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Supplement of

Spatiotemporal variability in the oxidative potential of ambient fine particulate matter in the Midwestern United States

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Section S1. Comparison of five Hi-Vol samplers before and after the sampling campaign

Out of five samplers used in our study, two were old samplers (about 5 years old, used in various sampling campaigns) and three were brand new, which were bought from TISCH Environmental (Cleves, OH, US) a month before the sampling. These new samplers were factory calibrated and installed at three farther sites, i.e. Chicago (CHI), Indianapolis (IND) and St. Louis (STL). The other two old samplers were installed at Champaign (CMP) and Bondville (BON). For the sole purpose of this discussion, we will name them as CHI (N), IND (N), STL (N), CMP (O) and BON (O). Since the new samplers were factory calibrated, we had more confidence in them, therefore, we chose one of those samplers, i.e. CHI (N), as a reference and compared the responses of other two old samplers, i.e. CMP (O) and BON (O), by running them in pairs, i.e. first CHI (N) and CMP (O) pair, followed by CHI (N) and BON (O) pair, at a site in Urbana in April 2018 (due to some practical constraint, we couldn't run all three of them together). We collected 9 sets of 24-hours integrated Hi-Vol $PM_{2.5}$ samples on quartz filters from each pair, and analyzed them for the DTT assay using the same extraction and analysis procedure as used in our current study. The comparison of OP^{DTT} response was conducted by the orthogonal fit regression analysis of $OP^{DTT}v$ of $PM_{2.5}$ samples collected from CHI (N) and old samplers (**Figure S1**). The correlations between the old samplers and CHI (N) sampler were excellent ($R^2 = 0.92 – 0.94$) with slopes almost equal to 1 (1.02 – 1.03), indicating that the samplers collect identical $PM_{2.5}$, and had negligible internal difference in sample collection.

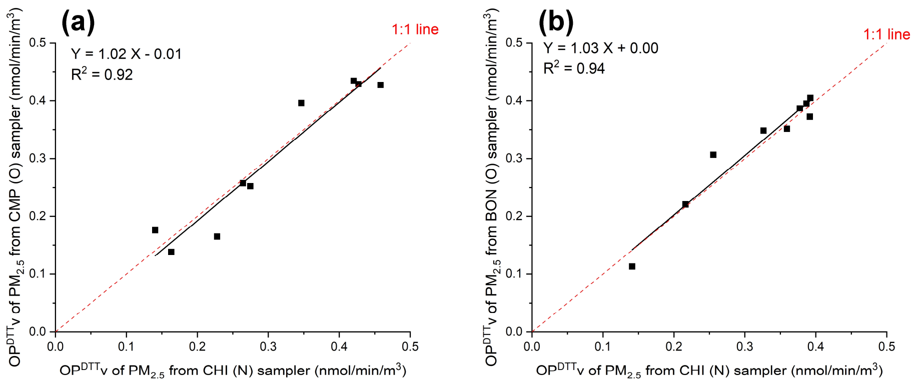

Figure S1. Comparison of OP^{DTT} of $PM_{2.5}$ samples collected from CHI (N) sampler with old samplers: (a) CMP (O) sampler; (b) BON (O) sampler.

After the sampling campaign, we again moved the new samplers [i.e. CHI (N), STL (N) and IND (N)] back to CMP site, kept them side-by-side, and collected 9 Hi-Vol samples (24-hours integrated) from each sampler. All these samples were extracted in DI and analyzed for OP^{DTT} in the same manner as used in our current study. The comparison of the reference sampler [i.e. CHI (N)] with other two new samplers was also conducted by orthogonal fit (Figure S2). Excellent correlations ($R^2 = 0.93 – 0.95$) and consistent slopes (1.05 – 1.06, close to 1) both showed a good consistency of three new samplers.

