

## **Supplemental Materials:**

### **Fine-particle air pollution and mortality: Importance of specific sources and chemical species**

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Supplemental Table 1. Summary of Pearson coefficients and COD<sup>\*</sup> values between monitoring sites during the 2008-2009 measurement campaign.

Pearson Coefficients between sites							
Site-to-Site		PM <sub>2.5</sub> mass	OC	EC	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>
Site 1	Site 2	0.85	0.92	0.82	0.81	0.91	0.88
Site 1	Site 3	0.93	0.90	0.83	0.92	0.94	0.95
Site 1	Site 4	0.93	0.94	0.90	0.93	0.95	0.95
Site 1	Site 5	0.93	0.92	0.81	0.90	0.95	0.95
Site 1	Site 6	0.90	0.92	0.85	0.94	0.94	0.95
Site 2	Site 3	0.86	0.92	0.80	0.73	0.94	0.89
Site 2	Site 4	0.80	0.92	0.80	0.81	0.90	0.87
Site 2	Site 5	0.83	0.92	0.82	0.82	0.96	0.95
Site 2	Site 6	0.87	0.93	0.79	0.83	0.92	0.91
Site 3	Site 4	0.91	0.88	0.87	0.86	0.94	0.93
Site 3	Site 5	0.96	0.93	0.88	0.95	0.96	0.96
Site 3	Site 6	0.91	0.97	0.93	0.91	0.96	0.96
Site 4	Site 5	0.90	0.92	0.84	0.87	0.94	0.93
Site 4	Site 6	0.91	0.92	0.86	0.92	0.95	0.95
Site 5	Site 6	0.90	0.93	0.85	0.93	0.95	0.97
COD values between sites							
Site-to-Site		PM <sub>2.5</sub> mass	OC	EC	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>
Site 1	Site 2	0.20	0.18	0.34	0.28	0.18	0.22
Site 1	Site 3	0.21	0.21	0.41	0.33	0.16	0.24
Site 1	Site 4	0.20	0.17	0.39	0.28	0.17	0.21
Site 1	Site 5	0.22	0.19	0.41	0.33	0.14	0.22
Site 1	Site 6	0.21	0.20	0.33	0.28	0.16	0.22
Site 2	Site 3	0.19	0.16	0.19	0.22	0.18	0.23
Site 2	Site 4	0.19	0.15	0.17	0.21	0.20	0.21
Site 2	Site 5	0.19	0.15	0.18	0.23	0.17	0.20
Site 2	Site 6	0.18	0.16	0.19	0.22	0.20	0.22
Site 3	Site 4	0.15	0.15	0.15	0.19	0.16	0.19
Site 3	Site 5	0.11	0.11	0.13	0.12	0.12	0.15
Site 3	Site 6	0.14	0.10	0.16	0.19	0.15	0.19
Site 4	Site 5	0.15	0.13	0.15	0.18	0.17	0.17
Site 4	Site 6	0.15	0.15	0.17	0.16	0.16	0.16
Site 5	Site 6	0.14	0.11	0.18	0.19	0.15	0.15

\*COD (Coefficient of Divergence): COD<sub>jk</sub> =  $\sqrt{\frac{1}{n} \sum_{i=1}^n \left( \frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2}$ , where 'x<sub>ij</sub>' is the chemical

concentrations per sampling interval (*i*) collected at site 'j', 'j' and 'k' represent two sampling sites, and 'n' is the number of sampling intervals. The COD provides a measure of the degree of uniformity among sampling sites, (i.e., the COD value is getting closer to zero when the average PM<sub>2.5</sub> mass and chemicals measured at two sites are uniform, but approaching to one when the measurements are dissimilar).

Supplemental Table 2. Summary statistics and mass concentrations ( $\mu\text{g}/\text{m}^3$ ) of  $\text{PM}_{2.5}$  and the 20 species used in the present study.

	Daily Mean <sup>b)</sup>	Standard Deviation	BDLs <sup>c)</sup> (%)
$\text{PM}_{2.5}$ mass	43.36	24.83	0.0
OC	9.94	5.00	0.0
EC	3.25	2.04	0.0
$\text{SO}_4^{2-}$	8.50	7.41	0.0
$\text{NO}_3^-$	7.21	5.82	0.0
$\text{NH}_4^+$	5.50	4.35	0.0
Na	0.15	0.10	4.1
Mg	0.06	0.07	12.9
Al	0.26	0.32	5.8
Si	0.70	0.85	0.0
Cl	0.45	0.64	7.5
K	0.40	0.31	0.0
Ca	0.19	0.19	1.0
Ti	0.02	0.03	13.7
Mn	0.02	0.02	5.2
Fe	0.36	0.31	0.0
Ni <sup>a)</sup>	1.91	2.28	53.9
Cu	0.02	0.01	2.3
Zn	0.11	0.08	0.0
Br	0.01	0.01	47.3
Pb	0.05	0.04	15.3

a) nanogram per cubic meter

b) data below the limit of detection were replaced by half of the reported detection limit values

c) Below detection limits

Supplemental Table 3. Correlation between daily concentrations of PM<sub>2.5</sub> mass and chemical species

