

Supplementary Information for:

Assessing PM_{2.5} Model Performance for the Conterminous U.S. with Comparison to Model Performance Statistics from 2007-2015

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Table S1. Median (range) of performance statistics for PM_{2.5} at AQS (FRM/FEM) sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	3.47 (2.41 to 9.06)	36.2 (22.5 to 79.3)	8.46 (6.43 to 14.57)	51.2 (42.1 to 87.7)	0.68 (0.45 to 0.74)
	Spring	1.21 (-0.48 to 2.72)	14.4 (-5.2 to 34.9)	5.19 (3.99 to 9.46)	44.0 (34.7 to 62.5)	0.59 (0.46 to 0.68)
	Summer	-2.12 (-4.82 to 1.25)	-18.2 (-35.4 to 15.0)	6.07 (4.06 to 8.39)	36.9 (35.2 to 54.6)	0.66 (0.22 to 0.71)
	Fall	1.90 (0.38 to 3.34)	26.5 (4.9 to 43.3)	6.07 (4.34 to 10.20)	45.2 (39.1 to 69.2)	0.70 (0.37 to 0.73)
	All	1.26 (-0.29 to 3.09)	15.0 (-3.1 to 36.3)	6.94 (5.45 to 10.86)	44.2 (40.3 to 64.6)	0.63 (0.42 to 0.67)
SE	Winter	2.06 (-0.07 to 3.54)	24.1 (-0.8 to 38.9)	6.07 (4.85 to 7.41)	47.4 (34.7 to 54.4)	0.58 (0.53 to 0.61)
	Spring	-1.31 (-3.41 to 0.78)	-13.9 (-30.6 to 8.7)	4.67 (4.08 to 9.57)	34.2 (31.7 to 42.3)	0.59 (0.48 to 0.68)
	Summer	-3.70 (-6.64 to 0.25)	-28.3 (-52.4 to 2.6)	6.06 (4.14 to 9.27)	37.1 (35.2 to 53.1)	0.60 (0.47 to 0.68)
	Fall	0.53 (-2.35 to 1.78)	6.2 (-28.0 to 20.4)	4.32 (4.04 to 7.02)	36.5 (29.9 to 42.1)	0.65 (0.57 to 0.70)
	All	-0.44 (-2.87 to 1.15)	-5.2 (-29.7 to 13.1)	5.39 (4.51 to 8.40)	39.3 (35.8 to 43.9)	0.52 (0.46 to 0.61)
Ohio Valley	Winter	1.81 (0.84 to 4.55)	17.1 (7.9 to 37.7)	5.75 (5.32 to 8.29)	38.4 (33.2 to 50.4)	0.66 (0.52 to 0.70)
	Spring	-0.14 (-1.21 to 1.21)	-1.0 (-11.9 to 11.0)	5.04 (4.07 to 7.13)	35.9 (32.8 to 39.6)	0.60 (0.47 to 0.65)
	Summer	-2.23 (-3.87 to -0.24)	-15.5 (-29.2 to -2.0)	5.63 (4.30 to 7.24)	32.1 (29.3 to 37.5)	0.59 (0.55 to 0.63)
	Fall	1.61 (0.12 to 3.84)	15.5 (1.2 to 37.4)	6.23 (4.56 to 7.90)	38.9 (33.2 to 50.9)	0.67 (0.63 to 0.76)
	All	0.53 (-0.90 to 1.29)	3.8 (-8.0 to 13.6)	6.00 (4.61 to 6.82)	36.2 (33.4 to 40.6)	0.59 (0.55 to 0.64)
MW	Winter	1.18 (-0.56 to 4.79)	10.5 (-4.3 to 49.1)	6.14 (4.75 to 8.31)	38.3 (29.8 to 61.7)	0.70 (0.61 to 0.75)
	Spring	0.23 (-1.15 to 1.70)	2.4 (-12.3 to 19.8)	5.12 (4.10 to 5.58)	40.2 (32.6 to 43.6)	0.65 (0.48 to 0.73)
	Summer	-1.35 (-3.36 to 0.22)	-15.3 (-37.8 to 2.8)	4.54 (4.05 to 5.61)	37.5 (33.5 to 43.6)	0.60 (0.56 to 0.73)
	Fall	1.40 (-0.36 to 3.98)	20.2 (-3.7 to 48.5)	4.92 (4.12 to 7.16)	42.1 (34.3 to 58.3)	0.75 (0.64 to 0.82)
	All	0.51 (-1.10 to 2.17)	6.2 (-11.4 to 25.1)	5.16 (4.82 to 6.36)	40.2 (35.4 to 48.9)	0.64 (0.63 to 0.74)
South	Winter	1.84 (-0.14 to 4.10)	23.0 (-1.7 to 45.8)	7.32 (5.10 to 13.03)	54.3 (40.6 to 69.0)	0.43 (0.30 to 0.49)
	Spring	-1.61 (-4.75 to -0.74)	-16.3 (-44.4 to -7.6)	6.02 (4.24 to 7.46)	42.9 (35.8 to 52.6)	0.37 (0.19 to 0.51)
	Summer	-2.89 (-5.93 to -0.76)	-24.4 (-52.6 to -6.7)	6.39 (5.59 to 7.78)	44.0 (38.0 to 54.9)	0.42 (0.30 to 0.58)
	Fall	1.21 (-1.70 to 3.09)	15.9 (-19.6 to 28.2)	5.68 (3.76 to 10.29)	44.2 (35.5 to 52.4)	0.55 (0.50 to 0.63)
	All	-0.13 (-2.96 to 0.71)	-1.6 (-31.0 to 6.5)	6.55 (5.35 to 9.83)	46.7 (40.7 to 51.8)	0.37 (0.31 to 0.52)
SW	Winter	-0.37 (-2.65 to 3.13)	-4.6 (-29.8 to 50.6)	9.07 (6.50 to 12.84)	66.0 (53.1 to 86.0)	0.44 (0.27 to 0.61)
	Spring	-0.61 (-3.65 to 1.04)	-9.4 (-49.8 to 20.6)	5.36 (3.60 to 6.52)	55.7 (48.1 to 63.1)	0.25 (0.12 to 0.44)
	Summer	-2.23 (-3.29 to -1.12)	-28.6 (-46.0 to -18.5)	5.16 (3.80 to 6.12)	52.5 (43.1 to 53.7)	0.30 (0.22 to 0.59)
	Fall	0.58 (-1.74 to 2.32)	10.5 (-24.4 to 38.5)	4.86 (4.06 to 6.60)	57.6 (43.1 to 76.0)	0.47 (0.34 to 0.56)
	All	-0.23 (-2.48 to 0.82)	-3.7 (-35.6 to 12.1)	6.54 (4.77 to 8.01)	57.7 (48.9 to 68.6)	0.40 (0.29 to 0.52)
