Supplementary Materials: Insight into the Source and Evolution of Oxalic Acid: Characterization of Particulate Organic Diacids in a Mega-City, Shanghai from 2008 to 2020

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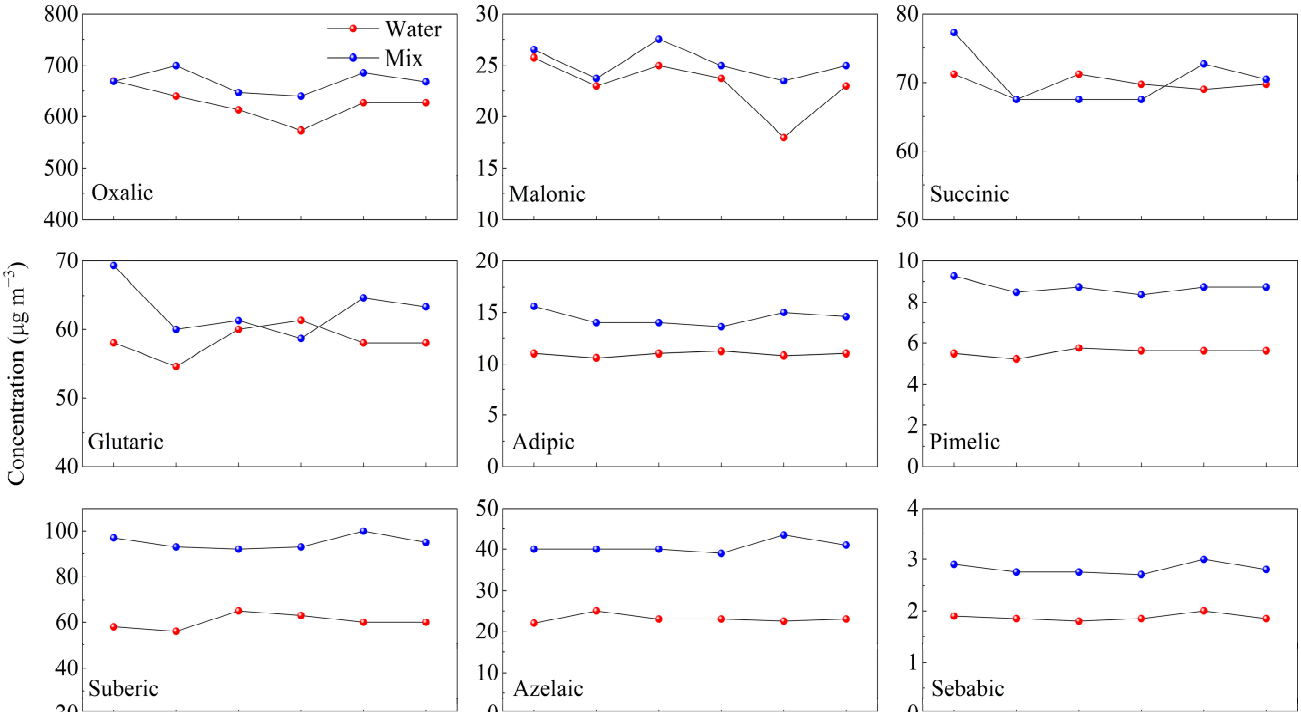


Figure S1. Comparison of the measured concentrations of different diacids in $PM_{2.5}$ extracted with Water (100% water) and Mix (50% methanol/water solution (*v*/*v*)).

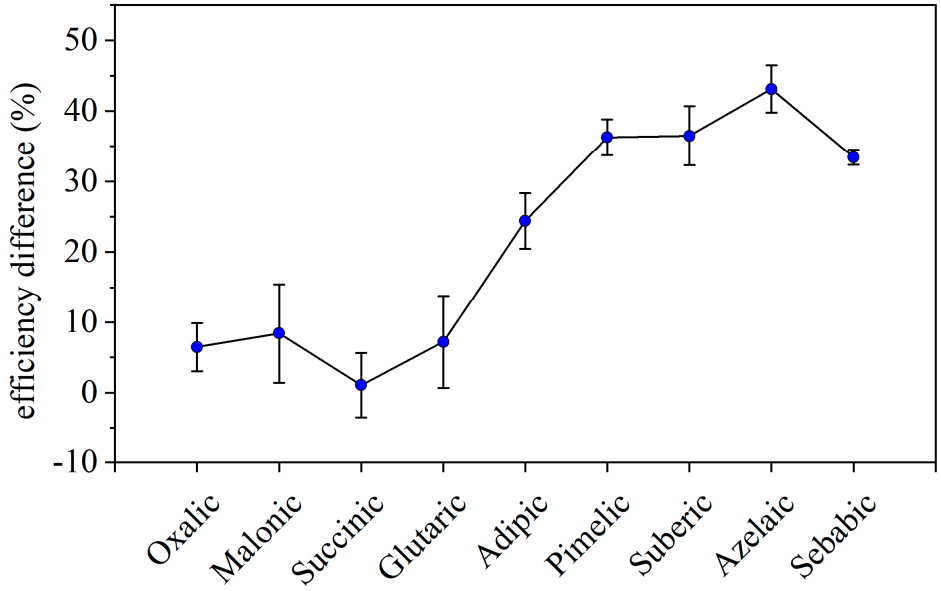


Figure S2. Differences in concentrations of particulate diacids measured by extraction with 50% methanol/water solution (v/v) compared to 100% water.

