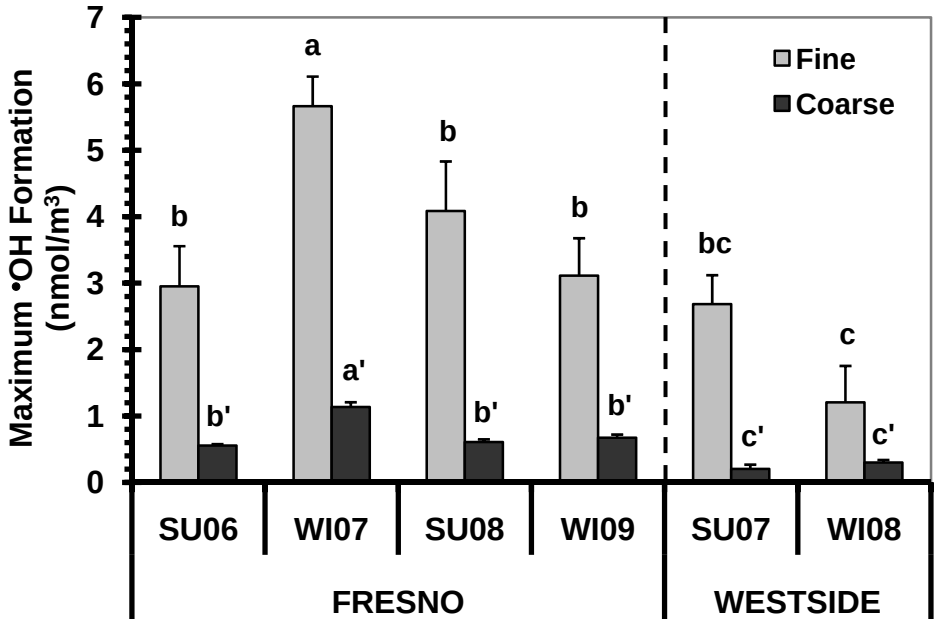
Supplementary Material

Table S1. Dates and substrates of sample collection, and time in frozen storage prior to ROS analysis.

Sample ID[a]	Dates of Collection	Substrate[b]		Days in -20 °C Storage Prior to Analysis[c]						
				HOOH				OH		
				w/Asc		w/o Asc		w/Asc		w/o Asc
		fine	coarse	fine	coarse	fine	Coarse	fine	coarse	fine & coarse
Fresno Samples										
FRSU06	5-9 & 12-16 Sep 2006	pure Teflon	baked foil	962, 1025, 1031	1090, 1095, 1112	1069	1118, 1125, 1146	1272, 1397, 1404, 1432	1272, 1374, 1399, 1432	1418, 1421, 1427
FRWI07	13-17 & 20-24 Feb 2007	pure Teflon	baked foil	808, 865, 877	929, 934, 951	908	957, 964, 985	1111, 1236, 1243, 1271	1111, 1213, 1238, 1271	1257, 1260, 1266
FRSU08	24-28 & 31 Aug -4 Sep 2008	pure Teflon	baked foil	243, 306, 312	371, 376, 393	350	399, 406, 427	553, 678, 685, 713	553, 655, 680, 713	699, 702, 708
FRWI09	10-14 & 17-21 Jan 2009	TX 40	baked foil	111, 168, 180	232, 237, 254	211	260, 267, 288	414, 539, 546, 574	414, 516, 541, 574	560, 563, 569
Westside Samples										
WESU07	14-18 & 21-25 Aug 2007	TX 40	baked foil	619, 682, 688	747, 752, 769	726	775, 782, 803	929, 1054, 1061, 1089	929, 1031, 1056, 1089	1075, 1078, 1084
WEWI08	6-10 & 13-17 Feb 2008	pure Teflon	baked foil	450, 507, 519	571, 576, 593	550	599, 606, 627	753, 878, 885, 913	753, 855, 880, 913	899, 902, 908

[a] Sample nomenclature: FR = Fresno, WE = Westside, SU = summer, WI = winter, ## = year (20xx). [b] TX 40 was Teflon-coated borosilicate glass microfibers, and the foil was baked at 400 °C for 24 h before collecting samples. [c] Multiple entries represent multiple days of analysis, with samples typically being analyzed in triplicate, with one analysis on each of three different days.