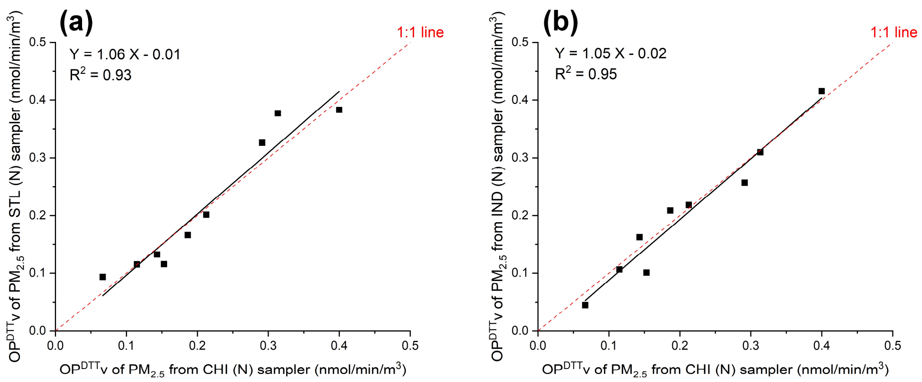

Figure S2. Comparison of OP^{DTT} of $PM_{2.5}$ samples collected from CHI (N) sampler with other new samplers: (a) STL (N) sampler; (b) IND (N) sampler.

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. Precision of SAMERA for methanol-soluble OP measurements compared with water-soluble OP measurements.

Endpoint	Unit	Average	Standard Deviation	CoV (%)	CoV (%) for the water-soluble $PM_{2.5}$ extract (Yu et al., 2020)
OP^{AA}	nmol/min/m^3	0.132	0.018	13.51	11.87
OP^{GSH}	nmol/min/m^3	0.098	0.010	10.65	7.89
$OP^{OH\text{-}SLF}$	pmol/min/m^3	0.740	0.011	14.49	10.56
OP^{DTT}	nmol/min/m^3	0.187	0.017	8.89	10.52
$OP^{OH\text{-}DTT}$	pmol/min/m^3	0.216	0.023	10.88	13.28

Table S3. Results of 1-way ANOVA test for assessing the temporal and spatial variability of $PM_{2.5}$ mass concentrations.

Variability	Sampling Site/Season	F value	Significantly different group(s)
Temporal	CHI	1.95	
	STL	1.79	
	IND	0.33	
	CMP	3.25*	Fall 2018
	BON	0.82	
Spatial	Summer 2018	3.48*	STL
	Fall 2018	3.13*	CHI, STL, IND, CMP
	Winter 2018	5.01**	CHI
	Spring 2019	3.35*	BON

Asterisks – * and ** indicate significant ($P < 0.05$) and highly 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 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	
	$OP^{OH\text{-}SLF}m$	21.84**	Summer 2018, Fall 2018, Spring 2019, Winter 2018
	$OP^{OH\text{-}SLF}v$	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	
	$OP^{OH\text{-}DTT}m$	7.26**	Summer 2018, Winter 2018, Fall 2018, Spring 2019
	$OP^{OH\text{-}DTT}v$	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	
	$OP^{OH\text{-}SLF}m$	4.25**	Summer 2018, Winter 2018, Spring 2019
	$OP^{OH\text{-}SLF}v$	5.33**	Summer 2018, Fall 2018, Winter 2018, Spring 2019
	$OP^{DTT}m$	1.83	
	$OP^{DTT}v$	0.56	
	$OP^{OH\text{-}DTT}m$	0.12	
	$OP^{OH\text{-}DTT}v$	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	
	$OP^{OH\text{-}SLF}m$	3.16*	Summer 2018, Winter 2018, Spring 2019
	$OP^{OH\text{-}SLF}v$	2.75*	Summer 2018, Winter 2018, Spring 2019
	$OP^{DTT}m$	1.29	
	$OP^{DTT}v$	0.33	
	$OP^{OH\text{-}DTT}m$	4.28**	Summer 2018, Winter 2018, Fall 2018, Spring 2019
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	5.47**	Summer 2018, Fall 2018, Winter 2018
	$OP^{OH\text{-}SLF}v$	7.59**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	$OP^{DTT}m$	0.70	
	$OP^{DTT}v$	1.55	
	$OP^{OH\text{-}DTT}m$	8.06**	Summer 2018, Winter 2018, Fall 2018, Spring 2019
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	16.82**	Summer 2018, Spring 2019, Fall 2018, Winter 2018
	$OP^{OH\text{-}SLF}v$	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
	$OP^{OH\text{-}DTT}m$	2.10	Winter 2018, Fall 2018
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	8.60**	CHI, CMP, BON, STL, IND
	$OP^{OH\text{-}SLF}v$	4.83**	CMP, CHI, STL, IND
	$OP^{DTT}m$	6.97**	CMP, STL, IND
	$OP^{DTT}v$	2.21	CMP, STL, IND
	$OP^{OH\text{-}DTT}m$	5.92**	CHI, IND, CMP, BON, STL
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	1.46	CMP, IND
	$OP^{OH\text{-}SLF}v$	0.46	
	$OP^{DTT}m$	13.39**	CMP, CHI, BON, STL, IND
	$OP^{DTT}v$	0.51	
	$OP^{OH\text{-}DTT}m$	3.52*	CHI, STL, IND, BON, CMP
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	2.23	CMP, CHI
	$OP^{OH\text{-}SLF}v$	1.78	STL, BON
	$OP^{DTT}m$	4.33**	CMP, STL, IND
	$OP^{DTT}v$	3.23*	CHI, STL, IND, BON
	$OP^{OH\text{-}DTT}m$	2.60*	IND, BON, STL
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	3.20*	CMP, CHI, STL, IND, BON
	$OP^{OH\text{-}SLF}v$	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
	$OP^{OH\text{-}DTT}m$	2.57*	IND, BON, CMP
	$OP^{OH\text{-}DTT}v$	1.89	STL, IND, CMP