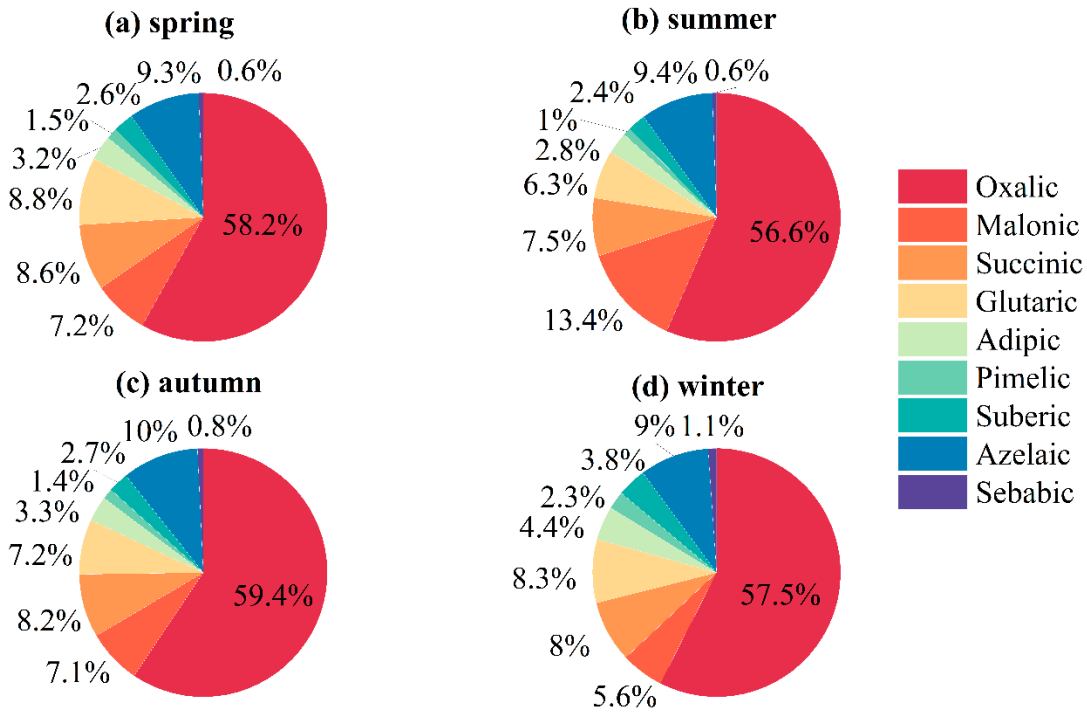


Figure S3. Seasonal variation of the diacid content in $PM_{2.5}$ at the BS site.

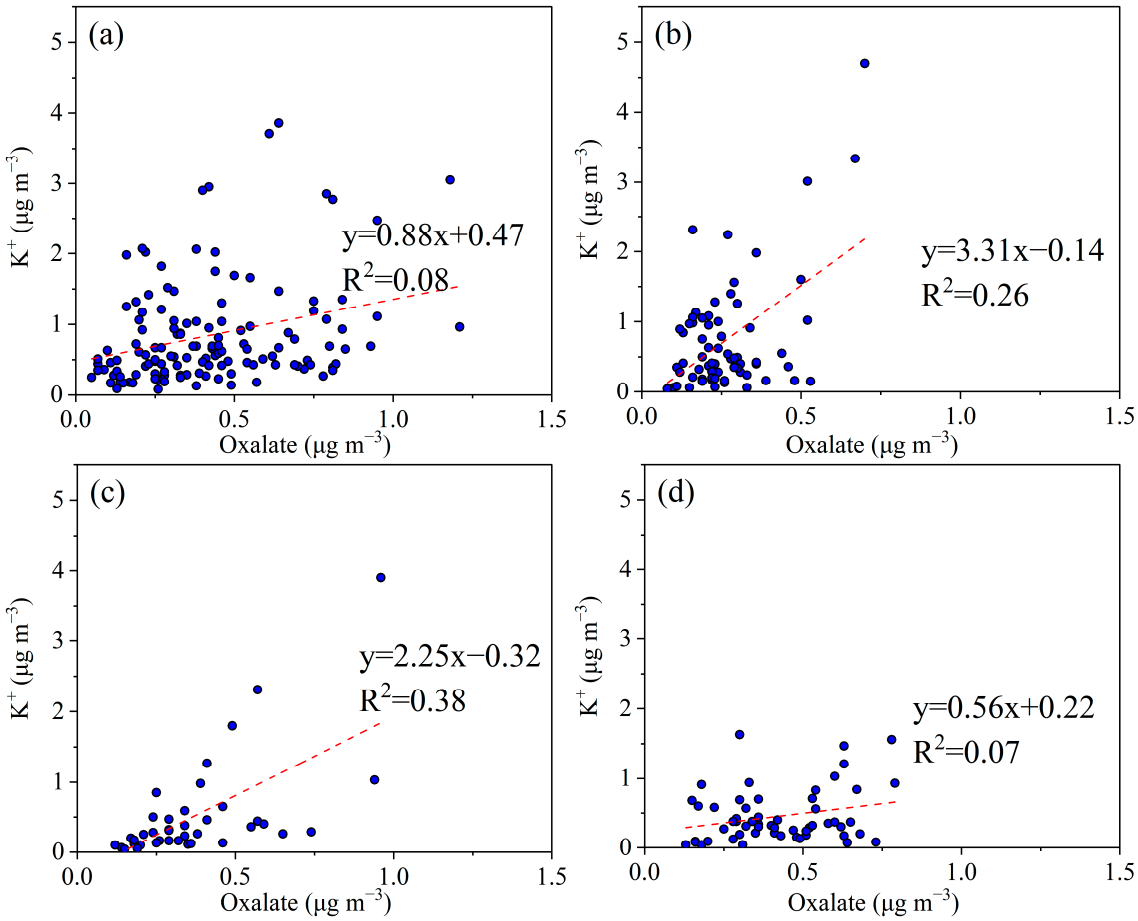


Figure S4. Correlation between oxalate and K^+ in (a):2008; (b):2010; (c):2013; (d):2014 at the BS site.