A


Maximum •OH Formation (nmol/m³)
7
6
5
4
3
2
1
0
Fine
Coarse
b
b'
a
a'
b
b'
b
b'
bc
c'
c
c'
SU06
WI07
SU08
WI09
SU07
WI08
FRESNO
WESTSIDE

B

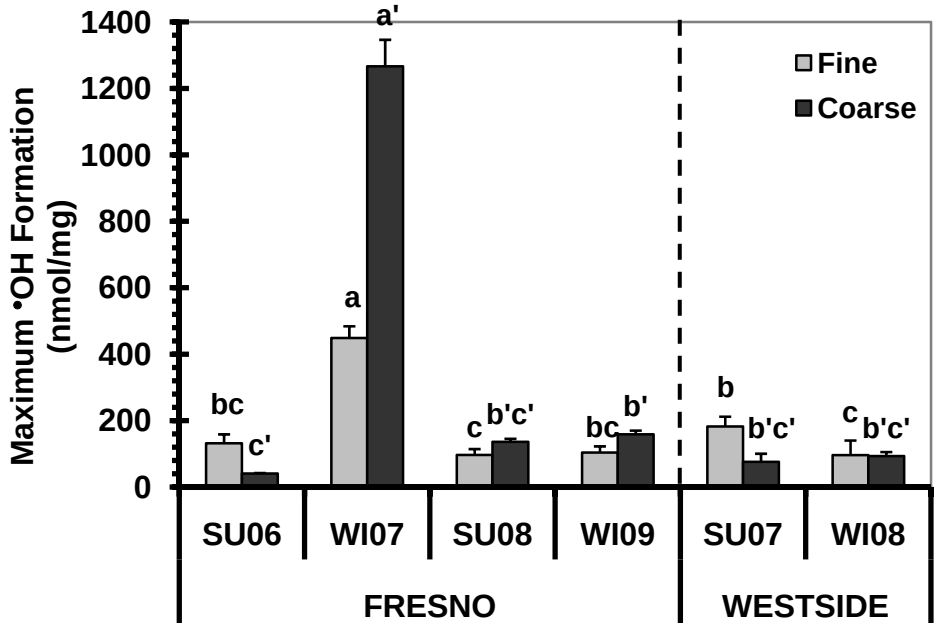
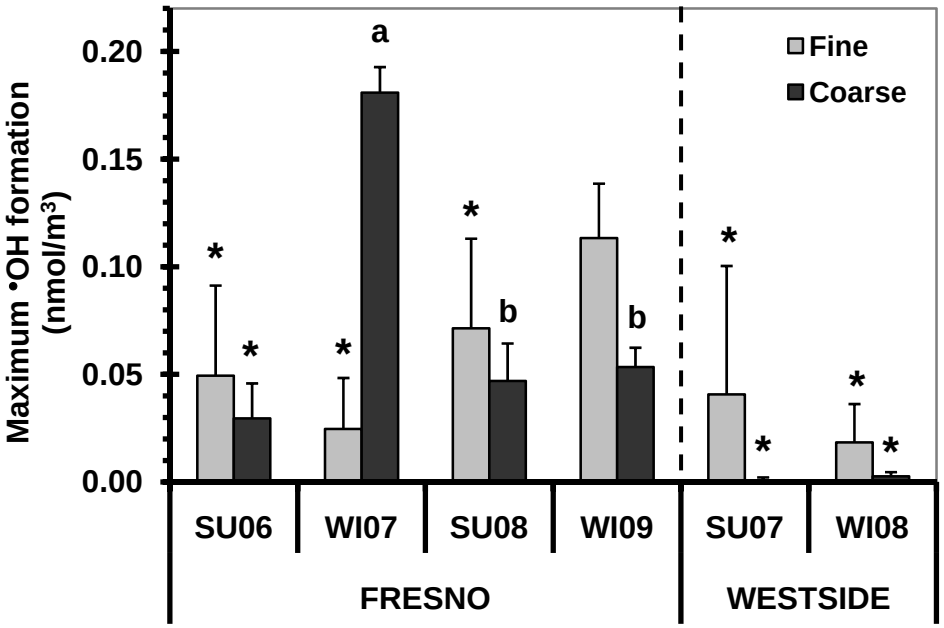

