.40	
	OP ^{OH-SLF} _m	4.25**	Summer 2018, Winter 2018, Spring 2019
	OP ^{OH-SLF} _v	5.33**	Summer 2018, Fall 2018, Winter 2018, Spring 2019
	OP ^{DTT} _m	1.83	
	OP ^{DTT} _v	0.56	
	OP ^{OH-DTT} _m	0.12	
OP ^{OH-DTT} _v	0.17		
Indianapolis, IN (IND)	OP ^{AA} _m	2.02	Summer 2018, Fall 2018
	OP ^{AA} _v	2.11	Summer 2018, Spring 2019, Fall 2018
	OP ^{GSH} _m	0.53	
	OP ^{GSH} _v	0.49	
	OP ^{OH-SLF} _m	3.16*	Summer 2018, Winter 2018, Spring 2019
	OP ^{OH-SLF} _v	2.75*	Summer 2018, Winter 2018, Spring 2019
	OP ^{DTT} _m	1.29	
	OP ^{DTT} _v	0.33	
	OP ^{OH-DTT} _m	4.28**	Summer 2018, Winter 2018, Fall 2018, Spring 2019
OP ^{OH-DTT} _v	2.57	Summer 2018, Winter 2018, Fall 2018	
Champaign, IL (CMP)	OP ^{AA} _m	2.59	Summer 2018, Winter 2018
	OP ^{AA} _v	2.77*	Summer 2018, Winter 2018
	OP ^{GSH} _m	3.44*	Spring 2019, Summer 2018, Winter 2018
	OP ^{GSH} _v	4.92**	Spring 2019, Summer 2018, Winter 2018, Fall 2018
	OP ^{OH-SLF} _m	5.47**	Summer 2018, Fall 2018, Winter 2018
	OP ^{OH-SLF} _v	7.59**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{DTT} _m	0.70	
	OP ^{DTT} _v	1.55	
	OP ^{OH-DTT} _m	8.06**	Summer 2018, Winter 2018, Fall 2018, Spring 2019
OP ^{OH-DTT} _v	6.18**	Summer 2018, Winter 2018, Spring 2019, Fall 2018	
Bondville, IL (BON)	OP ^{AA} _m	5.26**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{AA} _v	8.17**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{GSH} _m	8.16**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{GSH} _v	13.81**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{OH-SLF} _m	16.82**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{OH-SLF} _v	17.33**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	OP ^{DTT} _m	3.15*	Summer 2018, Spring 2019
	OP ^{DTT} _v	3.37*	Summer 2018, Winter 2018, Spring 2019
	OP ^{OH-DTT} _m	2.10	Winter 2018, Fall 2018
OP ^{OH-DTT} _v	1.34		

(b) Spatial variability

Season	Endpoint	F value	Significantly different group(s)
Summer 2018	OP ^{AA} _m	8.60**	CMP, BON, CHI, STL, IND
	OP ^{AA} _v	5.28**	CMP, CHI, STL, IND
	OP ^{GSH} _m	28.41**	CMP, BON, CHI, STL, IND
	OP ^{GSH} _v	9.30**	CMP, BON, CHI, STL, IND
	OP ^{OH-SLF} _m	8.60**	CHI, CMP, BON, STL, IND
	OP ^{OH-SLF} _v	4.83**	CMP, CHI, STL, IND
	OP ^{DTT} _m	6.97**	CMP, STL, IND
	OP ^{DTT} _v	2.21	CMP, STL, IND
	OP ^{OH-DTT} _m	5.92**	CHI, IND, CMP, BON, STL
	OP ^{OH-DTT} _v	4.70**	CHI, STL, IND, CMP, BON
Fall 2018	OP ^{AA} _m	12.08**	CMP, CHI, STL, IND, BON
	OP ^{AA} _v	3.81**	CMP, STL, IND, BON
	OP ^{GSH} _m	27.05**	CMP, CHI, BON, IND, STL
	OP ^{GSH} _v	4.07**	CMP, CHI, STL, IND
	OP ^{OH-SLF} _m	1.46	CMP, IND
	OP ^{OH-SLF} _v	0.46	
	OP ^{DTT} _m	13.39**	CMP, CHI, BON, STL, IND
	OP ^{DTT} _v	0.51	
	OP ^{OH-DTT} _m	3.52*	CHI, STL, IND, BON, CMP
	OP ^{OH-DTT} _v	4.00**	CHI, STL, IND, BON, CMP
Winter 2018	OP ^{AA} _m	2.21	CMP, CHI, STL, IND, BON
	OP ^{AA} _v	1.95	CMP, STL, IND, BON
	OP ^{GSH} _m	15.75**	CMP, CHI, STL, IND, BON
	OP ^{GSH} _v	12.37**	CMP, CHI, STL, IND, BON
	OP ^{OH-SLF} _m	2.23	CMP, CHI
	OP ^{OH-SLF} _v	1.78	STL, BON
	OP ^{DTT} _m	4.33**	CMP, STL, IND
	OP ^{DTT} _v	3.23*	CHI, STL, IND, BON
	OP ^{OH-DTT} _m	2.60*	IND, BON, STL
	OP ^{OH-DTT} _v	2.49*	CHI, IND, STL, CMP
Spring 2019	OP ^{AA} _m	5.20**	CMP, CHI, STL, IND, BON
	OP ^{AA} _v	4.92**	CMP, CHI, STL, IND, BON
	OP ^{GSH} _m	14.59**	CMP, CHI, STL, IND, BON
	OP ^{GSH} _v	10.74**	CMP, CHI, STL, IND, BON
	OP ^{OH-SLF} _m	3.20*	CMP, CHI, STL, IND, BON
	OP ^{OH-SLF} _v	3.19*	CMP, CHI, STL, IND, BON
	OP ^{DTT} _m	10.78**	CMP, CHI, BON, STL
	OP ^{DTT} _v	6.04**	CMP, CHI, STL, IND, BON
	OP ^{OH-DTT} _m	2.57*	IND, BON, CMP
	OP ^{OH-DTT} _v	1.89	STL, IND, CMP