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

Table S5. 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	
	$OP^{OH\text{-}SLF}m$	1.68	Summer 2018, Spring 2019
	$OP^{OH\text{-}SLF}v$	0.99	
	$OP^{DTT}m$	4.27*	Summer 2018, Fall 2018, Winter 2019
	$OP^{DTT}v$	1.53	
	$OP^{OH\text{-}DTT}m$	3.84*	Summer 2018, Fall 2018, Winter 2018, Spring 2019
	$OP^{OH\text{-}DTT}v$	3.37*	Summer 2018, Fall 2018
St. Louis, MO (STL)	$OP^{AA}m$	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
	$OP^{OH\text{-}SLF}m$	1.05	
	$OP^{OH\text{-}SLF}v$	1.23	
	$OP^{DTT}m$	1.14	
	$OP^{DTT}v$	1.87	Summer 2018, Winter 2019
	$OP^{OH\text{-}DTT}m$	0.50	
	$OP^{OH\text{-}DTT}v$	1.11	
Indianapolis, IN (IND)	$OP^{AA}m$	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	
	$OP^{OH\text{-}SLF}m$	3.49*	Fall 2018, Spring 2019, Winter 2018
	$OP^{OH\text{-}SLF}v$	2.41	Fall 2018, Winter 2018
	$OP^{DTT}m$	1.42	
	$OP^{DTT}v$	0.94	
	$OP^{OH\text{-}DTT}m$	0.20	
	$OP^{OH\text{-}DTT}v$	0.67	
Champaign, IL (CMP)	$OP^{AA}m$	1.64	Summer 2018, Winter 2018
	$OP^{AA}v$	2.95*	Summer 2018, Fall 2018, Winter 2018
	$OP^{GSH}m$	1.42	
	$OP^{GSH}v$	0.03	
	$OP^{OH\text{-}SLF}m$	1.00	
	$OP^{OH\text{-}SLF}v$	1.22	
	$OP^{DTT}m$	3.73*	Summer 2018, Winter 2018
	$OP^{DTT}v$	2.93*	Summer 2018, Fall 2018, Winter 2018
	$OP^{OH\text{-}DTT}m$	0.08	
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	4.30**	Summer 2018, Spring 2019, Winter 2018
	$OP^{OH\text{-}SLF}v$	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
	$OP^{OH\text{-}DTT}m$	2.24	
	$OP^{OH\text{-}DTT}v$	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	
	$OP^{OH\text{-}SLF}m$	2.80*	CHI, CMP, IND, STL
	$OP^{OH\text{-}SLF}v$	1.67	CHI, CMP, IND
	$OP^{DTT}m$	0.74	
	$OP^{DTT}v$	0.46	
	$OP^{OH\text{-}DTT}m$	3.75**	CHI, STL, CMP
	$OP^{OH\text{-}DTT}v$	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	
	$OP^{OH\text{-}SLF}m$	0.81	
	$OP^{OH\text{-}SLF}v$	0.97	
	$OP^{DTT}m$	0.33	
	$OP^{DTT}v$	2.50*	STL, CMP, BON
	$OP^{OH\text{-}DTT}m$	1.99	IND, STL, CMP
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	1.79	CHI, BON, STL
	$OP^{OH\text{-}SLF}v$	3.21*	CHI, IND, CMP, STL, BON
	$OP^{DTT}m$	0.86	
	$OP^{DTT}v$	2.45*	CHI, STL, CMP, BON
	$OP^{OH\text{-}DTT}m$	2.21	IND, CMP, BON, STL
	$OP^{OH\text{-}DTT}v$	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
	$OP^{OH\text{-}SLF}m$	0.46	
	$OP^{OH\text{-}SLF}v$	0.60	
	$OP^{DTT}m$	0.79	
	$OP^{DTT}v$	1.93	CHI, BON
	$OP^{OH\text{-}DTT}m$	2.15	BON, IND, CMP
	$OP^{OH\text{-}DTT}v$	1.63	IND, CMP