Maximum •OH Formation (nmol/mg)
1400
1200
1000
800
600
400
200
0
Fine
Coarse
bc
c'
a
a'
c
b'c'
bc
b'
b
b'c'
c
b'c'
SU06
WI07
SU08
WI09
SU07
WI08
FRESNO
WESTSIDE

Figure S1. Maximum ˙OH generation in the presence of 50 µM ascorbate. Panel (A) shows air-volume-normalized maximum levels of ˙OH formation, while (B) shows PM-mass-normalized maxima. Values are means ± SD, n = 3 to 4. Letters above bars indicate statistically different maxima: a > b > c for fine PM, while a’ > b’ > c’ for coarse PM.

A


Maximum •OH formation (nmol/m³)
0.20
0.15
0.10
0.05
0.00
Fine
Coarse
a
b
b
*
SU06
WI07
SU08
WI09
SU07
WI08
FRESNO
WESTSIDE

B

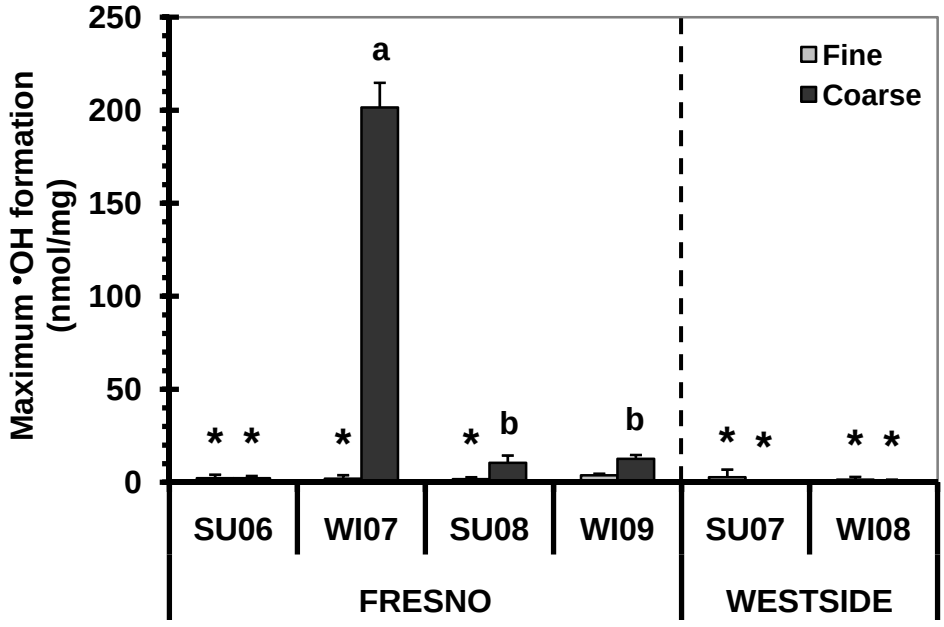

Maximum •OH formation (nmol/mg)
250
200
150
100
50
0
Fine
Coarse
a
b
b
*
SU06
WI07
SU08
WI09
SU07
WI08
FRESNO
WESTSIDE

Figure S2. Maximum ·OH generation in the absence of ascorbate. Panel (A) shows air-volume-normalized maximum levels of ·OH formation, while (B) shows PM-mass-normalized maxima. Values are means ± SD, n = 3. Letters above bars indicate statistically different maxima: a > b. An asterisk "*" indicates a value that is not statistically different from zero.