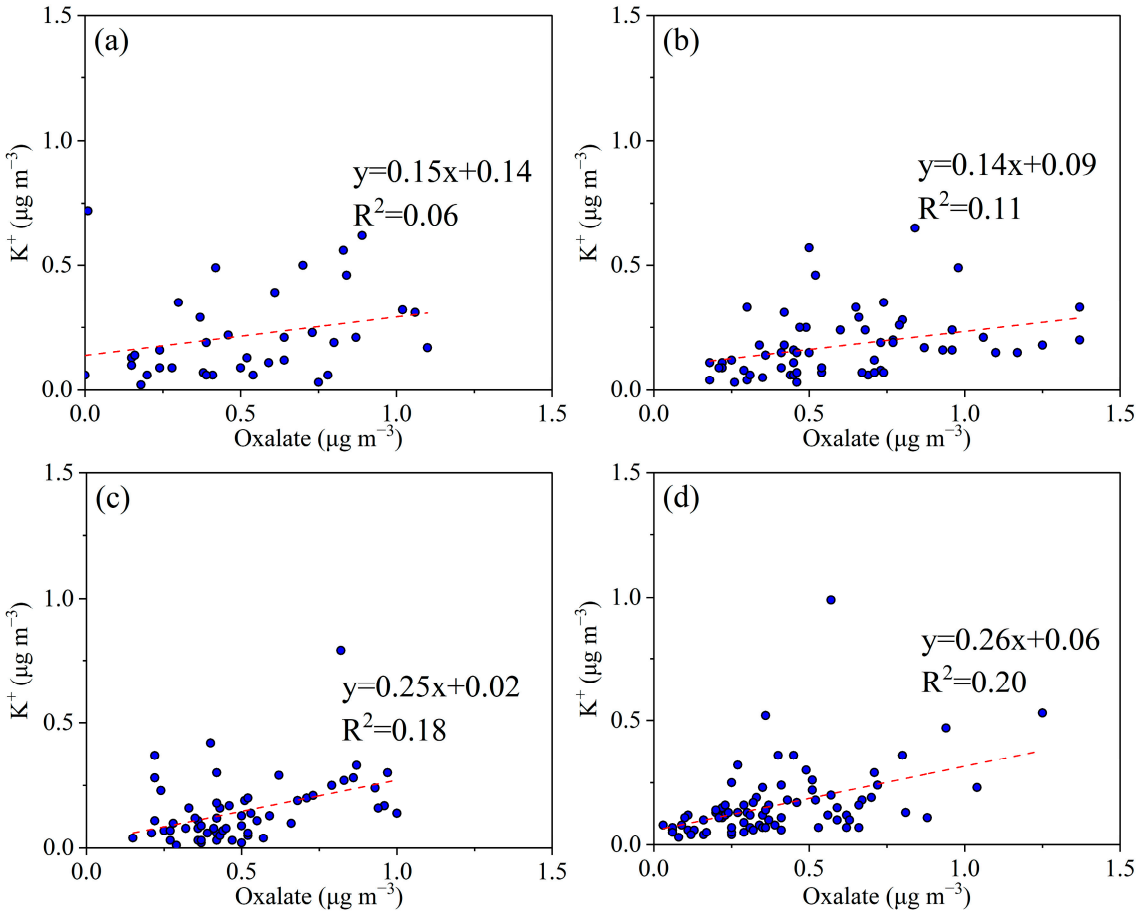


Figure S5. Correlation between oxalate and K^+ in (a):2016; (b):2017; (c):2018; (d):2019 at the PD site.

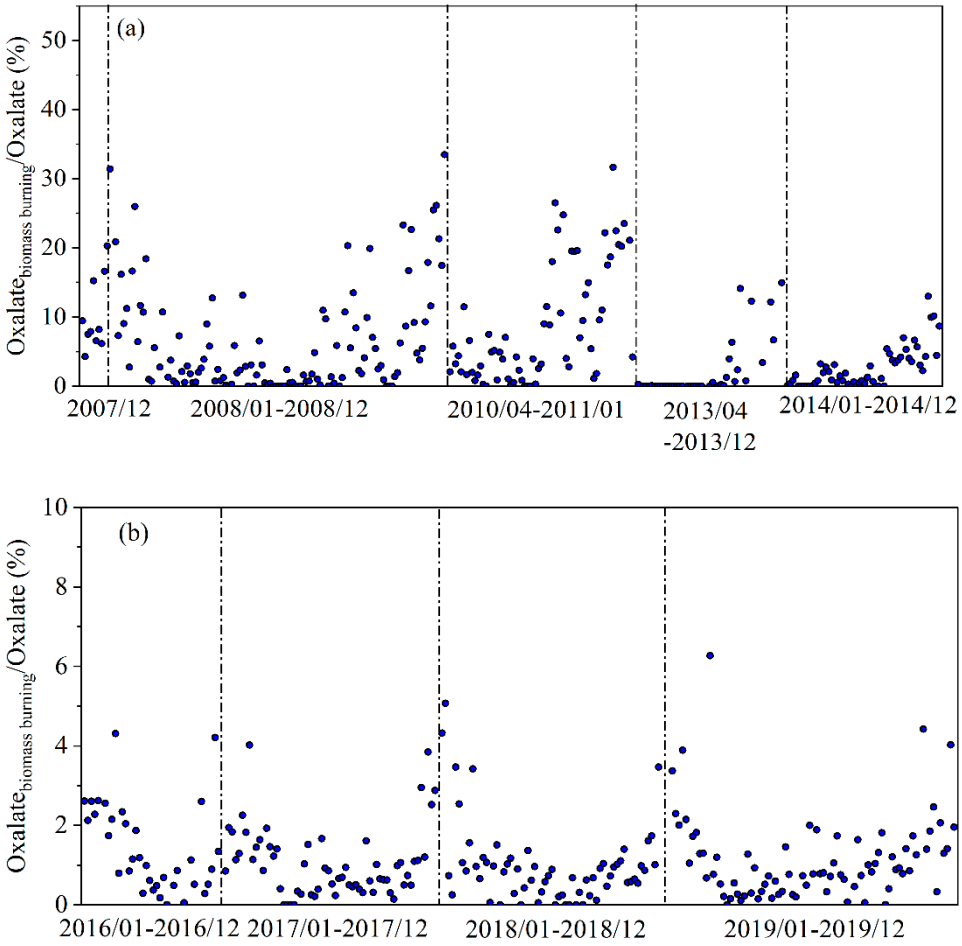


Figure S6. Time series of BB contribution to oxalate in BS (a) and PD (b).

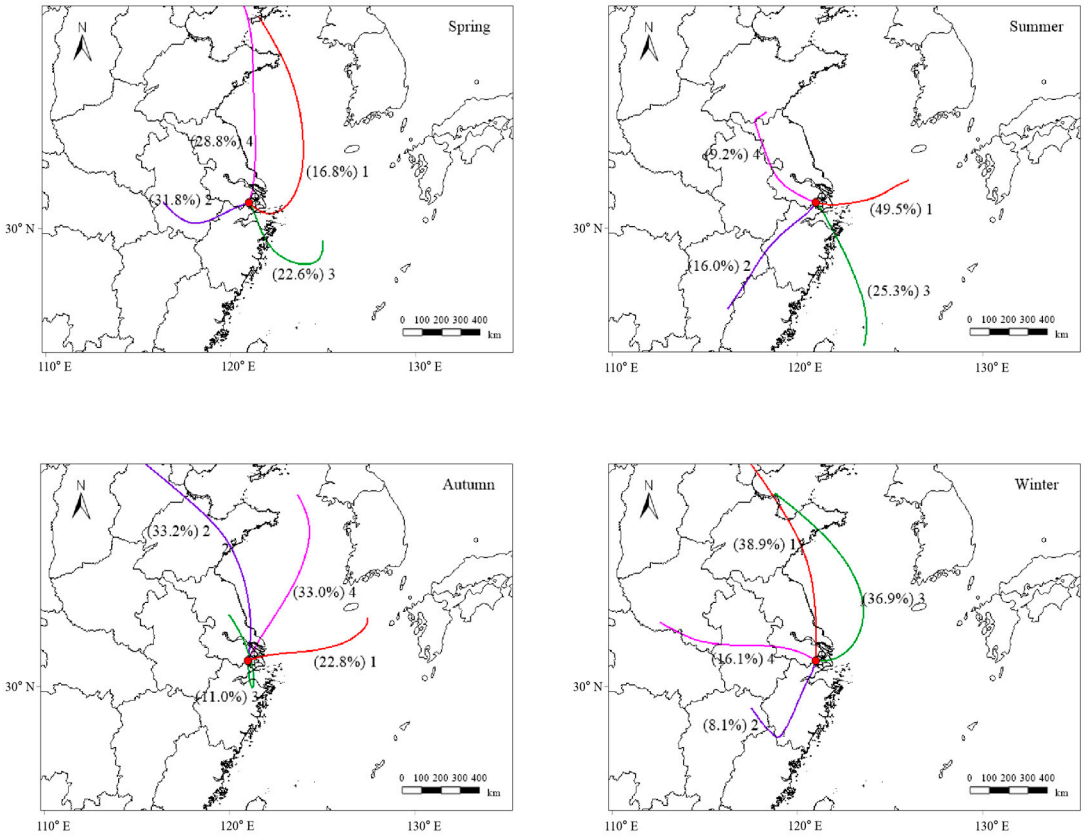
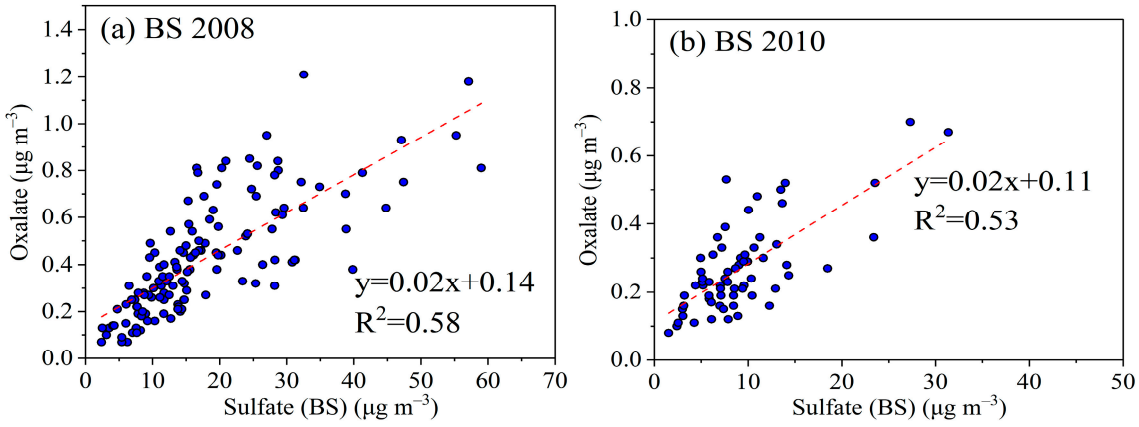