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

Table S6. 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 (μm)	Levels	Location	Location type	Sample size	Methodology
Fang et al. (2016)	≤ 2.5	0.2 - 5.2 $nmol \cdot min^{-1} \cdot m^{-3}$	Southeast US	Urban and rural	483	Ambient $PM_{2.5}$ samples were collected using a Hi-Vol sampler on quartz filters, extracted in DI and filtered through a syringe filter. OP^{AA} of filtered extracts was assessed with an AA-only assay (no other antioxidants involved; concentration of AA was 200 μM) with an automated system. AA was measured based on a photometric method (at 265 nm).
Mudway et al. (2005)	≤ 2.5	0.012 ± 0.0001 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	Eksaal, India	Biomass burning	3	Biomass burning samples were collected from dung-cake combustion, and extracted in Chelex-treated DI with 5% methanol. OP^{AA} of filtered extracts was assessed in a respiratory tract lining fluid (RTLF; composition was 200 μM AA, 200 μM GSH and 200 μM UA). AA was measured based on a photometric method (at 265 nm).
Künzli et al. (2006)	≤ 2.5	0.0096 ± 0.0025 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	19 European cities	Urban	716	Ambient $PM_{2.5}$ samples were collected using a Basel-Sampler, and extracted in metal-free DI. OP^{AA} was assessed in the same manner as Mudway et al. (2005).
Szigeti et al. (2016)	≤ 2.5	0.0017 – 0.04 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	8 European cities	Urban	22	Ambient and indoor $PM_{2.5}$ samples were collected using a Low-Vol sampler, and directly incubated in RTLF having same composition as in Mudway et al. (2005). AA was measured based on a photometric method (at 265 nm).
Godri et al. (2011)	1.0 – 1.9	0.0058 ± 0.0025 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	London, United Kingdom	Urban	14	Ambient size-segregated samples were collected using a MOUDI sampler, and extracted in Chelex-treated DI with 5% methanol. OP^{AA} was assessed in the same manner as Mudway et al. (2005).