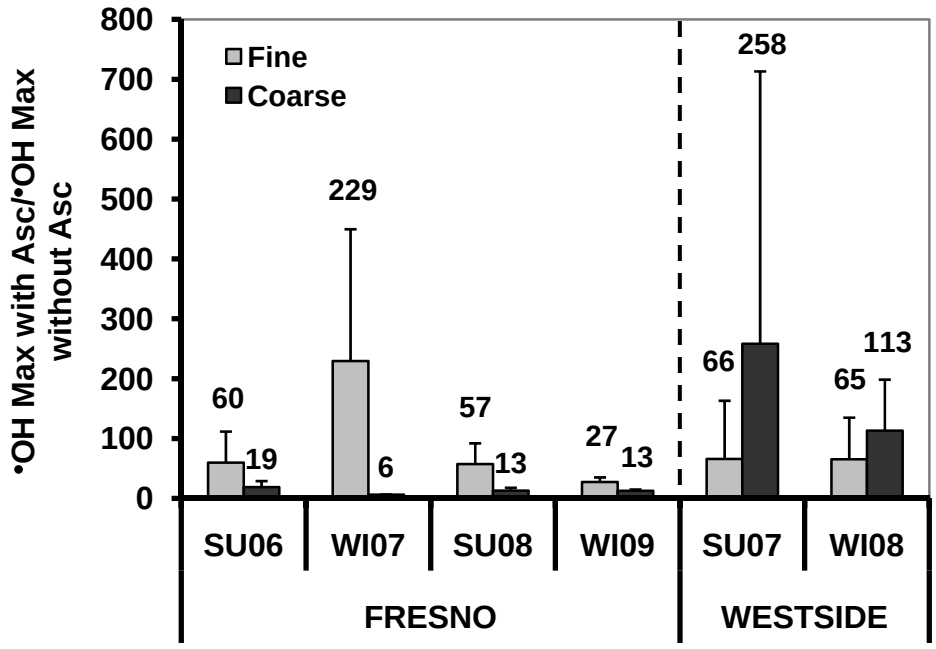


Figure S3. Ratios of the maximum •OH formation in SLF with ascorbate over the maximum •OH formation in SLF without ascorbate. Since the maximum •OH formation without ascorbate in all samples except the Fresno winter 2009 fine PM, the Fresno winter 2007, summer 2008, and winter 2009 coarse PM was not statistically different from zero (Fig. S2), we are likely underestimating the effect of ascorbate in amplifying •OH generation for the majority of the PM samples.