NRP	Winter	-2.18 (-5.29 to -0.77)	-35.5 (-51.8 to -12.3)	7.40 (6.25 to 10.79)	63.4 (59.1 to 64.8)	0.28 (0.23 to 0.37)
	Spring	-1.52 (-2.55 to 0.44)	-25.8 (-42.7 to 9.6)	4.26 (3.80 to 29.58)	50.8 (47.4 to 63.9)	0.53 (0.20 to 0.60)
	Summer	-2.05 (-3.48 to -0.91)	-26.5 (-57.2 to -8.8)	5.21 (4.42 to 17.61)	54.0 (48.5 to 73.8)	0.50 (0.24 to 0.80)
	Fall	-0.51 (-2.29 to -0.15)	-7.7 (-36.1 to -2.6)	5.97 (4.60 to 8.74)	55.4 (51.5 to 60.9)	0.40 (0.26 to 0.74)
	All	-1.50 (-2.92 to -0.99)	-23.3 (-45.1 to -15.1)	6.80 (5.05 to 18.06)	55.8 (53.5 to 66.0)	0.39 (0.24 to 0.70)
NW	Winter	-0.66 (-3.20 to 0.82)	-6.1 (-35.9 to 8.2)	10.20 (8.18 to 13.06)	77.6 (68.0 to 81.2)	0.29 (0.14 to 0.45)
	Spring	0.54 (-1.93 to 1.84)	11.8 (-35.2 to 40.5)	6.24 (4.10 to 6.72)	71.5 (57.8 to 85.5)	0.34 (0.25 to 0.48)
	Summer	-0.04 (-2.58 to 1.95)	-0.8 (-39.9 to 21.2)	7.24 (4.03 to 32.40)	62.6 (46.6 to 67.7)	0.39 (0.28 to 0.53)
	Fall	0.97 (-1.22 to 3.09)	9.8 (-18.4 to 34.9)	9.36 (5.78 to 15.33)	65.2 (52.3 to 82.3)	0.43 (0.31 to 0.56)
	All	0.06 (-1.80 to 1.55)	1.0 (-25.5 to 21.2)	9.09 (5.75 to 19.26)	69.8 (57.3 to 75.4)	0.40 (0.31 to 0.47)
West	Winter	-2.08 (-2.99 to 0.30)	-17.8 (-25.1 to 2.6)	9.66 (8.09 to 10.75)	49.5 (43.3 to 53.3)	0.61 (0.53 to 0.68)
	Spring	-1.05 (-3.03 to 1.68)	-12.7 (-34.0 to 22.1)	5.86 (4.17 to 6.75)	46.4 (38.3 to 56.0)	0.48 (0.44 to 0.61)
	Summer	-1.73 (-3.40 to 3.41)	-17.2 (-34.8 to 22.0)	6.58 (5.76 to 15.92)	46.8 (43.5 to 58.4)	0.48 (0.37 to 0.72)
	Fall	-1.33 (-2.43 to 0.56)	-13.4 (-21.1 to 5.8)	7.70 (6.36 to 16.85)	47.1 (44.0 to 53.9)	0.53 (0.37 to 0.56)
	All	-1.43 (-2.55 to 0.70)	-12.2 (-25.0 to 5.7)	7.95 (6.57 to 11.12)	47.6 (43.1 to 53.4)	0.53 (0.44 to 0.65)
All	Winter	1.00 (-0.49 to 3.12)	11.3 (-4.8 to 30.4)	8.13 (7.12 to 10.33)	51.9 (46.6 to 65.6)	0.51 (0.45 to 0.56)
	Spring	-0.17 (-2.12 to 0.49)	-2.1 (-22.9 to 6.8)	5.61 (4.95 to 8.86)	43.8 (42.3 to 45.5)	0.52 (0.35 to 0.60)
	Summer	-2.22 (-3.84 to -0.22)	-20.9 (-39.3 to -2.5)	6.19 (5.69 to 13.01)	42.3 (39.7 to 48.5)	0.55 (0.47 to 0.63)
	Fall	1.13 (-0.94 to 1.86)	14.5 (-11.2 to 20.8)	6.94 (5.37 to 9.22)	47.9 (41.3 to 52.9)	0.55 (0.44 to 0.60)
	All	0.12 (-1.74 to 0.61)	1.1 (-18.9 to 7.3)	7.09 (5.93 to 9.94)	46.4 (43.4 to 50.3)	0.51 (0.45 to 0.57)

Table S2. Median (range) of performance statistics for PM_{2.5} sulfate at CSN sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	-0.37 (-0.96 to 0.00)	-16.6 (-33.2 to 0.3)	1.28 (1.01 to 1.95)	41.0 (30.5 to 52.7)	0.48 (0.18 to 0.68)
	Spring	-0.07 (-0.94 to 0.01)	-4.0 (-32.0 to 0.7)	0.78 (0.57 to 1.44)	29.6 (26.3 to 37.0)	0.73 (0.57 to 0.86)
	Summer	-0.45 (-1.44 to 0.04)	-14.6 (-29.0 to 2.8)	1.48 (0.56 to 2.80)	29.9 (24.8 to 38.3)	0.82 (0.77 to 0.88)
	Fall	-0.14 (-0.57 to 0.12)	-7.8 (-22.3 to 10.8)	0.81 (0.63 to 1.72)	33.5 (28.5 to 37.3)	0.80 (0.63 to 0.87)
	All	-0.29 (-0.94 to 0.02)	-14.7 (-29.3 to 1.7)	1.06 (0.79 to 1.88)	31.9 (31.0 to 39.6)	0.72 (0.61 to 0.86)
SE	Winter	-0.13 (-0.65 to 0.08)	-5.4 (-28.8 to 6.1)	1.03 (0.74 to 1.29)	40.2 (32.6 to 44.6)	0.54 (0.41 to 0.65)
	Spring	-0.46 (-1.31 to 0.01)	-18.1 (-38.5 to 0.8)	0.95 (0.68 to 1.76)	29.4 (26.2 to 41.0)	0.65 (0.54 to 0.74)
	Summer	-0.71 (-1.84 to 0.12)	-21.2 (-40.0 to 6.6)	1.34 (0.52 to 2.81)	31.6 (26.8 to 42.0)	0.68 (0.61 to 0.76)
	Fall	-0.04 (-0.84 to 0.22)	-2.2 (-30.0 to 24.9)	0.82 (0.56 to 1.58)	31.9 (27.9 to 43.7)	0.72 (0.57 to 0.77)
	All	-0.36 (-1.16 to 0.07)	-15.1 (-35.4 to 5.7)	1.03 (0.64 to 1.91)	32.7 (30.4 to 40.5)	0.64 (0.60 to 0.76)
Ohio Valley	Winter	-0.65 (-1.23 to -0.49)	-32.0 (-42.2 to -19.3)	1.41 (1.24 to 2.24)	42.8 (36.5 to 47.2)	0.54 (0.28 to 0.64)
	Spring	-0.35 (-1.36 to -0.03)	-16.1 (-38.2 to -1.8)	1.13 (0.83 to 1.96)	31.3 (25.3 to 40.4)	0.69 (0.60 to 0.84)