Perrone et al. (2019)	≤ 2.5	0.006 ± 0.001 nmol·min^{-1}·µg^{-1} 0.136 ± 0.020 nmol·min^{-1}·m^{-3}	Lecce, Italy	Urban	39	Ambient $PM_{2.5}$ samples were collected using a low volume HYDRA-FAI dual sampler, and extracted in DI. OP^{AA} of filtered extracts was assessed with an AA-only assay similar as in Fang et al. (2016).
Gao et al. (2020a)	≤ 2.5	0.023 – 0.126 nmol·min^{-1}·m^{-3}	Atlanta, GA	Urban	349	Ambient $PM_{2.5}$ samples were collected using a Hi-Vol sampler on quartz filters, extracted in DI and filtered through a syringe filter. OP^{AA} was assessed in the same manner as Mudway et al. (2005).
Yang et al. (2014)	≤ 2.5	0.8 – 35.0 nmol·s^{-1}·m^{-3}	Rotterdam and Amsterdam, Netherland	Urban	10	Ambient $PM_{2.5}$ samples were collected using a Harvard Impactor and extracted in ultrapure water. OP^{AA} of filtered extracts was assessed AA-only assay similar as in Fang et al. (2016).
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	$PM_{2.5}$ sampling, preparation and OP^{AA} measurement were conducted in the same manner as the current study.
Yang et al. (2014)*	≤ 2.5	2.2 – 43.5 nmol·s^{-1}·m^{-3}	Rotterdam and Amsterdam, Netherland	Urban	20	Ambient $PM_{2.5}$ samples were collected using a Harvard Impactor and extracted in methanol. Filtered methanol extracts were evaporated using an evaporator set, and reconstituted with DI. OP^{AA} of water-reconstituted methanol extracts was assessed AA-only assay similar as in Fang et al. (2016).
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)	241	See section 2 (experimental methods).
This study*		0.004 – 0.029 nmol·min^{-1}·µg^{-1} median: 0.012 nmol·min^{-1}·µg^{-1} 0.030 – 0.311 nmol·min^{-1}·m^{-3} median: 0.134 nmol·min^{-1}·m^{-3}	Midwest US (5 sites)	Urban (4), rural (1)	241	

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

(b) OP^{GSH}

Reference	**PM size (µm)**	**Levels**	**Location**	**Location type**	**Sample size**	**Methodology**
Mudway et al. (2005)	≤ 2.5	0.0083 ± 0.0002 $nmol·min^{-1}·µg^{-1}$	Eksaal, India	Biomass burning	3	OP^{GSH} of filtered extracts was measured in RTLF. GSH was measured with a glutathione disulfide (GSSG)-reductase-5,5-dithio-bis-(2-nitrobenzoic acid) (DTNB) recycling assay, based on a photometric method (at 405 nm).
Künzli et al. (2006)	≤ 2.5	0.0041 ± 0.0017 $nmol·min^{-1}·µg^{-1}$	19 European cities	Urban	716	OP^{GSH} was assessed in the same manner as Mudway et al. (2005).
Szigeti et al. (2016)	≤ 2.5	0 – 0.0275 $nmol·min^{-1}·µg^{-1}$	8 European cities	Urban	22	Punches of filter samples were directly incubated in RTLF, and measured for OP^{GSH} in the same manner with Mudway et al. (2005).
Godri et al. (2011)	1.0 – 1.9	0.0042 ± 0.0033 $nmol·min^{-1}·µg^{-1}$	London, United Kingdom	Urban	14	OP^{GSH} was assessed in the same manner as Mudway et al. (2005).
Gao et al. (2020a)	≤ 2.5	0.025 – 0.067 $nmol·min^{-1}·m^{-3}$	Atlanta, GA	Urban	349	OP^{GSH} was assessed in the same manner as Mudway et al. (2005).
Yu et al. (2020)	≤ 2.5	0.001 – 0.040 $nmol·min^{-1}·µg^{-1}$ median: 0.010 $nmol·min^{-1}·µg^{-1}$ 0.008 – 0.463 $nmol·min^{-1}·m^{-3}$ median: 0.100 $nmol·min^{-1}·m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54	$PM_{2.5}$ sampling, preparation and OP^{GSH} measurement were conducted in the same manner as the current study.
This study	≤ 2.5	0.002 – 0.035 $nmol·min^{-1}·µg^{-1}$ median: 0.007 $nmol·min^{-1}·µg^{-1}$ 0.013 – 0.419 $nmol·min^{-1}·m^{-3}$ median: 0.074 $nmol·min^{-1}·m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	241	See section 2 (experimental methods).