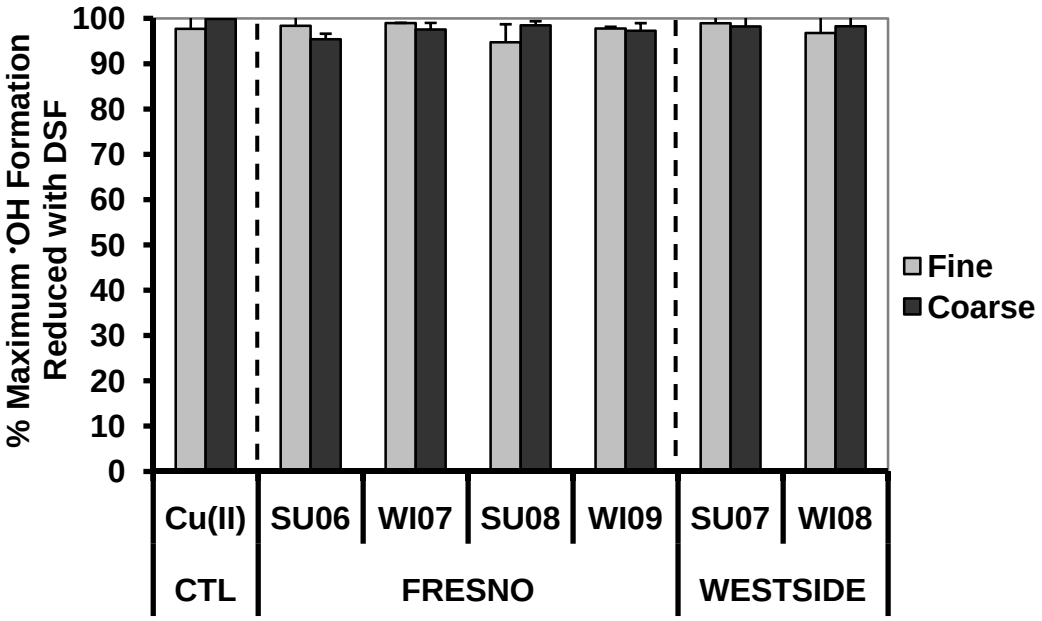


Figure S4. Inhibitory effect of the transition metal chelator DSF on the maximum ·OH generation in SLF with ascorbate for the positive control and the SJV PM. Values are means ± SD. n = 4 for extractions without added DSF, and n = 2 to 3 for extractions with added DSF.

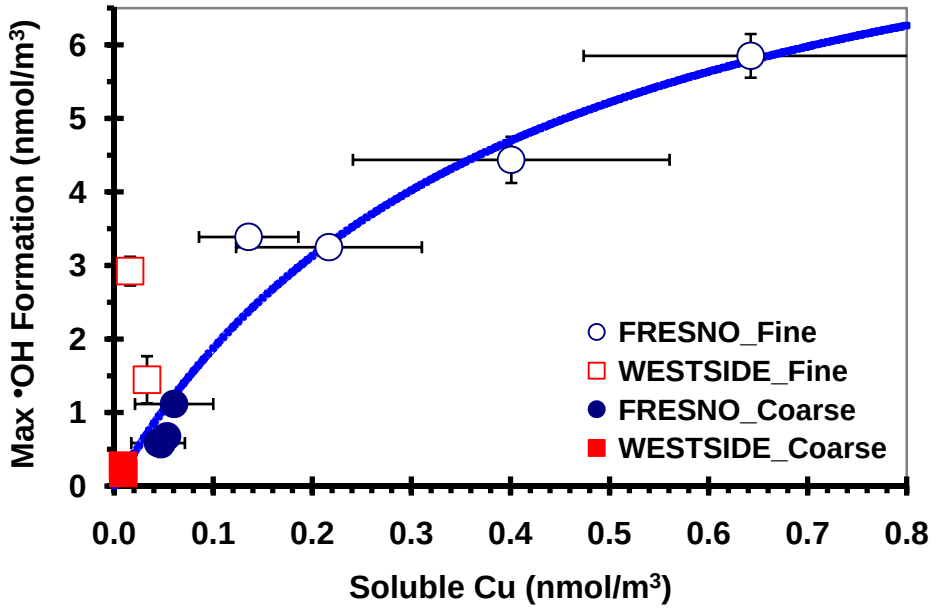
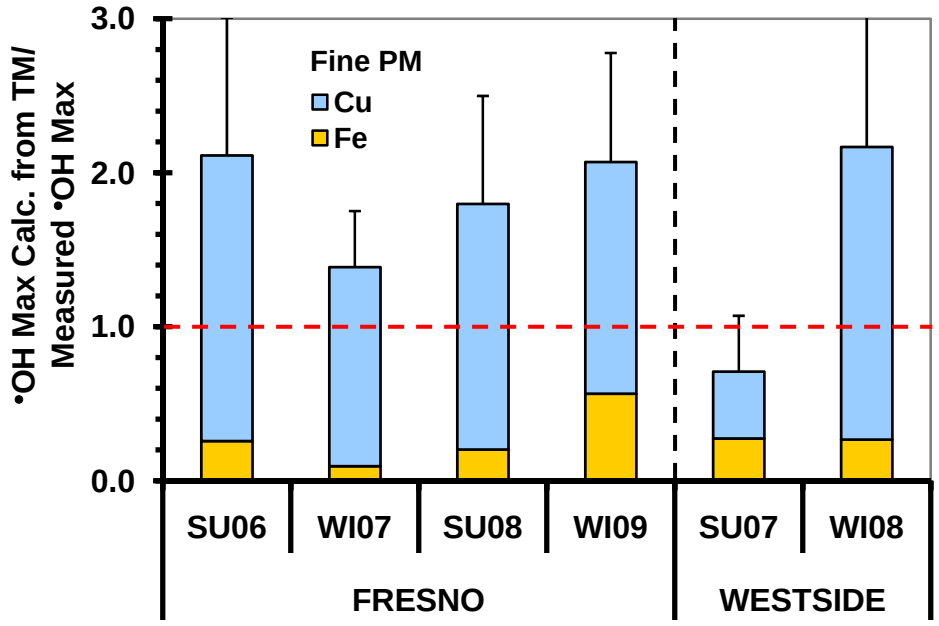