	PM <sub>2.5</sub>	OC	EC	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	Na	Mg	Al	Si	Cl	K	Ca	Ti	Mn	Fe	Ni	Cu	Zn	Br	Pb
PM <sub>2.5</sub>	1.00																				
OC	0.64	1.00																			
EC	0.43	0.61	1.00																		
SO <sub>4</sub> <sup>2-</sup>	0.71	0.22	0.14	1.00																	
NO <sub>3</sub> <sup>-</sup>	0.79	0.55	0.40	0.58	1.00																
NH <sub>4</sub> <sup>+</sup>	0.69	0.24	0.18	0.81	0.57	1.00															
Na	0.48	0.32	0.17	0.25	0.33	0.22	1.00														
Mg	0.40	0.15	-0.01	0.14	0.17	0.06	0.63	1.00													
Al	0.47	0.20	0.05	0.18	0.24	0.10	0.55	0.88	1.00												
Si	0.47	0.18	0.04	0.18	0.23	0.11	0.54	0.89	0.99	1.00											
Cl	0.41	0.54	0.32	0.04	0.37	0.11	0.49	0.19	0.24	0.23	1.00										
K	0.71	0.48	0.25	0.38	0.47	0.32	0.70	0.69	0.69	0.70	0.46	1.00									
Ca	0.53	0.25	0.09	0.23	0.29	0.13	0.57	0.84	0.91	0.91	0.29	0.77	1.00								
Ti	0.49	0.23	0.09	0.20	0.26	0.12	0.50	0.81	0.93	0.94	0.22	0.69	0.87	1.00							
Mn	0.72	0.43	0.19	0.41	0.50	0.38	0.63	0.66	0.67	0.69	0.39	0.86	0.75	0.71	1.00						
Fe	0.62	0.31	0.17	0.29	0.37	0.23	0.56	0.83	0.92	0.93	0.28	0.78	0.89	0.95	0.82	1.00					
Ni	0.49	0.48	0.42	0.24	0.42	0.26	0.28	0.11	0.22	0.20	0.44	0.30	0.24	0.28	0.36	0.33	1.00				
Cu	0.62	0.44	0.39	0.37	0.50	0.37	0.36	0.26	0.31	0.31	0.37	0.54	0.43	0.43	0.66	0.53	0.49	1.00			
Zn	0.65	0.41	0.26	0.45	0.50	0.40	0.48	0.38	0.37	0.38	0.37	0.67	0.52	0.44	0.76	0.56	0.41	0.71	1.00		
Br	0.46	0.37	0.21	0.23	0.37	0.22	0.35	0.19	0.28	0.27	0.47	0.36	0.28	0.30	0.43	0.35	0.44	0.47	0.39	1.00	
Pb	0.73	0.55	0.34	0.45	0.55	0.43	0.55	0.42	0.44	0.45	0.51	0.79	0.52	0.48	0.79	0.59	0.42	0.65	0.70	0.48	1.00

Supplemental Table 4. Excess risk (95% confidence interval) per IQR [ $(\text{Exp}(\text{RR} \times \text{IQR}) - 1) \times 100$ ] in single-pollutant models.

Pollutant	Lag	Mortality		
		All cause	Cardiovascular	Respiratory
PM2.5 mass	0	0.54 (-0.77 to 1.86)	2.76 (0.23 to 5.36)	-0.24 (-5.65 to 5.48)
	1	0.52 (-0.71 to 1.76)	0.51 (-1.81 to 2.89)	-0.06 (-5.13 to 5.28)
	2	-0.71 (-1.88 to 0.47)	-1.05 (-3.27 to 1.21)	1.81 (-3.19 to 7.06)
	3	-0.76 (-1.90 to 0.39)	-0.71 (-2.88 to 1.52)	2.05 (-2.79 to 7.13)
OC	0	0.64 (-0.80 to 2.09)	2.32 (-0.45 to 5.16)	-0.92 (-6.82 to 5.34)
	1	0.87 (-0.60 to 2.36)	1.78 (-1.01 to 4.65)	2.92 (-3.26 to 9.51)
	2	-0.63 (-2.00 to 0.77)	-0.90 (-3.49 to 1.76)	0.96 (-4.83 to 7.11)
	3	-0.94 (-2.26 to 0.41)	-0.71 (-3.22 to 1.86)	3.16 (-2.59 to 9.24)
EC	0	0.54 (-0.68 to 1.78)	1.20 (-1.12 to 3.57)	-2.37 (-7.62 to 3.19)
	1	0.17 (-1.08 to 1.43)	-0.14 (-2.50 to 2.27)	4.59 (-0.79 to 10.25)
	2	0.25 (-0.96 to 1.47)	-1.08 (-3.35 to 1.24)	3.62 (-1.62 to 9.13)
	3	-0.65 (-1.82 to 0.54)	-1.08 (-3.29 to 1.18)	1.84 (-3.38 to 7.36)
$\text{SO}_4^{2-}$	0	0.26 (-0.74 to 1.28)	1.26 (-0.69 to 3.24)	0.62 (-3.59 to 5.01)
	1	-0.01 (-0.95 to 0.93)	-0.45 (-2.23 to 1.37)	-0.18 (-4.08 to 3.88)
	2	-0.70 (-1.61 to 0.22)	-1.48 (-3.22 to 0.28)	2.63 (-1.29 to 6.71)
	3	-0.37 (-1.28 to 0.55)	-0.84 (-2.59 to 0.94)	-0.55 (-4.37 to 3.43)
$\text{NO}_3^-$	0	0.28 (-0.84 to 1.41)	1.69 (-0.45 to 3.87)	1.09 (-3.63 to 6.04)
	1	-0.26 (-1.33 to 0.82)	-1.66 (-3.68 to 0.40)	-0.35 (-4.75 to 4.26)
	2	-0.39 (-1.43 to 0.65)	-0.23 (-2.19 to 1.76)	1.29 (-3.02 to 5.80)
	3	-0.85 (-1.87 to 0.18)	0.00 (-1.92 to 1.96)	1.94 (-2.36 to 6.44)
$\text{NH}_4^+$	0	0.67 (-0.58 to 1.94)	2.45 (0.01 to 4.95)	-0.19 (-5.35 to 5.25)
	1	-0.13 (-1.29 to 1.05)	0.07 (-2.14 to 2.33)	2.27 (-2.62 to 7.41)
	2	-0.99 (-2.12 to 0.14)	-1.92 (-4.04 to 0.25)	1.50 (-3.31 to 6.56)
	3	-1.06 (-2.18 to 0.07)	-0.61 (-2.75 to 1.57)	-3.14 (-7.73 to 1.69)
Na	0	0.30 (-0.80 to 1.42)	-0.30 (-2.41 to 1.85)	-1.34 (-5.89 to 3.44)
	1	0.25 (-0.85 to 1.36)	0.51 (-1.58 to 2.65)	-0.31 (-4.80 to 4.40)
	2	-0.71 (-1.79 to 0.38)	-0.70 (-2.76 to 1.40)	4.99 (0.41 to 9.77)
	3	1.00 (-0.09 to 2.10)	1.64 (-0.45 to 3.77)	0.64 (-3.88 to 5.37)
Mg	0	0.39 (-0.29 to 1.07)	0.21 (-1.10 to 1.53)	-1.72 (-4.65 to 1.31)
	1	0.00 (-0.68 to 0.67)	-0.05 (-1.34 to 1.25)	-0.78 (-3.53 to 2.06)
	2	-0.09 (-0.76 to 0.59)	0.62 (-0.65 to 1.90)	1.71 (-1.01 to 4.51)
	3	0.16 (-0.51 to 0.84)	-0.57 (-1.88 to 0.76)	1.45 (-1.26 to 4.24)
Al	0	0.21 (-0.42 to 0.84)	0.01 (-1.20 to 1.23)	-1.95 (-4.66 to 0.85)
	1	0.17 (-0.44 to 0.79)	0.49 (-0.68 to 1.68)	-2.39 (-5.04 to 0.33)
	2	-0.33 (-0.95 to 0.29)	-0.24 (-1.41 to 0.95)	1.62 (-0.85 to 4.15)
	3	0.02 (-0.60 to 0.64)	-0.77 (-1.98 to 0.44)	2.12 (-0.30 to 4.59)
Si	0	0.18 (-0.44 to 0.80)	-0.02 (-1.21 to 1.19)	-1.78 (-4.45 to 0.97)
	1	0.13 (-0.49 to 0.74)	0.42 (-0.74 to 1.60)	-2.19 (-4.80 to 0.49)
	2	-0.29 (-0.90 to 0.33)	-0.22 (-1.38 to 0.95)	1.59 (-0.86 to 4.09)
	3	0.07 (-0.54 to 0.69)	-0.69 (-1.88 to 0.51)	2.16 (-0.22 to 4.60)