Asterisks - * and ** indicate significant ($P < 0.05$) and very significant ($P < 0.01$) differences, respectively.

Table S4. Results of 1-way ANOVA test for assessing the temporal and spatial variability of mass-normalized and volume-normalized OP endpoints for methanol-soluble PM_{2.5} samples.

(a) Temporal variability

Sampling Site	Endpoint	F value	Significantly different group(s)
Chicago, IL (CHI)	OP ^{AA} _m	1.03	
	OP ^{AA} _v	0.07	
	OP ^{GSH} _m	1.41	
	OP ^{GSH} _v	0.28	
	OP ^{OH-SLF} _m	1.68	Summer 2018, Spring 2019
	OP ^{OH-SLF} _v	0.99	
	OP ^{DTT} _m	4.27*	Summer 2018, Fall 2018, Winter 2019
	OP ^{DTT} _v	1.53	
St. Louis, MO (STL)	OP ^{OH-DTT} _m	3.84*	Summer 2018, Fall 2018, Winter 2018, Spring 2019
	OP ^{OH-DTT} _v	3.37*	Summer 2018, Fall 2018
	OP ^{AA} _m	2.16	Fall 2018, Spring 2019
	OP ^{AA} _v	3.41*	Summer 2018, Fall 2018, Spring 2019
	OP ^{GSH} _m	3.62*	Fall 2018, Summer 2018, Winter 2018, Spring 2019
	OP ^{GSH} _v	1.92	Fall 2018, Spring 2019
	OP ^{OH-SLF} _m	1.05	
	OP ^{OH-SLF} _v	1.23	
Indianapolis, IN (IND)	OP ^{DTT} _m	1.14	
	OP ^{DTT} _v	1.87	Summer 2018, Winter 2019
	OP ^{OH-DTT} _m	0.50	
	OP ^{OH-DTT} _v	1.11	
	OP ^{AA} _m	2.42	Summer 2018, Spring 2019
	OP ^{AA} _v	1.39	
	OP ^{GSH} _m	2.15*	Fall 2018, Spring 2019
	OP ^{GSH} _v	0.63	
Champaign, IL (CMP)	OP ^{OH-SLF} _m	3.49*	Fall 2018, Spring 2019, Winter 2018
	OP ^{OH-SLF} _v	2.41	Fall 2018, Winter 2018
	OP ^{DTT} _m	1.42	
	OP ^{DTT} _v	0.94	
	OP ^{OH-DTT} _m	0.20	
	OP ^{OH-DTT} _v	0.67	
	OP ^{AA} _m	1.64	Summer 2018, Winter 2018
	OP ^{AA} _v	2.95*	Summer 2018, Fall 2018, Winter 2018
Bondville, IL (BON)	OP ^{GSH} _m	1.42	
	OP ^{GSH} _v	0.03	
	OP ^{OH-SLF} _m	1.00	
	OP ^{OH-SLF} _v	1.22	
	OP ^{DTT} _m	3.73*	Summer 2018, Winter 2018
	OP ^{DTT} _v	2.93*	Summer 2018, Fall 2018, Winter 2018
	OP ^{OH-DTT} _m	0.08	
	OP ^{OH-DTT} _v	0.59	
Bondville, IL (BON)	OP ^{AA} _m	8.76**	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	OP ^{AA} _v	9.27**	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	OP ^{GSH} _m	1.51	
	OP ^{GSH} _v	1.58	Summer 2018, Winter 2018
	OP ^{OH-SLF} _m	4.30**	Summer 2018, Spring 2019, Winter 2018
	OP ^{OH-SLF} _v	4.70**	Summer 2018, Spring 2019, Winter 2018
	OP ^{DTT} _m	2.95*	Summer 2018, Spring 2019, Winter 2018
	OP ^{DTT} _v	4.28**	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	OP ^{OH-DTT} _m	2.24	
	OP ^{OH-DTT} _v	1.64	