	Summer	-0.65 (-1.48 to -0.02)	-17.7 (-32.6 to -0.6)	1.52 (1.00 to 2.95)	31.4 (27.3 to 38.8)	0.73 (0.68 to 0.78)
	Fall	-0.24 (-0.92 to 0.11)	-11.7 (-27.0 to 8.2)	0.91 (0.59 to 1.89)	30.6 (28.3 to 34.6)	0.79 (0.71 to 0.87)
	All	-0.43 (-1.25 to -0.11)	-18.0 (-34.6 to -6.2)	1.33 (0.99 to 2.15)	32.8 (31.0 to 39.9)	0.70 (0.62 to 0.82)
MW	Winter	-0.38 (-0.95 to -0.16)	-21.8 (-40.5 to -8.9)	1.02 (0.76 to 1.46)	40.0 (34.9 to 46.2)	0.59 (0.47 to 0.71)
	Spring	-0.06 (-0.85 to 0.05)	-3.5 (-35.0 to 3.5)	0.82 (0.55 to 1.31)	29.3 (25.6 to 39.8)	0.80 (0.67 to 0.91)
	Summer	-0.41 (-0.87 to 0.08)	-16.9 (-33.7 to 4.8)	1.18 (0.70 to 1.80)	33.5 (26.9 to 42.3)	0.80 (0.74 to 0.86)
	Fall	-0.13 (-0.64 to 0.18)	-8.6 (-29.2 to 14.3)	0.71 (0.62 to 1.48)	34.5 (28.0 to 39.9)	0.81 (0.71 to 0.90)
	All	-0.18 (-0.82 to -0.00)	-10.5 (-34.2 to -0.4)	0.93 (0.66 to 1.47)	33.9 (30.2 to 40.4)	0.79 (0.71 to 0.84)
South	Winter	-0.15 (-0.65 to 0.10)	-11.8 (-34.8 to 8.3)	1.09 (0.69 to 2.17)	40.8 (35.7 to 51.2)	0.64 (0.40 to 0.70)
	Spring	-0.47 (-1.35 to -0.04)	-21.9 (-47.6 to -2.2)	1.00 (0.72 to 1.82)	32.4 (28.1 to 48.7)	0.63 (0.50 to 0.82)
	Summer	-0.65 (-1.49 to -0.18)	-25.8 (-49.9 to -9.7)	1.13 (0.75 to 2.05)	37.6 (29.5 to 52.0)	0.61 (0.53 to 0.70)
	Fall	-0.17 (-1.06 to 0.10)	-10.8 (-37.8 to 7.7)	0.80 (0.65 to 1.63)	33.0 (29.6 to 43.1)	0.71 (0.58 to 0.83)
	All	-0.36 (-1.14 to -0.02)	-15.6 (-43.4 to -0.9)	1.10 (0.73 to 1.87)	34.6 (31.7 to 47.1)	0.66 (0.54 to 0.70)
SW	Winter	-0.04 (-0.28 to 0.16)	-5.9 (-33.7 to 28.9)	0.59 (0.34 to 1.19)	53.5 (47.7 to 69.4)	0.33 (0.12 to 0.65)
	Spring	-0.12 (-0.35 to 0.35)	-14.6 (-36.7 to 57.1)	0.43 (0.24 to 0.61)	34.5 (28.4 to 65.3)	0.55 (0.38 to 0.65)
	Summer	-0.39 (-0.46 to -0.11)	-34.8 (-45.2 to -13.3)	0.58 (0.39 to 0.68)	43.5 (38.2 to 50.0)	0.43 (0.09 to 0.53)
	Fall	-0.08 (-0.37 to 0.10)	-12.9 (-37.8 to 17.2)	0.42 (0.29 to 0.54)	40.3 (31.8 to 52.7)	0.53 (0.24 to 0.78)
	All	-0.11 (-0.32 to 0.11)	-14.9 (-36.1 to 17.9)	0.49 (0.43 to 0.66)	43.3 (39.7 to 52.0)	0.38 (0.29 to 0.55)
NRP	Winter	-0.20 (-0.57 to -0.05)	-21.6 (-45.5 to -6.0)	0.67 (0.48 to 0.90)	43.9 (35.6 to 59.7)	0.72 (0.43 to 0.77)
	Spring	-0.17 (-0.54 to 0.09)	-12.4 (-40.3 to 11.2)	0.66 (0.35 to 0.80)	35.1 (26.1 to 42.6)	0.76 (0.61 to 0.84)
	Summer	-0.19 (-0.52 to 0.18)	-17.6 (-38.3 to 21.3)	0.64 (0.41 to 1.16)	40.4 (25.8 to 58.7)	0.78 (0.50 to 0.90)
	Fall	-0.03 (-0.41 to 0.13)	-3.5 (-32.3 to 17.8)	0.52 (0.35 to 1.15)	38.0 (30.3 to 49.4)	0.78 (0.63 to 0.91)
	All	-0.17 (-0.51 to 0.02)	-15.9 (-39.2 to 3.3)	0.73 (0.40 to 0.88)	39.0 (31.3 to 51.0)	0.76 (0.56 to 0.81)
NW	Winter	0.26 (-0.15 to 0.44)	47.8 (-14.1 to 103.4)	0.69 (0.55 to 2.13)	70.8 (57.9 to 127.8)	0.31 (0.13 to 0.68)
	Spring	0.20 (-0.13 to 0.53)	36.3 (-14.7 to 99.2)	0.50 (0.35 to 0.70)	51.4 (37.7 to 103.8)	0.55 (0.44 to 0.70)
	Summer	0.06 (-0.23 to 0.42)	5.9 (-20.4 to 62.3)	0.58 (0.39 to 0.91)	38.4 (30.5 to 81.4)	0.60 (0.14 to 0.73)
	Fall	0.18 (0.06 to 0.49)	20.1 (-8.6 to 108.3)	0.67 (0.37 to 0.90)	57.0 (37.7 to 113.3)	0.42 (0.29 to 0.60)
	All	0.15 (-0.03 to 0.45)	17.8 (-4.0 to 87.6)	0.66 (0.43 to 1.07)	52.6 (43.5 to 103.2)	0.46 (0.19 to 0.52)
West	Winter	0.11 (-0.27 to 0.24)	16.2 (-29.2 to 39.5)	0.64 (0.41 to 0.98)	63.6 (46.5 to 66.7)	0.46 (0.29 to 0.65)
	Spring	-0.18 (-0.62 to 0.24)	-17.5 (-39.1 to 24.2)	0.61 (0.50 to 1.18)	38.7 (35.1 to 47.5)	0.68 (0.54 to 0.73)
	Summer	-0.62 (-0.76 to -0.03)	-33.0 (-44.5 to -2.1)	1.05 (0.61 to 2.02)	43.1 (32.6 to 48.3)	0.56 (0.49 to 0.65)
	Fall	-0.20 (-0.72 to 0.20)	-20.1 (-43.9 to 27.3)	0.55 (0.46 to 1.20)	42.2 (35.4 to 48.5)	0.61 (0.57 to 0.74)
	All	-0.23 (-0.57 to 0.14)	-19.7 (-36.8 to 14.5)	0.80 (0.56 to 1.41)	44.3 (39.0 to 47.3)	0.60 (0.49 to 0.68)