Cl	0	0.12 (-0.98 to 1.24)	1.28 (-0.81 to 3.41)	-2.12 (-6.67 to 2.64)
	1	0.14 (-0.99 to 1.28)	-0.63 (-2.77 to 1.56)	1.83 (-2.89 to 6.79)
	2	-0.26 (-1.36 to 0.85)	0.91 (-1.20 to 3.05)	1.46 (-3.03 to 6.16)
	3	0.29 (-0.81 to 1.39)	1.44 (-0.62 to 3.55)	-1.12 (-5.74 to 3.73)
K	0	0.79 (-0.36 to 1.95)	1.75 (-0.46 to 4.02)	-1.15 (-5.92 to 3.87)
	1	0.34 (-0.78 to 1.48)	0.42 (-1.72 to 2.61)	-3.40 (-7.99 to 1.42)
	2	-0.44 (-1.55 to 0.67)	0.04 (-2.07 to 2.19)	4.19 (-0.47 to 9.07)
	3	0.49 (-0.61 to 1.61)	0.86 (-1.26 to 3.03)	2.57 (-2.08 to 7.43)
Ca	0	0.57 (-0.20 to 1.35)	0.59 (-0.88 to 2.08)	-3.70 (-6.95 to -0.34)
	1	0.41 (-0.34 to 1.17)	0.38 (-1.06 to 1.84)	-2.80 (-5.93 to 0.43)
	2	-0.14 (-0.89 to 0.62)	0.11 (-1.31 to 1.55)	2.27 (-0.77 to 5.41)
	3	0.35 (-0.40 to 1.11)	-0.19 (-1.63 to 1.27)	2.35 (-0.66 to 5.45)
Ti	0	0.14 (-0.55 to 0.83)	0.02 (-1.30 to 1.36)	-2.62 (-5.64 to 0.50)
	1	-0.12 (-0.80 to 0.57)	0.47 (-0.82 to 1.78)	-2.52 (-5.47 to 0.53)
	2	-0.29 (-0.97 to 0.39)	-0.12 (-1.40 to 1.17)	1.52 (-1.21 to 4.32)
	3	-0.12 (-0.80 to 0.56)	-0.99 (-2.31 to 0.35)	2.17 (-0.48 to 4.90)
Mn	0	0.55 (-0.58 to 1.68)	1.16 (-0.99 to 3.36)	-1.91 (-6.55 to 2.96)
	1	0.37 (-0.72 to 1.48)	0.30 (-1.78 to 2.43)	-1.62 (-6.14 to 3.13)
	2	-0.16 (-1.23 to 0.92)	0.25 (-1.78 to 2.33)	3.09 (-1.39 to 7.78)
	3	0.11 (-0.97 to 1.20)	0.86 (-1.21 to 2.97)	3.00 (-1.48 to 7.68)
Fe	0	0.32 (-0.50 to 1.14)	0.69 (-0.86 to 2.26)	-3.08 (-6.63 to 0.60)
	1	0.11 (-0.69 to 0.93)	0.30 (-1.23 to 1.86)	-2.34 (-5.75 to 1.20)
	2	-0.16 (-0.96 to 0.64)	-0.05 (-1.56 to 1.47)	2.21 (-1.01 to 5.53)
	3	-0.06 (-0.85 to 0.74)	-0.76 (-2.30 to 0.79)	2.71 (-0.44 to 5.96)
Ni	0	-0.56 (-2.01 to 0.90)	2.50 (-0.30 to 5.38)	-3.18 (-8.98 to 3.00)
	1	0.49 (-0.99 to 1.99)	-0.34 (-3.13 to 2.52)	0.86 (-5.24 to 7.35)
	2	-0.18 (-1.64 to 1.31)	-1.11 (-3.86 to 1.72)	4.60 (-1.65 to 11.24)
	3	-0.57 (-1.98 to 0.87)	-0.79 (-3.46 to 1.96)	-4.02 (-9.72 to 2.04)
Cu	0	0.24 (-0.90 to 1.41)	2.30 (0.08 to 4.58)	-3.81 (-8.52 to 1.15)
	1	0.37 (-0.80 to 1.55)	-0.52 (-2.71 to 1.71)	0.71 (-4.16 to 5.83)
	2	-0.01 (-1.12 to 1.11)	-0.01 (-2.13 to 2.15)	-0.35 (-5.00 to 4.52)
	3	0.06 (-1.04 to 1.18)	0.59 (-1.54 to 2.76)	0.99 (-3.68 to 5.90)
Zn	0	0.49 (-0.53 to 1.51)	1.97 (0.01 to 3.96)	-3.00 (-7.20 to 1.39)
	1	0.52 (-0.48 to 1.53)	0.42 (-1.49 to 2.37)	-0.13 (-4.29 to 4.20)
	2	-0.10 (-1.07 to 0.89)	-0.40 (-2.27 to 1.49)	1.36 (-2.68 to 5.58)
	3	0.27 (-0.72 to 1.26)	1.49 (-0.39 to 3.41)	1.42 (-2.60 to 5.61)
Br	0	0.12 (-0.88 to 1.13)	1.13 (-0.75 to 3.04)	1.15 (-2.93 to 5.40)
	1	0.12 (-0.89 to 1.14)	0.05 (-1.89 to 2.04)	1.20 (-2.90 to 5.47)
	2	-0.30 (-1.29 to 0.71)	-0.93 (-2.83 to 1.01)	3.99 (0.03 to 8.11)
	3	0.57 (-0.42 to 1.56)	1.06 (-0.81 to 2.96)	1.14 (-2.94 to 5.40)
Pb	0	0.47 (-0.66 to 1.61)	2.08 (-0.11 to 4.32)	-0.54 (-5.22 to 4.36)
	1	0.07 (-1.03 to 1.17)	-0.15 (-2.23 to 1.98)	-1.06 (-5.57 to 3.68)
	2	0.11 (-0.97 to 1.19)	1.90 (-0.16 to 4.01)	3.34 (-1.21 to 8.10)
	3	0.21 (-0.86 to 1.30)	1.51 (-0.56 to 3.64)	4.07 (-0.47 to 8.83)