(c) $OP^{OH\text{-}SLF}$

Reference	PM size (μm)	Levels	Location	Location type	Sample size	Methodology
Vidrio et al. (2009)	≤ 2.5	0.253 ± 0.135 pmol·min^{-1}·μg^{-1}	Davis, CA	Urban	~90	Ambient $PM_{2.5}$ samples were collected using IMPROVE Version II samplers on Teflo filters, directly incubated in SLF (composition was 114 mM NaCl, 10 mM sodium benzoate, 10 mM total phosphate to buffer the solution at pH 7.4, 200 μM AA and 300 μM CA) with desferoxamine (DSF) for 24 hours, and measured for ·OH generation. ·OH was captured by sodium benzoate and measured based on a photometric method (at 256 nm) using a high-performance liquid chromatography (HPLC).
Ma et al. (2015)	≤ 2.5	0.092 ± 0.019 pmol·min^{-1}·μg^{-1}	Guangzhou, China	Urban	72	Ambient $PM_{2.5}$ samples were collected using a Low-Vol sampler on Teflon filters. $OP^{OH\text{-}SLF}$ was measured in the same manner as in Vidrio et al. (2009).
Yu et al. (2020)	≤ 2.5	0.085 – 0.967 pmol·min^{-1}·μg^{-1} median: 0.307 pmol·min^{-1}·μg^{-1} 0.857 – 7.884 pmol·min^{-1}·m^{-3} median: 3.559 pmol·min^{-1}·m^{-3}	Midwest US (5 sites)	Urban (4), rural (1)	54	$PM_{2.5}$ sampling, preparation and $OP^{OH\text{-}SLF}$ measurement were conducted in the same manner as the current study.
This study	≤ 2.5	0.040 – 1.217 pmol·min^{-1}·μg^{-1} median: 0.142 pmol·min^{-1}·μg^{-1} 0.269 – 12.13 pmol·min^{-1}·m^{-3} median: 1.449 pmol·min^{-1}·m^{-3}	Midwest US (5 sites)	Urban (4), rural (1)	241	See section 2 (experimental methods).

(d) OP^{DTT}

Reference	PM size (μm)	Levels	Location	Location type	Sample size	Methodology
Fang et al. (2015)	≤ 2.5	0.010 – 0.097 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.024 – 0.041 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.05 – 0.81 $nmol \cdot min^{-1} \cdot m^{-3}$ median: 0.23 – 0.31 $nmol \cdot min^{-1} \cdot m^{-3}$	Southeast US	Urban and rural	503	Ambient $PM_{2.5}$ samples were collected using a Hi-Vol sampler on quartz filters, extracted in DI and filtered through a syringe filter. Filtered extracts were then incubated in a mixture of 100 μM DTT and 0.5 mM potassium phosphate buffer (K-PB; pH = 7.4). DTT was captured by DTNB and measured based on a photometric method (at 412 nm) using an automated system.
Xiong et al. (2017)	≤ 2.5	0.1 – 0.18 $nmol \cdot min^{-1} \cdot m^{-3}$	Urbana, IL	Urban	10	Ambient $PM_{2.5}$ samples were collected with Hi-Vol sampler on quartz filters, extracted in Milli-Q water, and filtered through a syringe filter. OP^{DTT} were assessed in the same manner with Fang et al. (2015).
Cho et al. (2005)	≤ 2.5	0.013 – 0.047 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.029 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	Los Angeles basin, CA	Urban	11	Ambient size-segregated samples were collected using a VACES in conjunction with a BioSampler. Collected suspensions were then incubated in a mixture of 100 μM DTT and 0.5 mM potassium phosphate buffer (K-PB; pH = 7.4). DTT was captured by DTNB and measured based on a photometric method (at 412 nm) at designated time points within 90 min.
Charrier and Anastasio (2012)	≤ 2.5	0.02 – 0.061 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.029 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	San Joaquin, CA	Urban, rural	6	Ambient $PM_{2.5}$ samples were collected on Teflon filters, but the filter extraction method was not reported. DTT assay was conducted by incubating the aqueous sample extracts in 100 μM DTT. DTT was captured by DTNB and measured based on a photometric method (at 412 nm) at four time points within 16 min.
Gao et al. (2017)	≤ 2.5	0.09 – 0.30 $nmol \cdot min^{-1} \cdot m^{-3}$ median: 0.19 $nmol \cdot min^{-1} \cdot m^{-3}$	Atlanta, GA (2 sites)	Urban	66	$PM_{2.5}$ sampling, preparation and OP^{DTT} measurement were conducted in the same manner as