Figure S7. Clusters of backward trajectories for air parcels arriving in Shanghai during 2019.



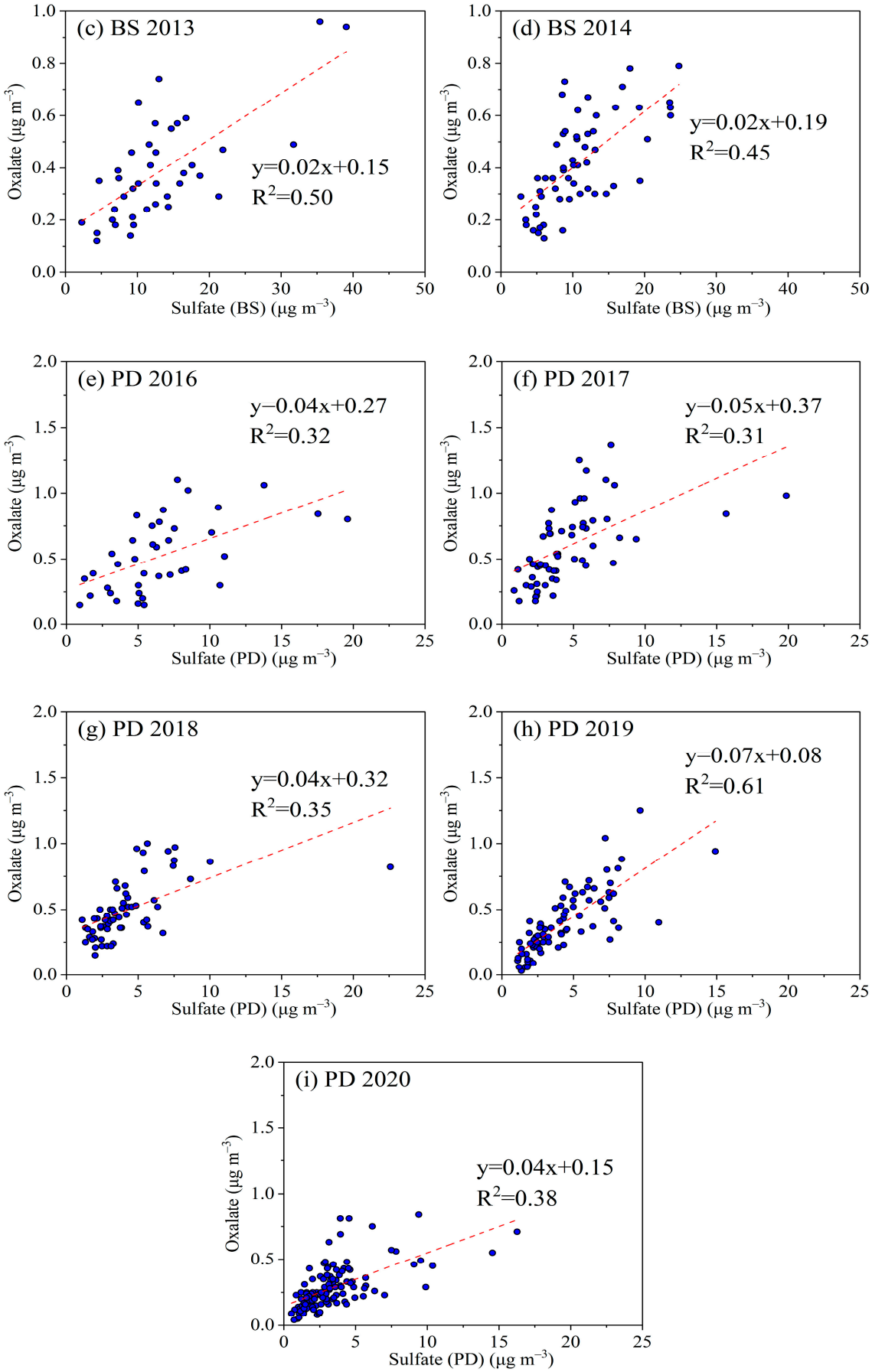
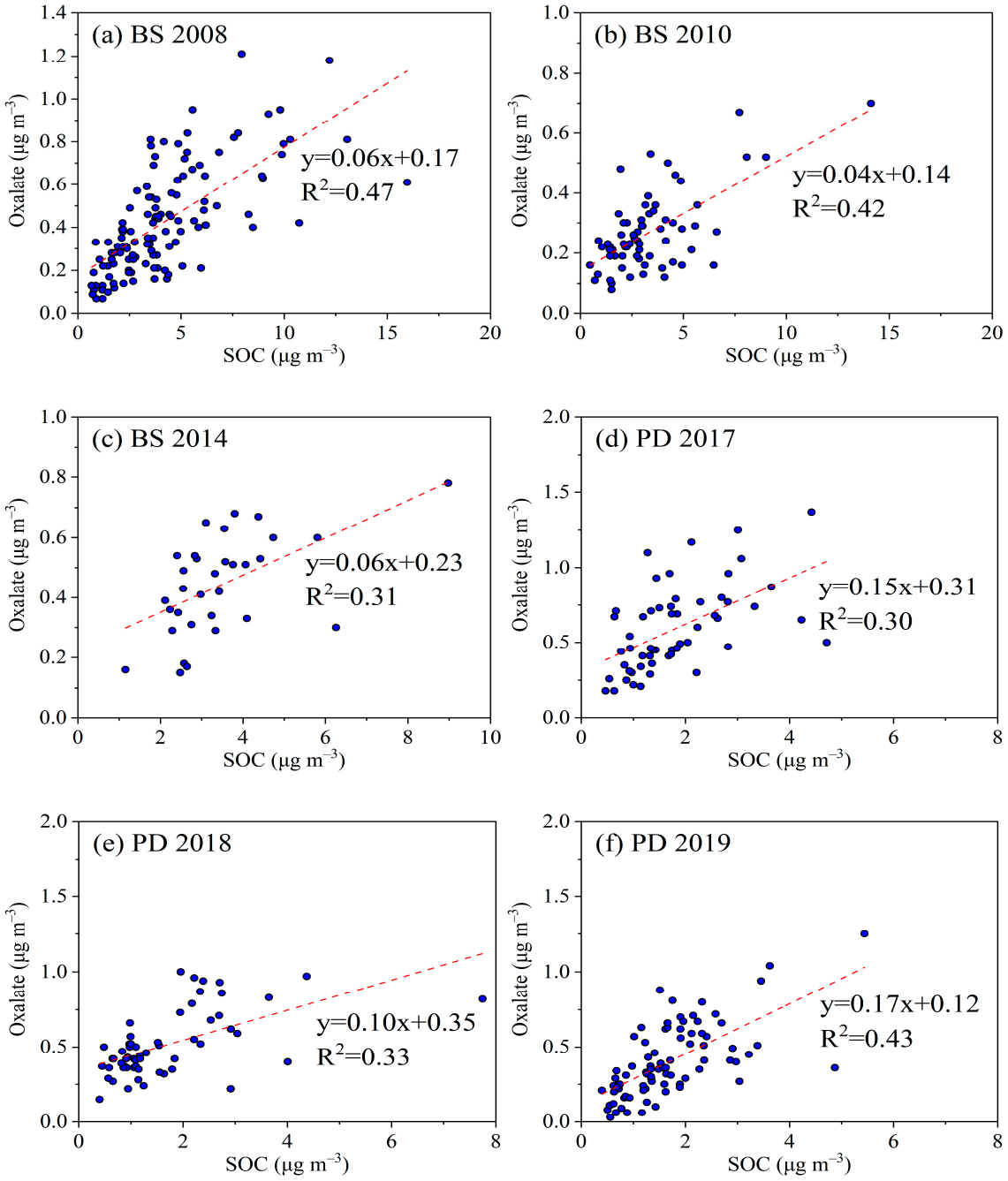