All	Winter	-0.27 (-0.76 to -0.10)	-15.7 (-34.9 to -7.5)	1.13 (1.00 to 1.55)	43.1 (37.7 to 47.3)	0.58 (0.42 to 0.63)
	Spring	-0.22 (-1.00 to 0.04)	-11.2 (-37.2 to 3.1)	0.90 (0.67 to 1.54)	31.9 (27.5 to 40.7)	0.73 (0.68 to 0.84)
	Summer	-0.54 (-1.22 to -0.01)	-19.2 (-34.7 to -0.7)	1.36 (0.80 to 2.43)	32.9 (28.9 to 41.1)	0.79 (0.73 to 0.84)
	Fall	-0.16 (-0.70 to 0.15)	-11.3 (-28.7 to 15.3)	0.76 (0.58 to 1.53)	34.1 (31.0 to 37.8)	0.79 (0.71 to 0.84)
	All	-0.27 (-0.92 to 0.02)	-13.2 (-34.0 to 1.8)	1.06 (0.79 to 1.73)	33.9 (32.9 to 41.2)	0.74 (0.67 to 0.83)

Table S3. Median (range) of performance statistics for PM_{2.5} sulfate at IMPROVE sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	-0.17 (-0.44 to 0.08)	-16.2 (-25.5 to 7.2)	0.71 (0.44 to 1.16)	35.5 (24.8 to 45.6)	0.69 (0.42 to 0.83)
	Spring	0.01 (-0.39 to 0.29)	0.8 (-21.3 to 29.2)	0.72 (0.37 to 0.98)	29.7 (27.1 to 47.1)	0.78 (0.61 to 0.87)
	Summer	-0.21 (-1.06 to 0.09)	-11.9 (-29.6 to 8.2)	1.16 (0.42 to 2.19)	34.9 (29.9 to 42.8)	0.84 (0.70 to 0.89)
	Fall	-0.01 (-0.25 to 0.14)	-1.1 (-17.0 to 15.5)	0.55 (0.32 to 1.17)	33.5 (31.1 to 41.2)	0.84 (0.75 to 0.90)
	All	-0.07 (-0.47 to 0.14)	-6.3 (-23.4 to 13.3)	0.77 (0.42 to 1.42)	33.5 (31.0 to 40.2)	0.78 (0.68 to 0.86)
SE	Winter	-0.14 (-0.47 to 0.02)	-10.7 (-26.1 to 1.3)	0.78 (0.63 to 1.07)	36.0 (32.0 to 39.4)	0.60 (0.53 to 0.66)
	Spring	-0.36 (-0.94 to -0.14)	-17.0 (-32.8 to -9.2)	0.93 (0.60 to 1.69)	30.1 (24.2 to 37.4)	0.66 (0.59 to 0.77)
	Summer	-0.83 (-1.58 to -0.19)	-25.9 (-39.1 to -10.2)	1.34 (0.55 to 2.80)	32.9 (26.3 to 42.4)	0.73 (0.64 to 0.79)
	Fall	-0.25 (-0.74 to 0.10)	-11.2 (-30.2 to 11.2)	0.82 (0.45 to 1.53)	34.3 (28.9 to 38.8)	0.73 (0.56 to 0.80)
	All	-0.40 (-0.91 to -0.11)	-18.2 (-33.0 to -8.1)	0.99 (0.57 to 1.88)	32.3 (30.0 to 39.5)	0.70 (0.63 to 0.77)
Ohio Valley	Winter	-0.52 (-0.80 to -0.33)	-25.8 (-40.0 to -22.9)	0.98 (0.66 to 1.32)	37.3 (33.8 to 43.3)	0.67 (0.59 to 0.74)
	Spring	-0.41 (-1.15 to -0.14)	-16.7 (-34.2 to -9.3)	1.09 (0.66 to 1.77)	31.0 (25.6 to 36.1)	0.76 (0.59 to 0.84)
	Summer	-0.98 (-1.75 to -0.17)	-27.8 (-35.6 to -8.7)	1.56 (0.78 to 3.07)	34.0 (28.5 to 38.6)	0.77 (0.73 to 0.83)
	Fall	-0.28 (-0.89 to 0.08)	-16.2 (-29.3 to 7.2)	0.87 (0.45 to 1.99)	32.0 (29.4 to 37.8)	0.76 (0.65 to 0.88)
	All	-0.51 (-1.09 to -0.14)	-21.3 (-33.8 to -9.4)	1.16 (0.66 to 2.10)	32.0 (30.5 to 38.4)	0.76 (0.74 to 0.87)
MW	Winter	-0.28 (-0.54 to -0.09)	-26.4 (-37.3 to -6.6)	0.72 (0.50 to 0.87)	38.3 (35.4 to 46.5)	0.71 (0.44 to 0.75)
	Spring	-0.10 (-0.57 to 0.07)	-9.0 (-33.3 to 8.0)	0.56 (0.37 to 0.99)	32.2 (27.7 to 38.1)	0.81 (0.66 to 0.87)
	Summer	-0.21 (-0.71 to 0.11)	-18.0 (-37.9 to 11.7)	0.59 (0.41 to 1.49)	34.8 (29.3 to 44.5)	0.86 (0.80 to 0.92)
	Fall	-0.07 (-0.59 to 0.15)	-7.5 (-34.6 to 17.3)	0.54 (0.32 to 1.31)	38.0 (33.6 to 44.7)	0.83 (0.75 to 0.91)
	All	-0.13 (-0.51 to 0.01)	-10.9 (-34.1 to 1.6)	0.61 (0.40 to 1.11)	36.1 (30.3 to 39.5)	0.80 (0.74 to 0.88)
South	Winter	-0.17 (-0.41 to -0.01)	-14.3 (-36.2 to -1.2)	0.72 (0.46 to 0.91)	38.5 (37.4 to 48.5)	0.69 (0.55 to 0.80)
	Spring	-0.40 (-0.86 to -0.10)	-23.0 (-46.2 to -8.9)	0.88 (0.60 to 1.34)	36.2 (28.4 to 47.3)	0.72 (0.60 to 0.79)
	Summer	-0.70 (-1.19 to -0.54)	-37.5 (-45.4 to -29.6)	1.06 (0.81 to 1.98)	41.0 (33.6 to 48.5)	0.73 (0.58 to 0.81)
	Fall	-0.25 (-0.62 to -0.05)	-19.0 (-34.1 to -4.3)	0.69 (0.49 to 1.69)	34.5 (27.8 to 42.2)	0.77 (0.64 to 0.88)
	All	-0.37 (-0.73 to -0.22)	-24.3 (-40.5 to -17.3)	0.86 (0.66 to 1.40)	37.9 (33.6 to 43.9)	0.74 (0.65 to 0.81)
SW	Winter	0.11 (0.02 to 0.18)	33.7 (3.9 to 51.8)	0.24 (0.21 to 0.37)	53.6 (37.3 to 69.2)	0.56 (0.51 to 0.76)