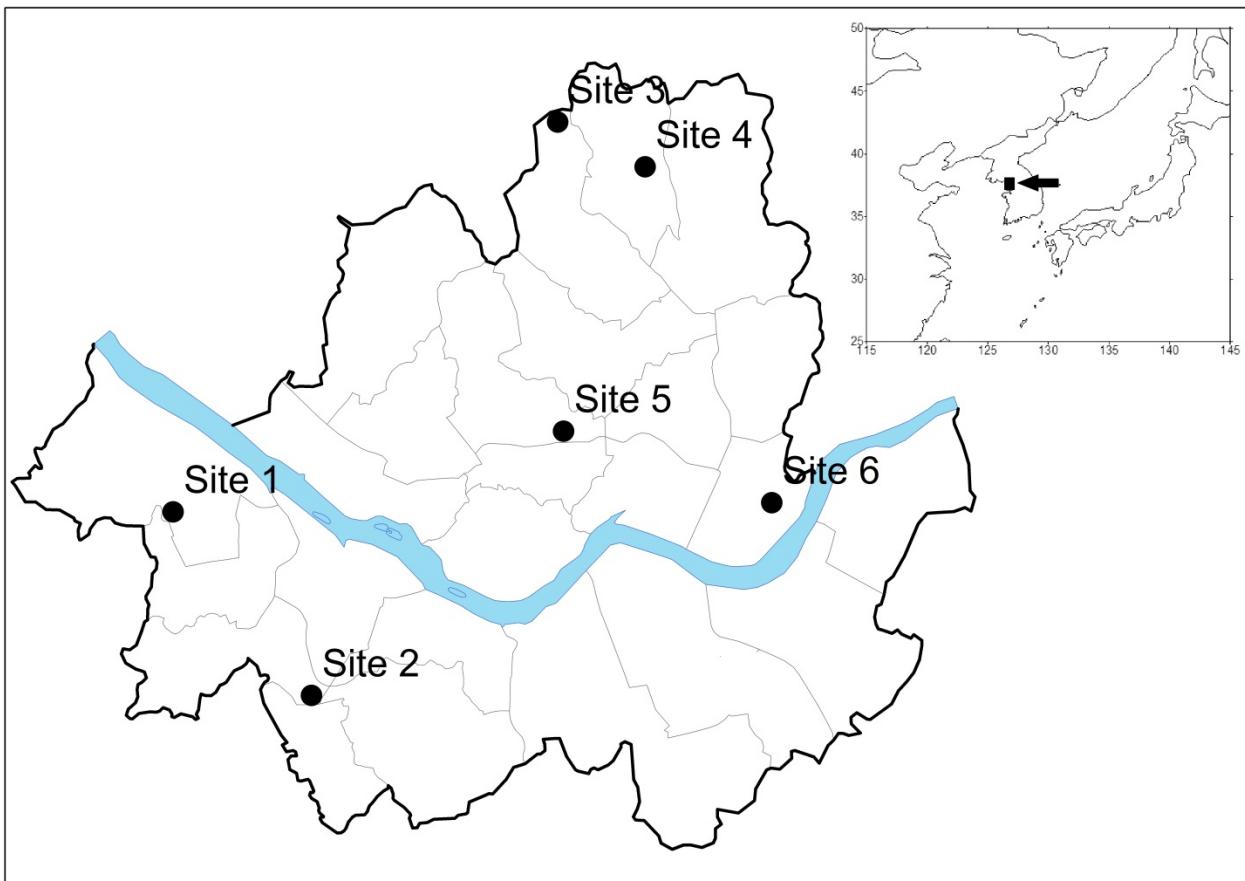
Supplemental Table 5. Excess risk (95% confidence interval) per IQR [ $(\text{Exp}(\text{RR} \times \text{IQR}) - 1) \times 100$ ] in multi-pollutant models.