						Fang et al. (2015).
Gao et al. (2020a) and Gao et al. (2020b)	≤ 2.5	0.005 – 0.070 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ average: 0.024 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ 0.05 – 0.48 $nmol\cdot min^{-1}\cdot m^{-3}$ average: 0.22 $nmol\cdot min^{-1}\cdot m^{-3}$	Atlanta, GA	Urban	349	$PM_{2.5}$ sampling, preparation and OP^{DTT} measurement were conducted in the same manner as Fang et al. (2015).
Hu et al. (2008)	0.25 – 2.5	0.014 – 0.024 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ median: 0.019 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ 0.10 – 0.16 $nmol\cdot min^{-1}\cdot m^{-3}$ median: 0.14 $nmol\cdot min^{-1}\cdot m^{-3}$	Los Angeles harbor, CA	Urban	6	Ambient size-segregated samples were collected with Sioutas samplers on Zefluor and Quartz filters, and extracted in Milli-Q water. DTT assay was conducted by incubating the PM suspensions in 100 μM DTT at pH = 7.4 adjusted by K-PB. DTT was captured by DTNB and measured based on a photometric method (at 412 nm) at designated time points within 30 min.
Cesari et al. (2019)	≤ 2.5	0.012 ± 0.008 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ 0.19 ± 0.10 $nmol\cdot min^{-1}\cdot m^{-3}$	Sarno, Italy	Urban	~50	Ambient $PM_{2.5}$ samples were collected using a Low-Vol sequential sampler on quartz filters, extracted in DI and filtered through a syringe filter. DTT assay was conducted by incubating the extracts in DTT (concentration not reported) at pH = 7.4 adjusted by K-PB. DTT was captured by DTNB and measured based on a photometric method (at 412 nm) at designated time points (details not reported).
Paraskevopoulou et al. (2019)	≤ 2.5	0.028 ± 0.014 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ 0.33 ± 0.20 $nmol\cdot min^{-1}\cdot m^{-3}$	Athens, Greece	Urban	361	Ambient $PM_{2.5}$ samples were collected using a Dichotomous Partisol sampler on quartz filters, extracted in DI and filtered through a syringe filter. OP^{DTT} was assessed in the same manner as Fang et al. (2015).
Perrone et al. (2019)	≤ 2.5	0.010 ± 0.001 $nmol\cdot min^{-1}\cdot \mu g^{-1}$ 0.228 ± 0.024 $nmol\cdot min^{-1}\cdot m^{-3}$	Lecce, Italy	Urban	39	Ambient $PM_{2.5}$ samples were collected using a low volume HYDRA-FAI dual sampler, and extracted in DI. DTT assay was conducted by incubating the aqueous sample extracts in 100 μM DTT. DTT was captured by DTNB and measured based on a photometric method (at 412 nm) at five time points within 40 min.

Yang et al. (2014)	≤ 2.5	0.4 – 7.2 $nmol \cdot s^{-1} \cdot m^{-3}$	Rotterdam and Amsterdam, Netherland	Urban	10	Ambient $PM_{2.5}$ samples were collected using a Harvard Impactor and extracted in ultrapure water. OP^{DTT} of water-soluble extracts was assessed in the same manner as Hu et al. (2008).
Yu et al. (2020)	≤ 2.5	0.004 – 0.193 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.014 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.041 – 1.282 $nmol \cdot min^{-1} \cdot m^{-3}$ median: 0.146 $nmol \cdot min^{-1} \cdot m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54	$PM_{2.5}$ sampling, preparation and OP^{DTT} measurement were conducted in the same manner as the current study.
Verma et al. (2012)*	≤ 2.5	0.020 – 0.054 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.034 $nmol \cdot min^{-1} \cdot \mu g^{-1}$	Atlanta, GA	Urban	8	Ambient $PM_{2.5}$ samples were collected using a Hi-Vol sampler on quartz filters, extracted in both methanol and water, and filtered through a syringe filter. Methanol extracts were evaporated to nearly dryness using a rotary evaporator and reconstituted to 15 mL with 0.1 M K-PB (pH = 7.4). Reconstituted methanol extracts were incubated in 100 μM DTT and 0.5 M K-PB (pH = 7.4). DTT was captured by DTNB and measured based on a photometric method (at 412 nm) at seven time points within 20 min.
Gao et al. (2017)*	≤ 2.5	0.14 – 0.47 $nmol \cdot min^{-1} \cdot m^{-3}$ median: 0.30 $nmol \cdot min^{-1} \cdot m^{-3}$	Atlanta, GA (2 sites)	Urban	66	Method 1: Ambient $PM_{2.5}$ samples were extracted in a stepwise manner with DI and methanol. Both extracts were filtered through a syringe filter. Methanol extracts were evaporated to ~200 μL using high-purity nitrogen and reconstituted with DI. Total OP was calculated by adding the OP of both extracts. Method 2: Samples were extracted in methanol. Punches were removed after sonication. The remaining suspensions were analyzed for OP^{DTT} without being filtered through a syringe filter. Method 3: Samples were sonicated in K-PB (pH = 7.4). The mixture was analyzed for OP^{DTT} without removing inside punches or being filtered through a syringe filter. OP^{DTT} measurement was conducted in the same