	Spring	-0.12 (-0.35 to 0.37)	-18.5 (-41.3 to 73.9)	0.42 (0.24 to 0.52)	34.1 (29.2 to 83.4)	0.56 (0.40 to 0.64)
	Summer	-0.39 (-0.46 to -0.19)	-38.2 (-49.9 to -24.3)	0.54 (0.41 to 0.60)	48.6 (38.9 to 53.4)	0.62 (0.32 to 0.76)
	Fall	-0.14 (-0.30 to 0.05)	-21.9 (-37.8 to 10.6)	0.33 (0.27 to 0.46)	40.7 (32.1 to 49.4)	0.68 (0.56 to 0.90)
	All	-0.13 (-0.25 to 0.09)	-22.3 (-35.6 to 17.6)	0.43 (0.36 to 0.45)	42.9 (39.8 to 56.0)	0.53 (0.46 to 0.74)
NRP	Winter	-0.03 (-0.12 to 0.08)	-7.9 (-25.5 to 23.4)	0.38 (0.28 to 0.64)	52.6 (49.3 to 58.3)	0.68 (0.47 to 0.79)
	Spring	-0.06 (-0.38 to 0.23)	-10.9 (-42.8 to 43.8)	0.40 (0.32 to 0.64)	38.3 (29.2 to 61.4)	0.73 (0.60 to 0.82)
	Summer	-0.06 (-0.25 to 0.22)	-11.9 (-35.5 to 43.8)	0.35 (0.25 to 0.68)	41.3 (30.0 to 65.6)	0.68 (0.38 to 0.78)
	Fall	-0.02 (-0.18 to 0.20)	-3.1 (-31.9 to 62.4)	0.33 (0.23 to 0.46)	41.4 (35.8 to 79.9)	0.70 (0.47 to 0.81)
	All	-0.02 (-0.24 to 0.16)	-3.3 (-35.4 to 37.2)	0.40 (0.30 to 0.57)	44.4 (33.5 to 64.4)	0.72 (0.51 to 0.78)
NW	Winter	0.15 (0.09 to 0.21)	64.9 (34.4 to 142.5)	0.33 (0.24 to 0.64)	98.7 (67.7 to 162.4)	0.45 (0.28 to 0.56)
	Spring	0.12 (-0.08 to 0.42)	26.9 (-13.5 to 104.5)	0.34 (0.28 to 0.57)	48.3 (37.8 to 106.9)	0.68 (0.57 to 0.78)
	Summer	0.04 (-0.14 to 0.35)	7.1 (-17.9 to 79.1)	0.36 (0.25 to 0.61)	39.9 (31.3 to 87.2)	0.64 (0.24 to 0.77)
	Fall	0.11 (-0.00 to 0.38)	28.4 (-0.4 to 152.4)	0.40 (0.29 to 0.84)	60.9 (46.6 to 161.7)	0.51 (0.29 to 0.65)
	All	0.10 (-0.02 to 0.34)	25.0 (-4.4 to 108.5)	0.40 (0.31 to 0.61)	54.8 (45.4 to 116.1)	0.56 (0.45 to 0.67)
West	Winter	0.16 (0.03 to 0.23)	56.6 (10.2 to 93.3)	0.34 (0.22 to 0.48)	87.4 (60.5 to 107.1)	0.51 (0.30 to 0.65)
	Spring	-0.08 (-0.35 to 0.32)	-12.1 (-38.5 to 50.4)	0.44 (0.35 to 0.77)	42.5 (38.4 to 68.4)	0.55 (0.45 to 0.65)
	Summer	-0.29 (-0.39 to 0.10)	-27.8 (-43.5 to 12.7)	0.63 (0.48 to 1.17)	48.2 (42.8 to 52.7)	0.38 (0.22 to 0.56)
	Fall	-0.10 (-0.27 to 0.19)	-15.9 (-35.9 to 43.0)	0.42 (0.30 to 0.51)	46.1 (40.5 to 72.2)	0.52 (0.36 to 0.61)
	All	-0.09 (-0.23 to 0.20)	-14.4 (-29.8 to 37.1)	0.48 (0.42 to 0.73)	50.4 (44.6 to 66.0)	0.48 (0.43 to 0.63)
All	Winter	-0.03 (-0.19 to 0.07)	-4.2 (-21.6 to 11.8)	0.55 (0.44 to 0.72)	45.0 (40.6 to 48.7)	0.75 (0.71 to 0.80)
	Spring	-0.09 (-0.46 to 0.18)	-10.6 (-33.9 to 23.6)	0.60 (0.41 to 0.95)	36.1 (30.5 to 50.1)	0.80 (0.66 to 0.88)
	Summer	-0.35 (-0.63 to -0.01)	-25.6 (-36.1 to -1.2)	0.86 (0.55 to 1.61)	40.4 (36.2 to 43.1)	0.83 (0.70 to 0.88)
	Fall	-0.11 (-0.34 to 0.13)	-13.7 (-28.2 to 22.6)	0.52 (0.45 to 0.95)	38.3 (35.7 to 50.5)	0.81 (0.70 to 0.87)
	All	-0.13 (-0.40 to 0.08)	-12.6 (-31.0 to 10.8)	0.65 (0.50 to 1.11)	38.6 (35.8 to 46.7)	0.81 (0.70 to 0.87)

Table S4. Median (range) of performance statistics for PM_{2.5} nitrate at CSN sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	r
NE	Winter	0.37 (-0.12 to 1.38)	16.4 (-4.8 to 65.8)	1.66 (1.42 to 2.33)	45.9 (41.6 to 77.5)	0.68 (0.66 to 0.75)
	Spring	0.01 (-0.28 to 0.63)	1.3 (-20.1 to 49.3)	1.12 (0.63 to 1.57)	55.5 (44.0 to 78.4)	0.70 (0.58 to 0.77)
	Summer	-0.17 (-0.28 to 0.11)	-29.0 (-58.2 to 13.8)	0.53 (0.40 to 1.25)	69.5 (55.5 to 85.2)	0.54 (0.39 to 0.61)
	Fall	0.39 (0.08 to 0.57)	40.2 (8.9 to 55.6)	1.21 (0.82 to 1.83)	74.3 (58.3 to 85.5)	0.69 (0.63 to 0.79)
	All	0.14 (-0.06 to 0.42)	11.2 (-5.0 to 36.6)	1.24 (0.89 to 1.59)	55.6 (49.7 to 70.1)	0.74 (0.69 to 0.78)
SE	Winter	0.71 (0.39 to 1.14)	76.7 (31.9 to 116.9)	1.45 (1.22 to 1.74)	93.7 (75.0 to 129.2)	0.62 (0.55 to 0.68)
	Spring	-0.02 (-0.20 to 0.38)	-3.9 (-30.2 to 46.8)	0.59 (0.39 to 1.28)	68.7 (55.6 to 105.1)	0.55 (0.37 to 0.72)
	Summer	-0.11 (-0.25 to 0.27)	-32.3 (-67.9 to 97.2)	0.31 (0.24 to 1.39)	72.6 (59.7 to 143.7)	0.22 (0.06 to 0.44)