(c) BS 2013
y=0.02x+0.15
R2=0.50
Oxalate (μg m−3)
Sulfate (BS) (μg m−3)
(d) BS 2014
y=0.02x+0.19
R2=0.45
Oxalate (μg m−3)
Sulfate (BS) (μg m−3)
(e) PD 2016
y=0.04x+0.27
R2=0.32
Oxalate (μg m−3)
Sulfate (PD) (μg m−3)
(f) PD 2017
y=0.05x+0.37
R2=0.31
Oxalate (μg m−3)
Sulfate (PD) (μg m−3)
(g) PD 2018
y=0.04x+0.32
R2=0.35
Oxalate (μg m−3)
Sulfate (PD) (μg m−3)
(h) PD 2019
y=0.07x+0.08
R2=0.61
Oxalate (μg m−3)
Sulfate (PD) (μg m−3)
(i) PD 2020
y=0.04x+0.15
R2=0.38
Oxalate (μg m−3)
Sulfate (PD) (μg m−3)

Figure S8. Oxalate correlation with sulfate: (a-i:BS2008-PD2020).



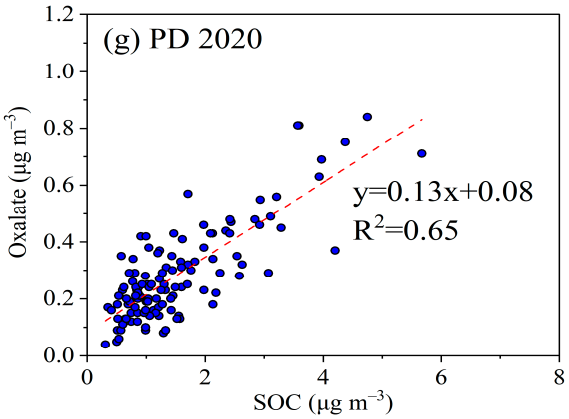
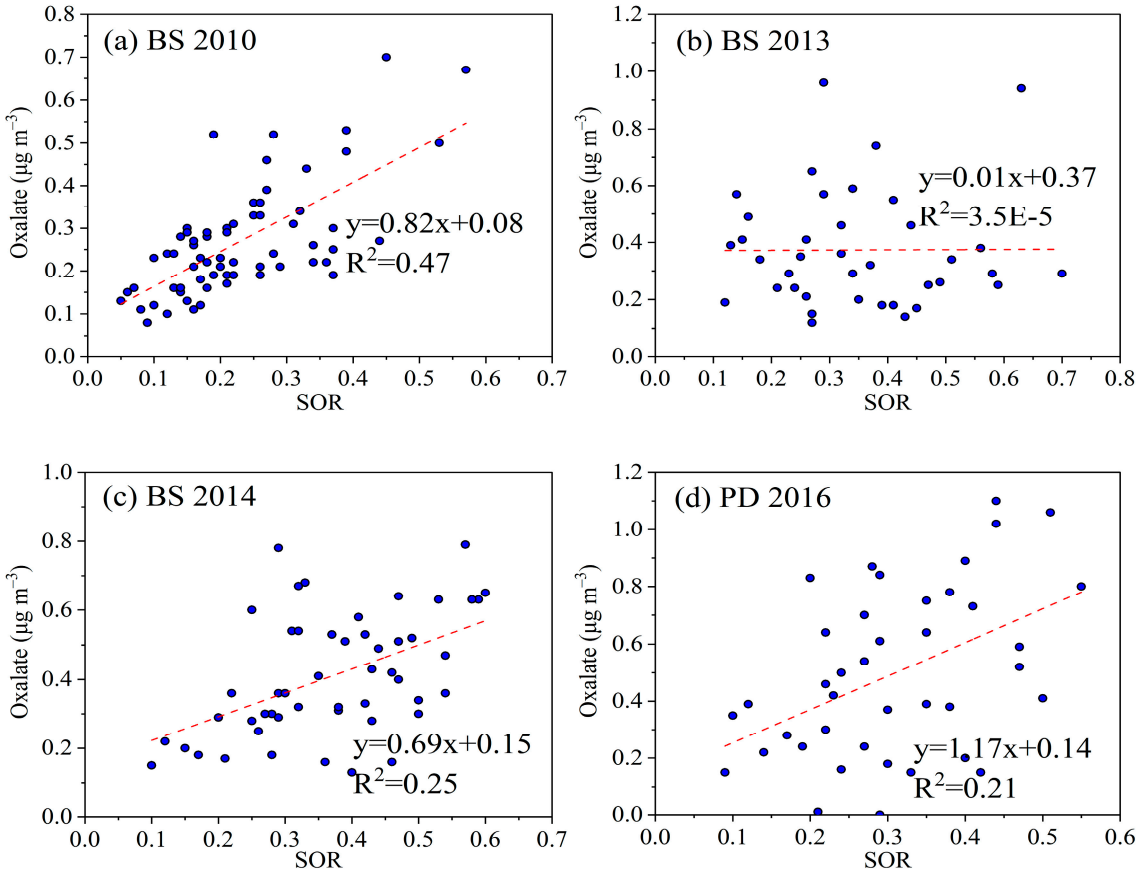