Pollutant	lag	Mortality		
		All cause	Cardiovascular	Respiratory
OC	0	0.74 (-1.61 to 3.15)	1.83 (-2.63 to 6.50)	-1.38 (-10.70 to 8.91)
	1	2.57 (0.13 to 5.08)	6.94 (2.17 to 11.94)	-0.71 (-10.42 to 10.04)
	2	-1.74 (-4.04 to 0.61)	-1.24 (-5.58 to 3.30)	-8.02 (-16.76 to 1.65)
	3	-0.68 (-2.98 to 1.67)	-0.92 (-5.22 to 3.58)	3.30 (-6.53 to 14.16)
EC	0	0.25 (-1.68 to 2.21)	-2.00 (-5.54 to 1.67)	-0.20 (-8.44 to 8.77)
	1	-0.93 (-2.84 to 1.03)	-1.91 (-5.49 to 1.81)	7.42 (-1.13 to 16.72)
	2	1.44 (-0.53 to 3.45)	-0.96 (-4.58 to 2.81)	9.52 (0.83 to 18.96)
	3	-0.05 (-1.99 to 1.92)	-1.33 (-4.92 to 2.40)	1.87 (-6.53 to 11.01)
$\text{SO}_4^{2-}$	0	-0.04 (-1.39 to 1.33)	0.01 (-2.57 to 2.66)	-0.36 (-6.01 to 5.62)
	1	-0.10 (-1.42 to 1.22)	-0.08 (-2.56 to 2.46)	2.61 (-2.93 to 8.46)
	2	-0.86 (-2.16 to 0.45)	-2.56 (-5.01 to -0.05)	0.86 (-4.57 to 6.61)
	3	-0.39 (-1.71 to 0.94)	-2.76 (-5.25 to -0.20)	-4.81 (-10.09 to 0.78)
$\text{NO}_3^-$	0	-0.08 (-1.74 to 1.61)	-0.01 (-3.14 to 3.21)	3.65 (-3.52 to 11.35)
	1	-1.39 (-3.00 to 0.26)	-4.24 (-7.22 to -1.16)	-2.70 (-9.18 to 4.24)
	2	0.17 (-1.47 to 1.83)	1.86 (-1.28 to 5.11)	-0.94 (-7.53 to 6.13)
	3	-0.82 (-2.45 to 0.84)	0.90 (-2.22 to 4.12)	3.85 (-3.10 to 11.31)
$\text{NH}_4^+$	0	0.64 (-0.86 to 2.15)	1.35 (-1.51 to 4.30)	-0.68 (-6.72 to 5.75)
	1	-0.78 (-2.20 to 0.66)	-0.70 (-3.38 to 2.05)	3.48 (-2.58 to 9.92)
	2	-1.26 (-2.67 to 0.17)	-2.41 (-5.06 to 0.31)	-0.89 (-6.73 to 5.31)
	3	-1.51 (-2.92 to -0.08)	-2.00 (-4.67 to 0.74)	-7.17 (-12.63 to -1.38)
Na	0	-0.84 (-2.63 to 0.99)	-3.17 (-6.50 to 0.27)	-0.50 (-7.98 to 7.60)
	1	0.34 (-1.47 to 2.19)	1.61 (-1.85 to 5.20)	0.32 (-7.11 to 8.36)
	2	-0.68 (-2.45 to 1.13)	-3.26 (-6.52 to 0.12)	5.94 (-1.69 to 14.18)
	3	1.54 (-0.28 to 3.39)	2.68 (-0.80 to 6.29)	0.81 (-6.65 to 8.87)
Mg	0	0.40 (-0.72 to 1.52)	0.82 (-1.32 to 3.00)	-3.09 (-7.80 to 1.87)
	1	-0.53 (-1.64 to 0.60)	-1.25 (-3.35 to 0.90)	2.22 (-2.50 to 7.17)
	2	0.32 (-0.79 to 1.45)	2.29 (0.18 to 4.44)	-1.57 (-6.04 to 3.11)
	3	-0.71 (-1.83 to 0.42)	-2.75 (-4.89 to -0.56)	1.34 (-3.27 to 6.17)
Al	0	-0.02 (-0.98 to 0.96)	-0.34 (-2.21 to 1.56)	-3.51 (-7.50 to 0.66)
	1	0.04 (-0.92 to 1.01)	0.70 (-1.14 to 2.58)	-2.08 (-6.12 to 2.13)
	2	-0.40 (-1.36 to 0.58)	-0.04 (-1.87 to 1.82)	-0.89 (-4.72 to 3.09)
	3	-0.62 (-1.58 to 0.35)	-2.26 (-4.09 to -0.39)	3.29 (-0.66 to 7.40)
Si	0	-0.11 (-1.08 to 0.87)	-0.48 (-2.35 to 1.43)	-3.12 (-7.14 to 1.07)
	1	-0.10 (-1.07 to 0.88)	0.44 (-1.41 to 2.33)	-1.63 (-5.71 to 2.62)
	2	-0.33 (-1.30 to 0.65)	-0.10 (-1.94 to 1.77)	-0.83 (-4.67 to 3.17)
	3	-0.50 (-1.47 to 0.48)	-2.15 (-3.99 to -0.27)	3.45 (-0.53 to 7.58)
Cl	0	-0.11 (-1.65 to 1.46)	-0.16 (-3.07 to 2.83)	-0.93 (-7.28 to 5.86)
	1	-0.53 (-2.06 to 1.03)	-1.66 (-4.55 to 1.31)	3.15 (-3.41 to 10.15)
	2	-0.15 (-1.69 to 1.40)	2.68 (-0.29 to 5.75)	-2.18 (-8.22 to 4.26)
	3	-0.09 (-1.61 to 1.46)	0.61 (-2.26 to 3.57)	-3.04 (-9.39 to 3.75)