	Fall	0.38 (0.21 to 0.64)	86.8 (44.3 to 119.4)	0.96 (0.77 to 1.60)	130.8 (99.9 to 144.4)	0.60 (0.52 to 0.78)
	All	0.24 (0.08 to 0.35)	46.3 (12.4 to 65.3)	1.01 (0.80 to 1.31)	92.0 (83.5 to 105.8)	0.67 (0.55 to 0.72)
Ohio Valley	Winter	0.05 (-0.93 to 0.77)	1.8 (-21.3 to 34.2)	1.77 (1.57 to 2.67)	41.6 (38.6 to 51.1)	0.68 (0.57 to 0.79)
	Spring	0.05 (-0.72 to 0.79)	3.7 (-41.7 to 48.1)	1.26 (0.75 to 2.07)	55.5 (46.9 to 83.7)	0.74 (0.64 to 0.78)
	Summer	-0.11 (-0.23 to 0.22)	-17.4 (-49.2 to 31.5)	0.70 (0.50 to 1.49)	76.8 (65.6 to 107.1)	0.43 (0.18 to 0.54)
	Fall	0.55 (0.05 to 0.82)	46.0 (4.8 to 61.0)	1.41 (0.67 to 1.93)	69.4 (60.9 to 94.5)	0.73 (0.59 to 0.88)
	All	0.15 (-0.25 to 0.42)	9.4 (-15.6 to 26.5)	1.25 (1.06 to 2.00)	52.1 (48.5 to 67.2)	0.77 (0.63 to 0.84)
MW	Winter	-0.25 (-1.14 to 0.85)	-5.5 (-26.5 to 27.0)	1.76 (1.23 to 3.46)	36.3 (29.6 to 40.8)	0.78 (0.57 to 0.86)
	Spring	-0.01 (-0.82 to 0.34)	-0.3 (-38.8 to 14.8)	1.50 (1.00 to 2.15)	48.0 (39.8 to 62.2)	0.71 (0.62 to 0.81)
	Summer	-0.07 (-0.22 to 0.34)	-12.2 (-44.4 to 42.8)	0.70 (0.45 to 1.10)	71.8 (58.4 to 89.6)	0.48 (0.35 to 0.61)
	Fall	0.54 (0.03 to 0.89)	36.1 (2.0 to 49.8)	1.42 (0.81 to 2.17)	57.8 (44.8 to 73.1)	0.79 (0.63 to 0.93)
	All	0.06 (-0.31 to 0.34)	3.9 (-17.2 to 20.7)	1.45 (1.03 to 2.18)	47.2 (42.6 to 50.9)	0.80 (0.71 to 0.88)
South	Winter	0.05 (-0.22 to 0.39)	4.0 (-14.3 to 21.7)	1.16 (0.91 to 1.81)	50.5 (44.8 to 59.6)	0.69 (0.61 to 0.85)
	Spring	-0.23 (-0.52 to 0.25)	-25.7 (-51.1 to 22.3)	0.79 (0.58 to 1.59)	60.5 (54.3 to 74.6)	0.74 (0.43 to 0.84)
	Summer	-0.17 (-0.30 to 0.40)	-52.8 (-80.8 to 107.4)	0.42 (0.29 to 1.90)	84.6 (59.5 to 186.5)	0.30 (0.11 to 0.46)
	Fall	0.06 (-0.01 to 0.66)	10.9 (-3.1 to 102.2)	0.61 (0.44 to 1.64)	79.3 (65.9 to 149.0)	0.63 (0.51 to 0.73)
	All	-0.04 (-0.10 to 0.33)	-6.5 (-13.1 to 31.3)	0.93 (0.66 to 1.40)	62.9 (58.0 to 79.4)	0.72 (0.45 to 0.79)
SW	Winter	-1.99 (-3.10 to -0.79)	-55.6 (-70.5 to -39.3)	4.41 (2.05 to 6.96)	66.3 (58.6 to 75.1)	0.50 (0.32 to 0.71)
	Spring	-0.31 (-0.55 to -0.13)	-38.4 (-58.0 to -22.5)	0.89 (0.52 to 1.22)	62.6 (50.8 to 73.4)	0.59 (0.34 to 0.77)
	Summer	-0.21 (-0.35 to -0.03)	-53.3 (-85.6 to -8.8)	0.42 (0.37 to 0.65)	85.2 (82.3 to 118.1)	0.21 (-0.09 to 0.57)
	Fall	-0.30 (-0.57 to 0.29)	-39.6 (-50.4 to 40.7)	1.22 (0.89 to 1.65)	71.5 (61.5 to 127.0)	0.53 (0.16 to 0.71)
	All	-0.70 (-1.12 to -0.32)	-52.6 (-65.7 to -29.1)	2.28 (1.18 to 3.53)	68.4 (65.4 to 81.0)	0.57 (0.48 to 0.70)
NRP	Winter	-0.26 (-1.21 to 0.72)	-12.6 (-32.8 to 34.6)	1.80 (1.06 to 2.80)	49.8 (40.5 to 70.6)	0.71 (0.68 to 0.86)
	Spring	-0.33 (-0.81 to -0.05)	-23.8 (-58.0 to -6.3)	1.00 (0.74 to 2.42)	49.8 (40.0 to 66.5)	0.80 (0.64 to 0.93)
	Summer	-0.16 (-0.18 to 0.04)	-40.8 (-57.9 to 14.2)	0.36 (0.31 to 0.87)	75.8 (64.1 to 95.7)	0.51 (0.24 to 0.63)
	Fall	0.37 (-0.03 to 0.71)	47.9 (-5.2 to 82.5)	1.20 (0.47 to 1.56)	86.5 (50.8 to 111.1)	0.79 (0.62 to 0.90)
	All	-0.14 (-0.34 to 0.27)	-10.6 (-34.3 to 26.0)	1.30 (0.81 to 1.95)	59.0 (45.2 to 72.8)	0.76 (0.64 to 0.88)
NW	Winter	-0.27 (-1.05 to 0.62)	-16.2 (-38.4 to 36.0)	1.87 (1.17 to 7.14)	75.5 (60.5 to 96.8)	0.36 (0.24 to 0.61)
	Spring	0.24 (-0.09 to 0.59)	56.1 (-12.6 to 106.7)	0.97 (0.46 to 1.64)	89.8 (57.3 to 146.2)	0.58 (0.37 to 0.71)
	Summer	-0.01 (-0.28 to 0.69)	-1.6 (-70.2 to 224.1)	0.53 (0.32 to 2.63)	90.6 (67.7 to 278.0)	0.35 (0.16 to 0.54)
	Fall	0.26 (-0.10 to 0.62)	19.7 (-9.7 to 96.8)	1.35 (0.85 to 2.52)	87.4 (65.9 to 132.5)	0.44 (0.29 to 0.55)
	All	0.10 (-0.29 to 0.54)	11.0 (-28.9 to 70.5)	1.26 (0.86 to 3.48)	87.0 (66.1 to 121.8)	0.41 (0.28 to 0.57)
West	Winter	-1.82 (-2.93 to -1.16)	-41.8 (-60.2 to -31.0)	4.86 (3.50 to 5.94)	57.9 (48.4 to 67.6)	0.63 (0.47 to 0.78)