Figure S9. Oxalate correlation with SOC (a-g:BS2008-PD2020).



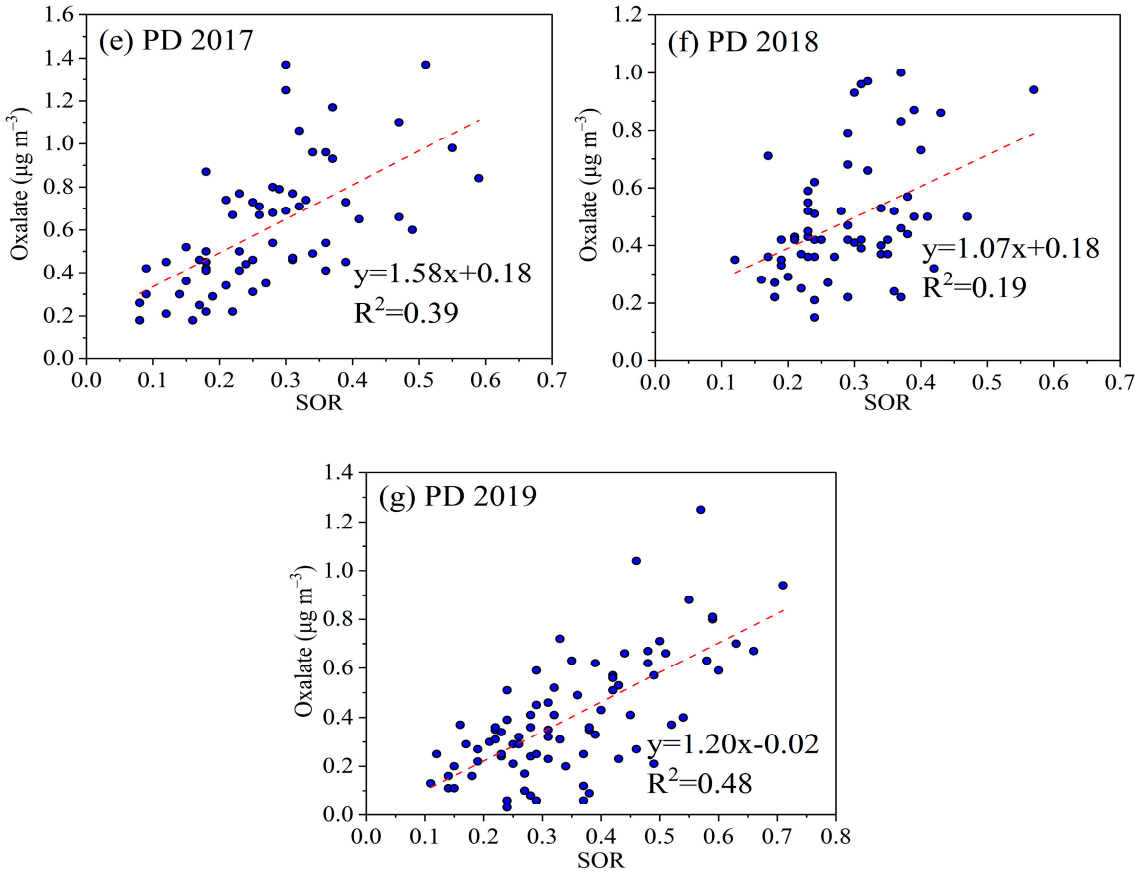


Figure S10. Oxalate correlation with SOR (a-g:BS2010-PD2019).

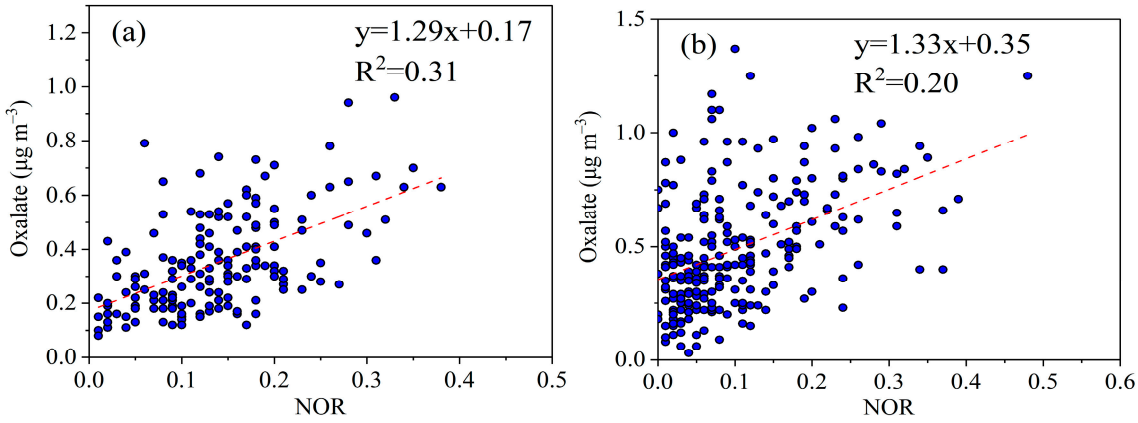


Figure S11. Oxalate correlation with NOR for BS (a) and PD(b).