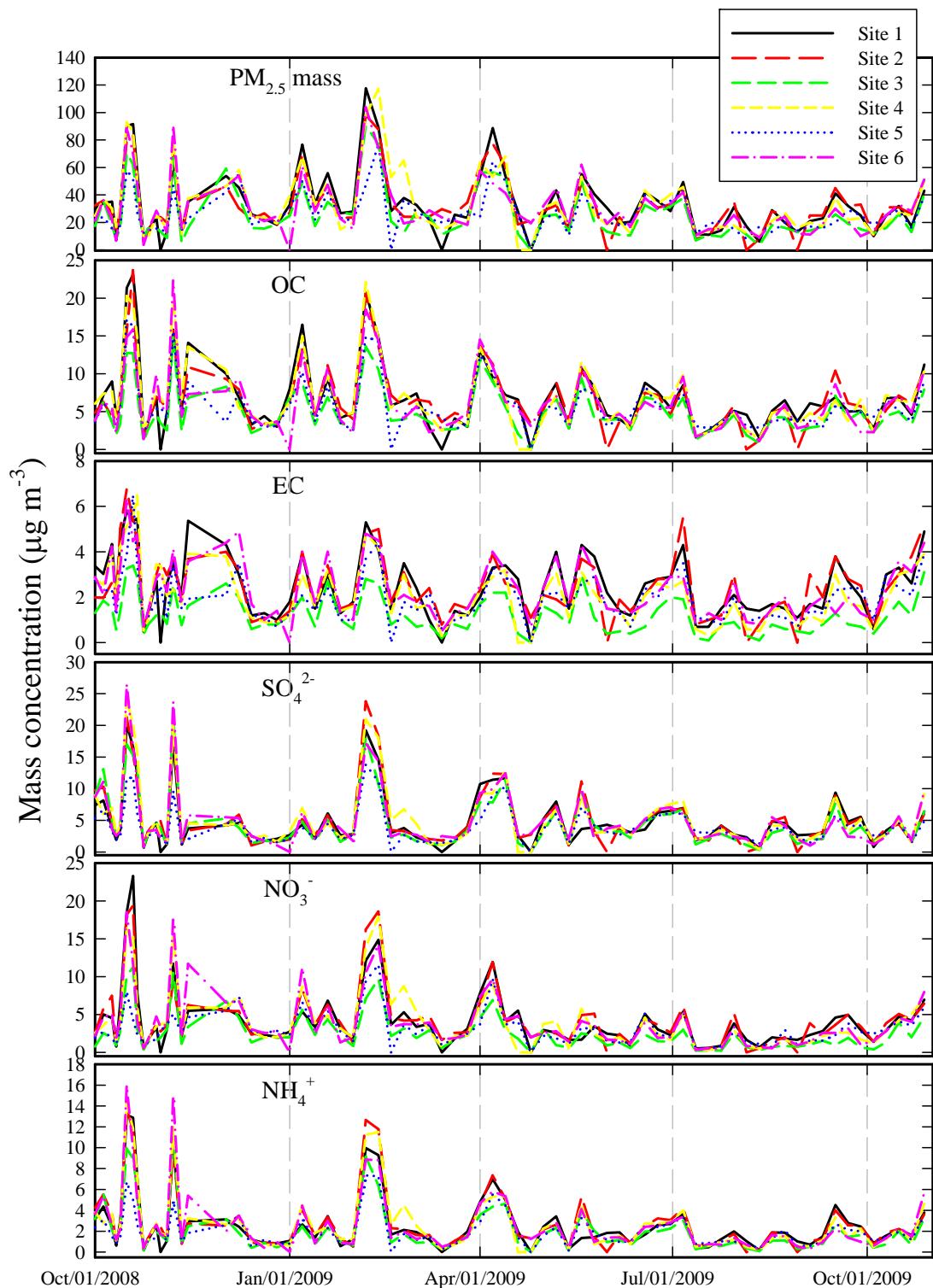
K	0	0.90 (-1.96 to 3.84)	3.80 (-1.74 to 9.66)	7.59 (-4.49 to 21.21)
	1	0.20 (-2.63 to 3.10)	1.15 (-4.26 to 6.85)	-9.86 (-20.19 to 1.80)
	2	-1.63 (-4.44 to 1.27)	-3.59 (-8.78 to 1.90)	-0.02 (-11.34 to 12.75)
	3	0.84 (-2.03 to 3.79)	0.63 (-4.74 to 6.29)	-6.98 (-17.85 to 5.33)
Ca	0	0.76 (-0.54 to 2.08)	0.45 (-2.05 to 3.02)	-7.39 (-12.58 to -1.88)
	1	0.45 (-0.84 to 1.76)	0.40 (-2.07 to 2.94)	-2.88 (-8.18 to 2.72)
	2	0.09 (-1.19 to 1.39)	0.99 (-1.46 to 3.50)	-0.03 (-5.14 to 5.36)
	3	-0.10 (-1.38 to 1.20)	-1.61 (-4.05 to 0.89)	3.44 (-1.83 to 8.99)
Ti	0	-0.19 (-1.23 to 0.86)	-0.76 (-2.75 to 1.26)	-3.77 (-8.04 to 0.71)
	1	-0.75 (-1.79 to 0.30)	0.59 (-1.39 to 2.61)	-2.27 (-6.69 to 2.36)
	2	-0.43 (-1.48 to 0.63)	0.00 (-1.97 to 2.01)	-0.88 (-5.06 to 3.49)
	3	-0.86 (-1.91 to 0.20)	-2.69 (-4.68 to -0.66)	2.93 (-1.33 to 7.38)
Mn	0	-0.46 (-3.29 to 2.46)	-4.52 (-9.67 to 0.92)	4.88 (-6.82 to 18.05)
	1	0.38 (-2.49 to 3.35)	0.29 (-5.13 to 6.02)	4.05 (-7.81 to 17.42)
	2	-0.15 (-3.02 to 2.80)	-0.15 (-5.54 to 5.55)	0.16 (-11.16 to 12.92)
	3	-0.77 (-3.65 to 2.19)	1.43 (-4.07 to 7.25)	-0.27 (-11.68 to 12.61)
Fe	0	-0.07 (-1.58 to 1.46)	0.01 (-2.89 to 2.99)	-6.05 (-12.18 to 0.51)
	1	-0.52 (-2.02 to 1.00)	0.34 (-2.50 to 3.28)	-2.37 (-8.73 to 4.44)
	2	-0.04 (-1.56 to 1.50)	0.44 (-2.41 to 3.38)	-0.87 (-6.88 to 5.53)
	3	-1.03 (-2.55 to 0.51)	-3.81 (-6.67 to -0.86)	5.08 (-1.18 to 11.72)
Ni	0	-1.16 (-2.99 to 0.70)	1.23 (-2.29 to 4.88)	-0.52 (-8.03 to 7.59)
	1	0.48 (-1.37 to 2.36)	-0.34 (-3.81 to 3.25)	-0.66 (-8.18 to 7.48)
	2	0.23 (-1.66 to 2.15)	-0.80 (-4.35 to 2.88)	5.76 (-2.22 to 14.40)
	3	-0.30 (-2.16 to 1.58)	-0.99 (-4.46 to 2.60)	-7.80 (-14.73 to -0.29)
Cu	0	-0.47 (-2.62 to 1.73)	0.88 (-3.23 to 5.17)	-2.63 (-11.48 to 7.11)
	1	0.47 (-1.69 to 2.67)	-1.38 (-5.36 to 2.77)	-0.19 (-8.96 to 9.43)
	2	-0.75 (-2.87 to 1.42)	-1.44 (-5.43 to 2.73)	-10.92 (-18.96 to -2.09)
	3	0.32 (-1.83 to 2.53)	-1.65 (-5.69 to 2.56)	-2.33 (-11.00 to 7.17)
Zn	0	0.14 (-1.63 to 1.94)	0.72 (-2.67 to 4.22)	-2.55 (-9.79 to 5.28)
	1	0.71 (-1.05 to 2.51)	2.57 (-0.81 to 6.07)	0.93 (-6.45 to 8.90)
	2	-0.53 (-2.27 to 1.23)	-2.83 (-6.11 to 0.56)	-0.88 (-7.97 to 6.76)
	3	0.15 (-1.63 to 1.95)	1.65 (-1.76 to 5.18)	0.66 (-6.28 to 8.12)
Br	0	0.32 (-0.88 to 1.53)	0.50 (-1.76 to 2.82)	3.07 (-1.60 to 7.95)
	1	-0.19 (-1.39 to 1.04)	-0.05 (-2.37 to 2.33)	1.26 (-3.55 to 6.32)
	2	-0.15 (-1.36 to 1.07)	-1.35 (-3.66 to 1.03)	4.39 (-0.45 to 9.46)
	3	1.00 (-0.20 to 2.21)	1.08 (-1.16 to 3.37)	0.98 (-3.94 to 6.15)
Pb	0	-0.09 (-2.35 to 2.23)	0.60 (-3.73 to 5.13)	0.50 (-8.67 to 10.59)
	1	-0.90 (-3.17 to 1.42)	-1.04 (-5.30 to 3.43)	-0.43 (-9.60 to 9.67)
	2	2.14 (-0.19 to 4.53)	9.37 (4.66 to 14.29)	5.51 (-4.07 to 16.05)
	3	0.14 (-2.12 to 2.46)	2.30 (-2.07 to 6.87)	10.06 (0.05 to 21.06)