	Spring	-0.61 (-1.19 to -0.10)	-39.3 (-53.3 to -6.4)	1.82 (1.25 to 3.23)	53.9 (47.3 to 59.3)	0.74 (0.64 to 0.79)
	Summer	-0.48 (-0.97 to -0.27)	-32.9 (-62.2 to -14.2)	1.47 (1.01 to 2.38)	54.7 (42.0 to 67.8)	0.78 (0.58 to 0.88)
	Fall	-1.03 (-1.88 to -0.56)	-40.7 (-52.8 to -29.9)	2.91 (2.46 to 4.76)	57.8 (53.8 to 68.7)	0.62 (0.48 to 0.83)
	All	-0.96 (-1.45 to -0.70)	-40.3 (-52.2 to -29.6)	3.02 (2.33 to 4.20)	58.0 (49.2 to 62.5)	0.67 (0.60 to 0.77)
All	Winter	-0.08 (-0.52 to 0.47)	-3.9 (-17.1 to 20.5)	2.56 (1.84 to 3.12)	50.7 (48.3 to 61.9)	0.61 (0.54 to 0.68)
	Spring	-0.15 (-0.35 to 0.27)	-13.5 (-28.5 to 18.5)	1.17 (0.95 to 1.80)	55.6 (50.8 to 71.3)	0.69 (0.62 to 0.77)
	Summer	-0.14 (-0.25 to 0.08)	-25.7 (-47.6 to 14.8)	0.69 (0.58 to 1.19)	69.7 (64.6 to 94.0)	0.57 (0.29 to 0.82)
	Fall	0.19 (0.08 to 0.43)	19.8 (9.1 to 35.5)	1.37 (1.07 to 2.26)	72.8 (66.9 to 82.2)	0.61 (0.51 to 0.78)
	All	-0.05 (-0.15 to 0.12)	-4.8 (-10.4 to 9.4)	1.60 (1.25 to 2.18)	61.1 (55.2 to 65.3)	0.66 (0.62 to 0.71)

Table S5. Median (range) of performance statistics for PM_{2.5} nitrate at IMPROVE sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	0.66 (0.41 to 1.15)	92.2 (59.7 to 194.6)	1.12 (0.88 to 1.87)	110.5 (81.5 to 200.7)	0.71 (0.58 to 0.80)
	Spring	0.08 (-0.01 to 0.48)	18.6 (-4.2 to 89.2)	0.55 (0.38 to 1.24)	90.7 (69.8 to 127.5)	0.64 (0.45 to 0.71)
	Summer	-0.02 (-0.08 to 0.09)	-10.8 (-47.7 to 48.0)	0.25 (0.20 to 0.71)	84.6 (78.8 to 118.8)	0.38 (0.27 to 0.48)
	Fall	0.19 (0.07 to 0.49)	66.4 (26.0 to 112.0)	0.68 (0.33 to 1.28)	113.5 (90.9 to 147.1)	0.69 (0.50 to 0.82)
	All	0.19 (0.14 to 0.47)	50.8 (36.2 to 113.0)	0.73 (0.55 to 1.18)	95.5 (81.6 to 155.0)	0.69 (0.63 to 0.75)
SE	Winter	0.42 (0.28 to 0.72)	65.2 (45.0 to 109.6)	1.07 (0.83 to 1.41)	108.6 (80.1 to 142.6)	0.65 (0.58 to 0.68)
	Spring	-0.01 (-0.17 to 0.24)	-1.5 (-43.4 to 40.9)	0.53 (0.41 to 1.22)	90.1 (58.0 to 122.1)	0.57 (0.44 to 0.68)
	Summer	-0.07 (-0.15 to 0.27)	-32.2 (-66.3 to 129.3)	0.30 (0.21 to 1.11)	87.2 (63.4 to 163.5)	0.18 (-0.07 to 0.42)
	Fall	0.17 (0.11 to 0.61)	66.1 (52.4 to 145.9)	0.72 (0.37 to 1.45)	132.2 (98.4 to 198.5)	0.62 (0.47 to 0.76)
	All	0.14 (0.09 to 0.27)	32.9 (22.8 to 56.3)	0.76 (0.51 to 1.11)	106.0 (76.5 to 129.2)	0.64 (0.51 to 0.68)
Ohio Valley	Winter	0.21 (-0.40 to 0.47)	10.8 (-22.6 to 30.2)	1.39 (1.02 to 1.86)	54.2 (47.2 to 66.9)	0.64 (0.59 to 0.72)
	Spring	0.01 (-0.46 to 0.83)	3.1 (-47.0 to 77.5)	1.01 (0.57 to 1.97)	74.9 (51.2 to 106.7)	0.60 (0.47 to 0.71)
	Summer	-0.02 (-0.14 to 0.36)	-6.5 (-63.5 to 139.8)	0.27 (0.19 to 1.39)	79.0 (53.8 to 187.4)	0.38 (0.22 to 0.64)
	Fall	0.48 (-0.01 to 1.04)	79.6 (-1.8 to 140.1)	1.15 (0.36 to 1.82)	110.8 (70.7 to 158.2)	0.70 (0.47 to 0.80)
	All	0.12 (-0.12 to 0.57)	14.5 (-13.8 to 60.5)	1.14 (0.69 to 1.51)	69.0 (54.7 to 92.1)	0.71 (0.48 to 0.77)
MW	Winter	0.16 (-0.65 to 0.69)	6.0 (-23.4 to 35.3)	1.62 (0.91 to 2.15)	47.0 (36.6 to 64.1)	0.79 (0.72 to 0.87)
	Spring	0.02 (-0.55 to 0.35)	2.3 (-44.2 to 28.6)	0.87 (0.82 to 1.64)	52.2 (41.9 to 65.4)	0.80 (0.64 to 0.88)
	Summer	0.02 (-0.11 to 0.21)	12.8 (-55.1 to 76.7)	0.33 (0.20 to 0.72)	75.7 (62.5 to 118.5)	0.64 (0.42 to 0.83)
	Fall	0.67 (0.05 to 0.91)	84.4 (7.2 to 113.1)	1.38 (0.56 to 1.95)	99.0 (66.4 to 133.3)	0.77 (0.64 to 0.89)
	All	0.12 (-0.16 to 0.33)	12.2 (-16.6 to 36.0)	1.22 (0.73 to 1.58)	57.8 (45.1 to 71.7)	0.80 (0.67 to 0.86)
South	Winter	-0.08 (-0.34 to 0.16)	-3.9 (-29.4 to 10.4)	1.15 (0.83 to 1.44)	49.7 (45.7 to 69.1)	0.75 (0.50 to 0.86)
	Spring	-0.08 (-0.41 to 0.20)	-16.3 (-55.4 to 20.6)	0.78 (0.49 to 1.09)	63.4 (43.8 to 81.0)	0.75 (0.41 to 0.92)
	Summer	-0.12 (-0.23 to 0.15)	-53.1 (-85.6 to 56.4)	0.27 (0.19 to 1.21)	88.0 (54.8 to 149.9)	0.22 (0.02 to 0.57)