Figure S5. Correlation between the air-volume-normalized maximum level of ˙OH generation in SLF with Asc and the the accompanying SLF-soluble Cu concentration. Values are means ± SD. n = 6 except for a few of the Cu concentrations where n = 4. The maximum levels of ˙OH formation by Fresno fine and coarse PM were strongly correlated with the SLF-soluble Cu concentrations in corresponding PM extracts: $y = 23.45x/(1+ 2.49x)$, $R^2 = 0.95$. No correlation was observed between the SLF-soluble Cu concentrations and the maximum levels of ˙OH formation from the Westside PM ($R^2 = 0.17$).

A


Fine PM
Cu
Fe
•OH Max Calc. from TM/
Measured •OH Max
3.0
2.0
1.0
0.0
SU06
WI07
SU08
WI09
SU07
WI08
FRESNO
WESTSIDE

B

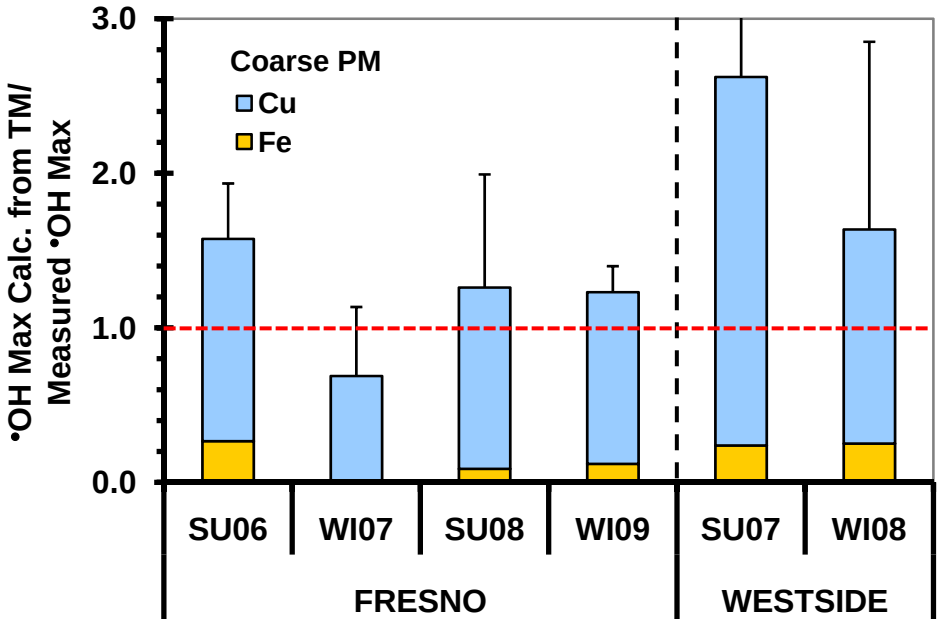

Coarse PM
Cu
Fe
•OH Max Calc. from TM/
Measured •OH Max
3.0
2.0
1.0
0.0
SU06
WI07
SU08
WI09
SU07
WI08
FRESNO
WESTSIDE

Figure S6. Contributions of SLF-soluble Cu (blue bars) and Fe (yellow bars) to the maximum $^{\bullet}$OH generation in SLF with Asc in fine (panel A) and coarse (panel B) particles. Values are means ± SD, n = 3. There are three samples whose error bars extend beyond the range of the y-axis (mean ± 1 SD): Fresno summer 2006 fine PM, 2.1 ± 0.9; Westside winter 2008 fine PM, 2.2 ± 1.1; Westside summer 2007 coarse PM, 2.6 ± 9.1.