Supplemental Table 6. Excess risk (95% confidence interval) of each source contribution per IQR [(Exp(RR×IQR)-1)\*100

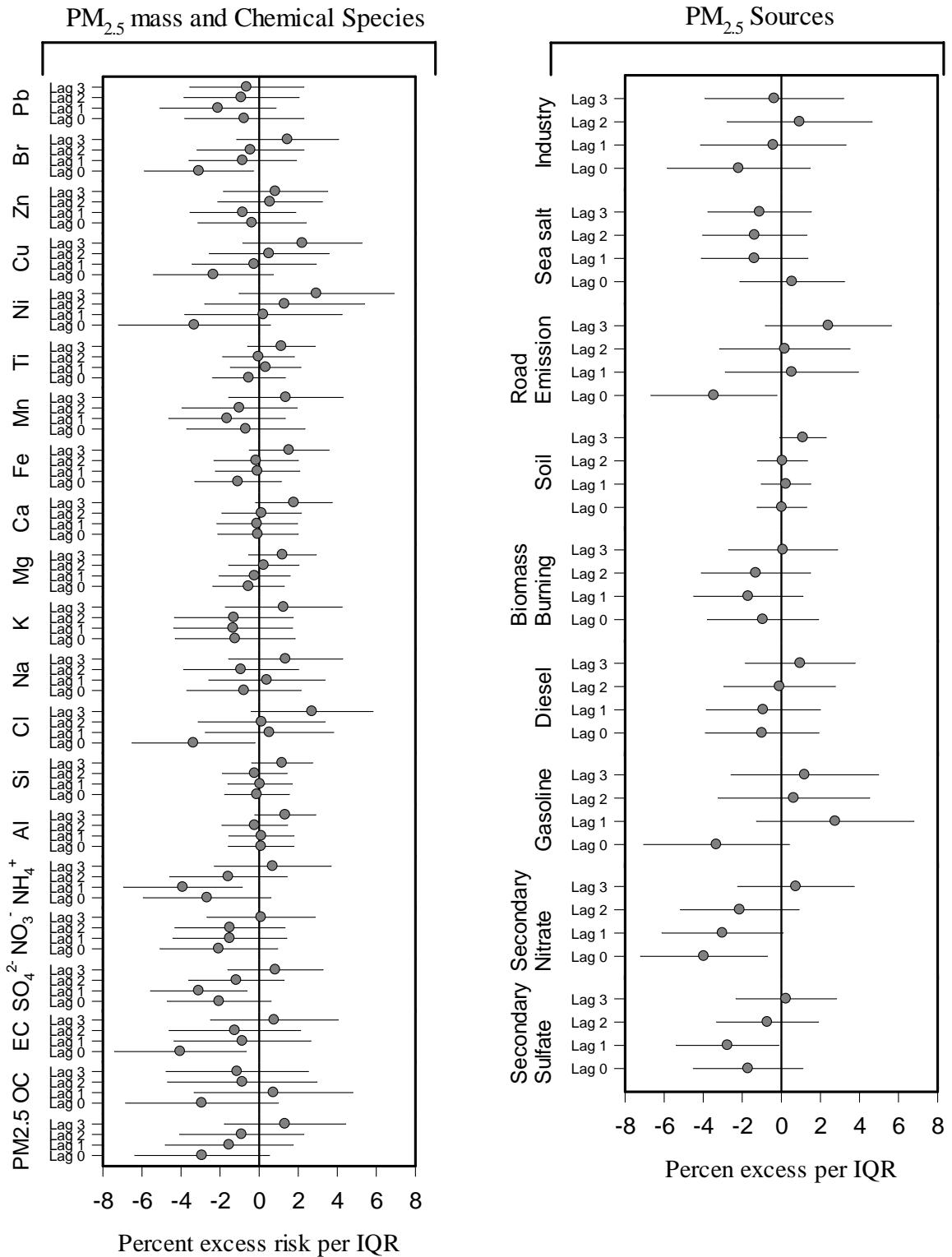
PM2.5 Sources	Lag day	Mortality		
		All cause	Cardiovascular	Respiratory
Secondary Nitrate	0	0.56 (-0.52 to 1.64)	1.38 (-0.67 to 3.47)	-0.52 (-5.04 to 4.22)
	1	0.06 (-0.97 to 1.10)	-0.74 (-2.68 to 1.25)	0.07 (-4.23 to 4.58)
	2	-0.13 (-1.14 to 0.89)	-0.82 (-2.72 to 1.12)	1.88 (-2.31 to 6.26)
	3	-1.14 (-2.13 to -0.13)	-0.74 (-2.62 to 1.16)	0.43 (-3.83 to 4.87)
Secondary Sulfate	0	0.45 (-0.53 to 1.44)	0.98 (-0.93 to 2.92)	1.18 (-2.93 to 5.47)
	1	-0.06 (-0.97 to 0.85)	-0.26 (-1.99 to 1.50)	-0.27 (-4.05 to 3.65)
	2	-0.74 (-1.63 to 0.16)	-1.72 (-3.41 to 0.01)	2.12 (-1.71 to 6.10)
	3	-0.57 (-1.46 to 0.33)	-1.45 (-3.16 to 0.30)	-0.13 (-3.86 to 3.76)
Gasoline	0	0.36 (-0.83 to 1.56)	0.41 (-1.86 to 2.72)	5.45 (0.49 to 10.66)
	1	0.48 (-0.69 to 1.67)	0.80 (-1.43 to 3.09)	-4.20 (-9.00 to 0.86)
	2	-0.61 (-1.79 to 0.58)	-1.02 (-3.23 to 1.25)	1.50 (-3.47 to 6.72)
	3	0.36 (-0.81 to 1.55)	0.45 (-1.77 to 2.73)	0.48 (-4.42 to 5.62)
Diesel	0	0.57 (-0.88 to 2.04)	0.90 (-1.85 to 3.73)	-3.42 (-9.32 to 2.86)
	1	0.28 (-1.20 to 1.79)	-0.02 (-2.82 to 2.87)	6.74 (0.23 to 13.67)
	2	0.42 (-1.04 to 1.90)	-1.66 (-4.38 to 1.13)	1.84 (-4.33 to 8.40)
	3	-0.93 (-2.37 to 0.53)	-0.74 (-3.47 to 2.06)	0.20 (-6.00 to 6.81)
Biomass	0	0.58 (-0.37 to 1.54)	1.86 (0.01 to 3.74)	-1.38 (-5.39 to 2.80)
	1	0.24 (-0.69 to 1.18)	-0.30 (-2.08 to 1.52)	-0.19 (-4.03 to 3.80)
	2	0.17 (-0.74 to 1.10)	1.03 (-0.73 to 2.82)	2.72 (-1.08 to 6.66)
	3	0.27 (-0.65 to 1.20)	1.67 (-0.12 to 3.49)	2.56 (-1.27 to 6.54)
Soil	0	0.14 (-0.35 to 0.62)	-0.04 (-0.97 to 0.91)	-2.00 (-4.16 to 0.22)
	1	0.08 (-0.40 to 0.56)	0.34 (-0.56 to 1.25)	-1.84 (-3.93 to 0.28)
	2	-0.15 (-0.63 to 0.33)	-0.06 (-0.96 to 0.86)	0.93 (-0.99 to 2.88)
	3	-0.01 (-0.49 to 0.47)	-0.75 (-1.70 to 0.20)	1.57 (-0.27 to 3.45)
Roadway emission	0	-0.01 (-1.17 to 1.16)	1.30 (-0.91 to 3.55)	-1.83 (-6.61 to 3.20)
	1	0.17 (-1.02 to 1.38)	-0.48 (-2.73 to 1.83)	2.20 (-2.81 to 7.48)
	2	-0.42 (-1.58 to 0.75)	0.65 (-1.56 to 2.91)	1.67 (-3.13 to 6.71)
	3	0.46 (-0.69 to 1.62)	1.88 (-0.29 to 4.10)	-1.28 (-6.11 to 3.81)
Aged sea salt	0	0.57 (-0.45 to 1.60)	-0.49 (-2.43 to 1.48)	-0.60 (-4.79 to 3.76)
	1	0.18 (-0.83 to 1.21)	0.16 (-1.77 to 2.13)	-0.60 (-4.74 to 3.71)
	2	-0.62 (-1.64 to 0.40)	-0.42 (-2.34 to 1.52)	2.25 (-1.91 to 6.59)
	3	0.40 (-0.62 to 1.42)	0.31 (-1.61 to 2.28)	0.04 (-4.13 to 4.39)
Industry	0	-0.24 (-1.42 to 0.95)	2.13 (-0.16 to 4.48)	-3.20 (-7.99 to 1.84)
	1	-0.32 (-1.52 to 0.90)	-0.61 (-2.88 to 1.72)	1.23 (-3.80 to 6.51)
	2	0.18 (-0.99 to 1.37)	0.04 (-2.19 to 2.33)	0.89 (-4.00 to 6.04)
	3	0.24 (-0.90 to 1.40)	0.33 (-1.84 to 2.55)	-0.73 (-5.42 to 4.19)