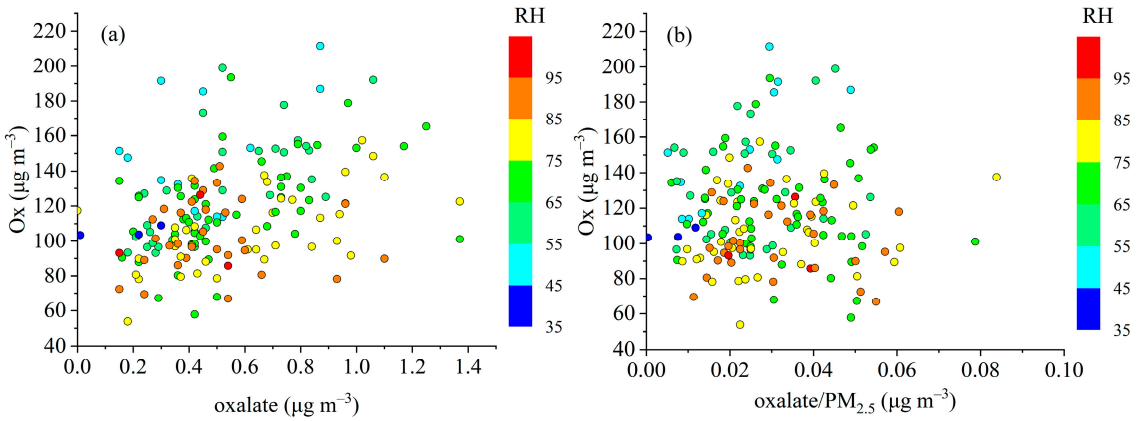


Figure S12. (a) RH distribution of oxalate and Ox; (b) RH distribution of oxalate/PM2.5 and Ox.

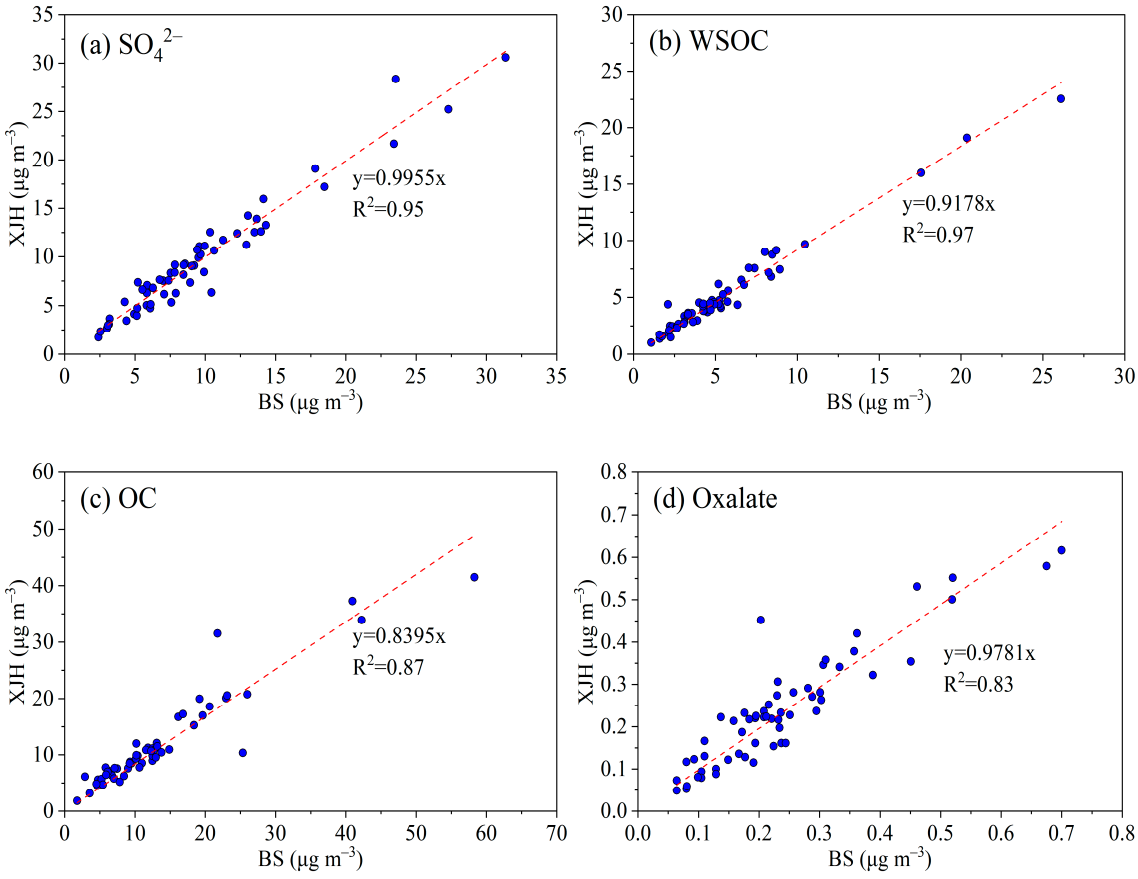


Figure S13. pollutants correlation in BS and XJH.

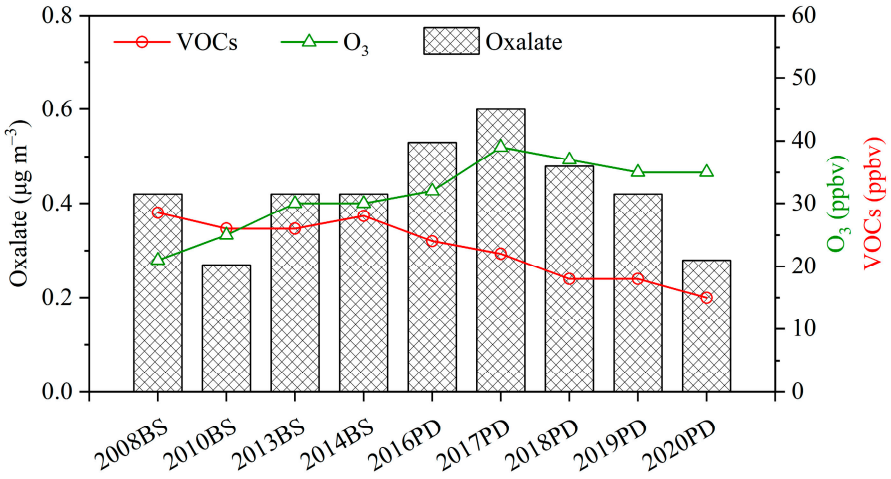


Figure S14. Interannual variation of the concentrations of oxalate, VOCs and O_3 in Shanghai (Data of VOCs of 2008-2014 was from Peng et al [1,2]; and VOCs of 2016-2020 and O_3 were from the Shanghai environmental monitoring center.).