						manner as Fang et al. (2015) using a modified automated system for analyzing suspensions with insoluble fractions.
Gao et al. (2020b)*	≤ 2.5	0.012 – 0.116 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ average: 0.027 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.13 – 0.58 $nmol \cdot min^{-1} \cdot m^{-3}$ average: 0.28 $nmol \cdot min^{-1} \cdot m^{-3}$	Atlanta, GA	Urban	349	$PM_{2.5}$ sampling, preparation and OP^{DTT} measurement were conducted in the same manner as Gao et al. (2017) (Method 3).
Yang et al. (2014)*	≤ 2.5	0.5 – 5.2 $nmol \cdot min^{-1} \cdot m^{-3}$	Rotterdam and Amsterdam, Netherland	Urban	20	Ambient $PM_{2.5}$ samples were collected using a Harvard Impactor and extracted in methanol. Filtered methanol extracts were evaporated using an evaporator set, and reconstituted with DI. OP^{DTT} of water-reconstituted methanol-soluble extracts was assessed in the same manner as Hu et al. (2008).
This study	≤ 2.5	0.004 – 0.032 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.014 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.029 – 0.561 $nmol \cdot min^{-1} \cdot m^{-3}$ median: 0.150 $nmol \cdot min^{-1} \cdot m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	241	See section 2 (experimental methods).
This study*	≤ 2.5	0.004 – 0.042 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.021 $nmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.031 – 0.639 $nmol \cdot min^{-1} \cdot m^{-3}$ median: 0.234 $nmol \cdot min^{-1} \cdot m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	241	

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

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

Reference	**PM size (μm)**	**Levels**	**Location**	**Location type**	**Sample size**	**Methodology**
Xiong et al. (2017)	≤ 2.5	0.2 – 0.6 $pmol \cdot min^{-1} \cdot m^{-3}$	Urbana, IL	Urban	10	$PM_{2.5}$ extracts were incubated in 100 μM DTT and K-PB (pH = 7.4) with 50 mM TPT. ·OH was captured by TPT and measured based on a fluorometric method (excitation/emission wavelength of 310/425 nm) at six time points within 120 min.
Yu et al. (2018)	≤ 2.5	0.2 – 1.1 $pmol \cdot min^{-1} \cdot m^{-3}$	Urbana, IL	Urban	10	$PM_{2.5}$ sampling, preparation and $OP^{OH\text{-}DTT}$ measurement were conducted in the same manner as Xiong et al. (2017).
Yu et al. (2020)	≤ 2.5	0.034 – 0.357 $pmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.082 $pmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.360 – 4.152 $pmol \cdot min^{-1} \cdot m^{-3}$ median: 1.054 $pmol \cdot min^{-1} \cdot m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	54	$PM_{2.5}$ sampling, preparation and $OP^{OH\text{-}DTT}$ measurement was conducted in the same manner as the current study.
This study	≤ 2.5	0.004 – 0.357 $pmol \cdot min^{-1} \cdot \mu g^{-1}$ median: 0.065 $pmol \cdot min^{-1} \cdot \mu g^{-1}$ 0.022 – 3.565 $pmol \cdot min^{-1} \cdot m^{-3}$ median: 0.722 $pmol \cdot min^{-1} \cdot m^{-3}$	Midwest US (5 sites)	Urban (4), rural (1)	241	See section 2 (experimental methods).

Table S7. Seasonal median of the ratio of methanol-soluble OPv to water-soluble OPv (M/W^{OP}) for $OP^{OH\text{-}SLF}v$ 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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