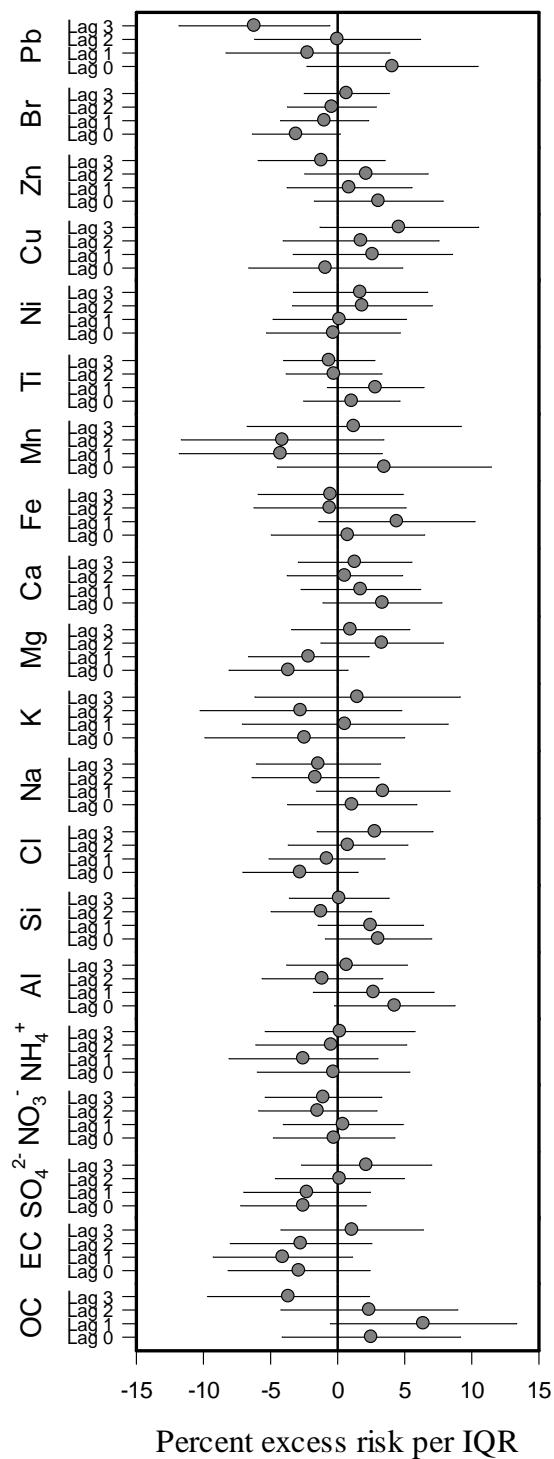
Supplemental Figure 1. Map of six urban sampling locations for the 2008-2009 measurement campaign of ambient PM<sub>2.5</sub> in Seoul. The integrated 24-hour PM<sub>2.5</sub> samples were simultaneously collected at each site from August 2008 to October 2009, with a 1-in-3 day sampling schedule for the 2008 calendar year and a 1-in-6 day sampling schedule for the 2009 calendar year. Total collected samples were about 50 to 60 at each sampling site. PM<sub>2.5</sub> total mass and five major chemical species, including OC, EC, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>, were determined by the same analytical methods as described in the main manuscript.



Supplemental Figure 2. Spatial and temporal trends in concentrations of PM<sub>2.5</sub> mass and major chemical species across the sites during the 2008-2009 measurement campaign.



Supplemental Figure 3. Percent excess risk (95% CI) in mortality for injury related events per IQR increase in PM<sub>2.5</sub> mass and chemical constituents as well as source apportionments in Seoul during 2003-2007 (single-pollutant model results).



Supplemental Figure 4. Percent excess risk (95% CI) in mortality for injury related events per IQR increase in PM2.5 chemical constituents in Seoul during 2003-2007 (multi-pollutant model results).