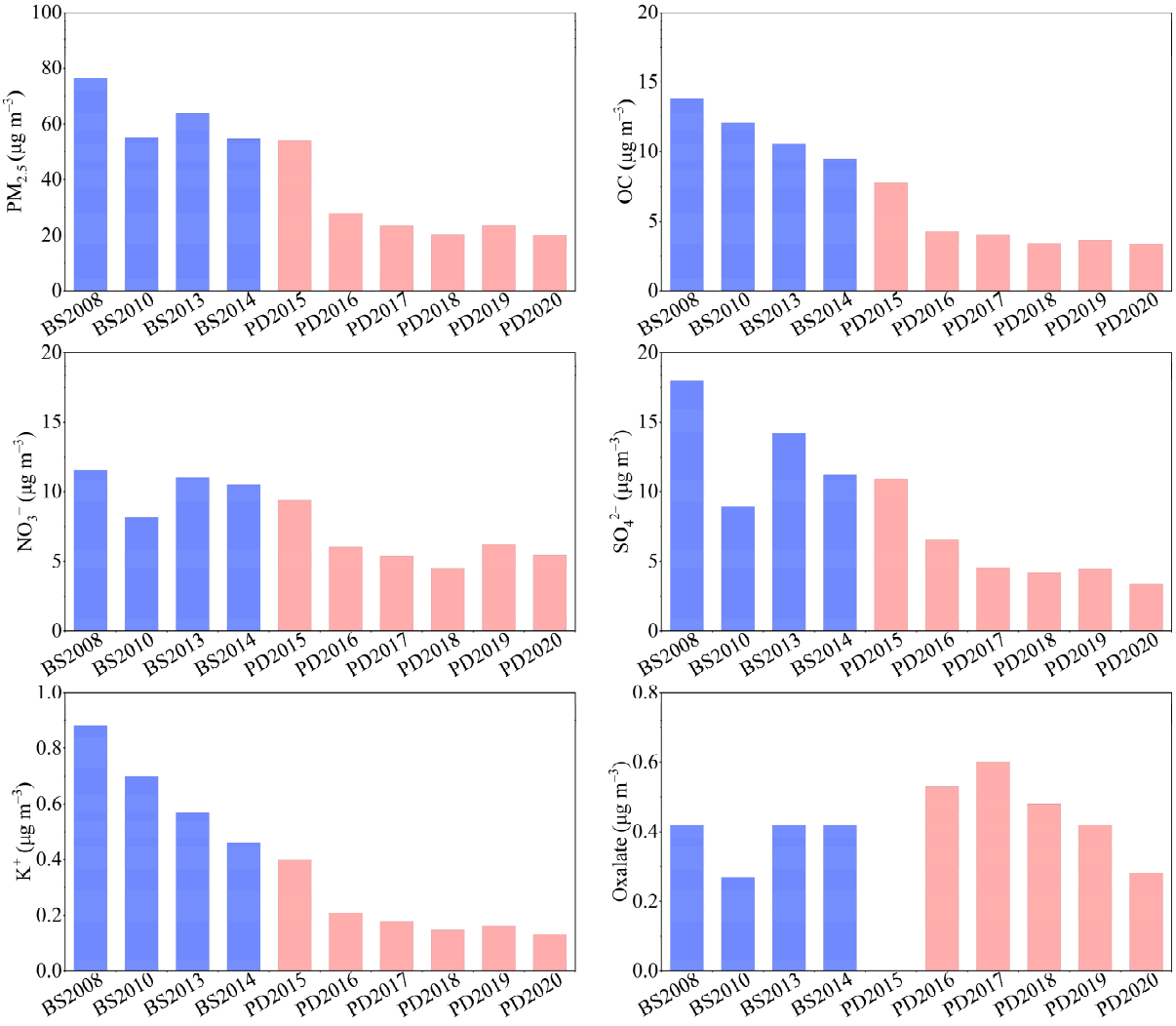


Figure S15. Evolution of particulate pollutants for Shanghai from 2008 to 2020. Blue bars represent BS and red bars represent PD.

Table S1. The ratio of Ca^{2+}/K^{+} in biomass burning aerosol reported in previous studies.

Sampling	Biomass type	Year	Region	K^+(mg kg^{-1})	Ca^{2+}(mg kg^{-1})	Ca^{2+}/K^+	References
Chamber	Corncob	2018.8-2019.2	Liaoning	174.66±46.01	18.47±2.14	0.11	[3]
Chamber	Corncob	2018.8-2019.2	Shanxi	254.70±41.23	4.03±0.57	0.02	[3]
Chamber	Corncob	2018.8-2019.2	Henan	208.17±53.47	44.92±34.51	0.22	[3]
Chamber	Corncob	2018.8-2019.2	Shandong	540.73±95.65	12.21±4.49	0.02	[3]
Chamber	Corncob	2018.8-2019.2	Hubei	269.02±45.58	22.31±11.15	0.08	[3]
Chamber	Wheat straw			530±250	82±72	0.15	[4]
Chamber	Rice straw			900±870	77±30	0.09	[4]
Chamber	Corn straw			280±120	88±43	0.31	[4]
Chamber	Wheat straw			350±180	30±20	0.09	[5]
Chamber	Corn straw			390±210	60±40	0.15	[5]
Chamber	Rice straw			390±280	30±10	0.08	[5]
Chamber	Wheat straw			490.21±359.74	19.35±75.43	0.04	[6]
Chamber	Corn straw			134.80±84.31	60.54±52.64	0.45	[6]
Chamber	Rice straw			246.10±297.61	65.79±60.95	0.27	[6]

Table S2. Annual $PM_{2.5}$ concentrations at 9 monitoring stations in Shanghai during 2015-2021**.**

Site	Huangpu	Hongkou	Xujiahui	Yangpu	Qingpu	Jing'an	Chuansha	PudongN	Zhangjiang	Baoshan	Average	CV
2015	55	52	53	54	63	55	50	52	51	56	54	6.7%
2016	48	45	44	46	53	49	40	44	47	46	46	7.1%
2017	38	40	38	39	39	44	40	36	37	40	39	5.3%
2018	36	35	37	37	42	35	34	34	36	34	36	6.4%
2019	34	35	37	37	37	38	35	34	34	36	36	4.0%
2020	33	32	33	32	36	32	30	30	32	33	32	5.1%
2021	/	28	29	27	29	29	26	28	29	28	28	3.1%

References

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2. Xu, J.; Tie, X.; Gao, W.; Lin, Y.; Fu, Q. Measurement and model analyses of the ozone variation during 2006 to 2015 and its response to emission change in megacity Shanghai, China. *Atmos. Chem. Phys.* **2019**, *19*, 9017–9035.
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5. Zhang, H.; Hu, J.; Qi, Y.; Li, C.; Chen, J.; Wang, X.; He, J.; Wang, S.; Hao, J.; Zhang, L. Emission characterization, environmental impact, and control measure of $PM_{2.5}$ emitted from agricultural crop residue burning in China. *J. Cleaner Prod*. **2017**, *149*, 629–635.
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