(b) Spatial variability

Season	Endpoint	F value	Significantly different group(s)
Summer 2018	OP ^{AA} _m	1.17	BON, STL
	OP ^{AA} _v	0.13	
	OP ^{GSH} _m	2.00	CMP, STL, IND
	OP ^{GSH} _v	0.40	
	OP ^{OH-SLF} _m	2.80*	CHI, CMP, IND, STL
	OP ^{OH-SLF} _v	1.67	CHI, CMP, IND
	OP ^{DTT} _m	0.74	
	OP ^{DTT} _v	0.46	
	OP ^{OH-DTT} _m	3.75**	CHI, STL, CMP
	OP ^{OH-DTT} _v	3.11*	CHI, IND, STL, CMP
Fall 2018	OP ^{AA} _m	0.62	
	OP ^{AA} _v	2.40	STL, CMP, BON
	OP ^{GSH} _m	2.55*	CMP, STL, BON, IND
	OP ^{GSH} _v	1.05	
	OP ^{OH-SLF} _m	0.81	
	OP ^{OH-SLF} _v	0.97	
	OP ^{DTT} _m	0.33	
	OP ^{DTT} _v	2.50*	STL, CMP, BON
	OP ^{OH-DTT} _m	1.99	IND, STL, CMP
	OP ^{OH-DTT} _v	2.28	IND, CMP, BON
Winter 2018	OP ^{AA} _m	1.06	
	OP ^{AA} _v	3.62**	CHI, STL, IND, BON
	OP ^{GSH} _m	6.31**	CMP, CHI, BON, STL, IND
	OP ^{GSH} _v	2.86*	CHI, CMP, IND, BON
	OP ^{OH-SLF} _m	1.79	CHI, BON, STL
	OP ^{OH-SLF} _v	3.21*	CHI, IND, CMP, STL, BON
	OP ^{DTT} _m	0.86	
	OP ^{DTT} _v	2.45*	CHI, STL, CMP, BON
	OP ^{OH-DTT} _m	2.21	IND, CMP, BON, STL
	OP ^{OH-DTT} _v	2.67*	CHI, IND, CMP, BON
Spring 2019	OP ^{AA} _m	1.60	
	OP ^{AA} _v	2.46*	CHI, CMP, BON
	OP ^{GSH} _m	7.44**	CMP, CHI, IND, STL
	OP ^{GSH} _v	4.33**	CMP, CHI, BON, IND, STL
	OP ^{OH-SLF} _m	0.46	
	OP ^{OH-SLF} _v	0.60	
	OP ^{DTT} _m	0.79	
	OP ^{DTT} _v	1.93	CHI, BON
	OP ^{OH-DTT} _m	2.15	BON, IND, CMP
	OP ^{OH-DTT} _v	1.63	IND, CMP

Asterisks - * and ** indicate significant ($P < 0.05$) and very significant ($P < 0.01$) differences, respectively.

Table S5. Seasonal median of the ratio of methanol-soluble OPv to water-soluble OPv (M/W^{OP}) for OP^{OH-SLF}v at five sampling sites.

	CHI	STL	IND	CMP	BON
Summer 2018	2.1	2.6	2.0	1.1	2.0
Fall 2018	3.5	4.9	5.5	2.7	4.6
Winter 2018	9.4	2.9	3.3	3.2	3.9
Spring 2019	3.2	2.7	7.2	4.1	3.9

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