	Fall	0.14 (-0.01 to 0.61)	43.1 (-3.8 to 158.3)	0.58 (0.35 to 1.42)	98.3 (69.8 to 196.8)	0.70 (0.35 to 0.78)
	All	-0.03 (-0.26 to 0.22)	-5.8 (-37.8 to 28.3)	0.85 (0.57 to 1.05)	66.8 (53.2 to 74.6)	0.78 (0.45 to 0.83)
SW	Winter	-0.11 (-0.16 to -0.03)	-33.2 (-44.8 to -13.6)	0.47 (0.33 to 0.64)	68.1 (63.0 to 74.4)	0.65 (0.57 to 0.77)
	Spring	-0.10 (-0.19 to -0.06)	-48.2 (-79.6 to -26.9)	0.24 (0.20 to 0.32)	78.8 (68.2 to 82.7)	0.38 (0.24 to 0.51)
	Summer	-0.13 (-0.16 to -0.08)	-81.8 (-87.6 to -51.1)	0.20 (0.13 to 0.33)	86.9 (67.2 to 104.5)	0.25 (0.14 to 0.47)
	Fall	-0.02 (-0.07 to 0.04)	-16.1 (-48.1 to 31.1)	0.22 (0.17 to 0.55)	92.5 (73.5 to 128.8)	0.40 (0.33 to 0.54)
	All	-0.09 (-0.17 to -0.05)	-44.5 (-67.9 to -24.7)	0.31 (0.23 to 0.42)	78.6 (71.3 to 87.4)	0.58 (0.51 to 0.67)
NRP	Winter	0.22 (-0.09 to 0.44)	29.7 (-26.6 to 124.4)	0.99 (0.48 to 1.52)	76.2 (57.8 to 173.2)	0.60 (0.41 to 0.76)
	Spring	-0.00 (-0.23 to 0.12)	-1.0 (-64.8 to 39.4)	0.49 (0.25 to 0.64)	69.4 (55.9 to 89.9)	0.71 (0.59 to 0.83)
	Summer	-0.06 (-0.07 to 0.09)	-56.6 (-75.6 to 59.5)	0.17 (0.12 to 0.85)	83.5 (78.4 to 152.8)	0.41 (0.28 to 0.53)
	Fall	0.20 (0.04 to 0.29)	111.4 (29.6 to 143.1)	0.60 (0.31 to 0.79)	147.0 (100.1 to 188.0)	0.70 (0.35 to 0.83)
	All	0.12 (-0.13 to 0.17)	30.7 (-46.7 to 65.5)	0.66 (0.43 to 0.85)	90.7 (67.6 to 115.3)	0.66 (0.37 to 0.74)
NW	Winter	0.03 (-0.13 to 0.13)	10.3 (-44.7 to 37.3)	0.75 (0.30 to 1.34)	92.6 (82.6 to 138.2)	0.43 (0.27 to 0.73)
	Spring	0.03 (-0.01 to 0.16)	16.6 (-5.3 to 117.5)	0.41 (0.29 to 0.75)	105.0 (75.7 to 155.7)	0.63 (0.48 to 0.71)
	Summer	0.01 (-0.12 to 0.17)	4.4 (-69.5 to 129.7)	0.38 (0.17 to 1.52)	105.4 (79.3 to 227.6)	0.45 (0.29 to 0.61)
	Fall	0.13 (-0.02 to 0.24)	71.4 (-7.7 to 117.0)	0.68 (0.36 to 0.79)	144.4 (89.6 to 160.6)	0.50 (0.39 to 0.63)
	All	0.05 (-0.06 to 0.15)	19.9 (-30.1 to 75.8)	0.59 (0.41 to 1.00)	111.2 (84.0 to 149.3)	0.43 (0.28 to 0.60)
West	Winter	-0.23 (-0.53 to -0.00)	-34.4 (-46.1 to -0.1)	1.67 (0.82 to 2.47)	68.2 (51.2 to 79.6)	0.75 (0.58 to 0.91)
	Spring	-0.23 (-0.32 to -0.05)	-37.7 (-51.6 to -11.6)	0.56 (0.52 to 1.04)	68.5 (56.9 to 73.1)	0.58 (0.48 to 0.68)
	Summer	-0.22 (-0.34 to 0.09)	-58.8 (-66.6 to 24.2)	0.43 (0.39 to 0.77)	76.9 (69.9 to 83.7)	0.51 (0.31 to 0.66)
	Fall	-0.24 (-0.47 to 0.02)	-42.1 (-55.7 to 4.9)	1.14 (0.76 to 2.17)	76.4 (71.7 to 82.2)	0.62 (0.50 to 0.71)
	All	-0.23 (-0.41 to 0.00)	-39.8 (-48.3 to 0.3)	1.01 (0.63 to 1.72)	73.2 (65.0 to 74.4)	0.68 (0.58 to 0.84)
All	Winter	0.11 (-0.06 to 0.23)	11.0 (-8.0 to 32.7)	1.17 (0.89 to 1.43)	70.1 (62.1 to 92.1)	0.70 (0.58 to 0.77)
	Spring	-0.02 (-0.16 to 0.09)	-4.1 (-37.3 to 15.5)	0.60 (0.46 to 0.98)	73.7 (66.1 to 86.9)	0.68 (0.51 to 0.75)
	Summer	-0.08 (-0.12 to 0.04)	-40.6 (-61.1 to 17.6)	0.35 (0.24 to 0.67)	86.5 (80.5 to 114.1)	0.37 (0.17 to 0.51)
	Fall	0.10 (0.03 to 0.25)	37.0 (10.4 to 67.6)	0.82 (0.52 to 1.26)	102.8 (91.3 to 129.7)	0.57 (0.45 to 0.69)
	All	0.03 (-0.07 to 0.11)	6.6 (-17.3 to 22.0)	0.83 (0.60 to 1.09)	81.2 (70.9 to 89.0)	0.66 (0.56 to 0.72)

Table S6. Median (range) of performance statistics for PM_{2.5} OC at CSN sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	1.76 (0.84 to 3.69)	116.1 (44.8 to 231.7)	2.46 (1.91 to 6.49)	122.4 (62.9 to 233.9)	0.61 (0.38 to 0.76)
	Spring	0.38 (-0.44 to 2.15)	24.3 (-24.5 to 192.8)	1.58 (1.15 to 4.60)	59.8 (48.4 to 199.4)	0.49 (0.34 to 0.64)
	Summer	-0.73 (-2.06 to 1.43)	-33.7 (-62.7 to 72.8)	1.92 (0.91 to 4.76)	42.0 (34.5 to 100.6)	0.49 (0.19 to 0.67)
	Fall	0.26 (-0.21 to 2.25)	17.4 (-10.0 to 183.3)	1.77 (1.01 to 5.45)	56.0 (38.7 to 188.6)	0.66 (0.31 to 0.74)
	All	0.40 (-0.47 to 2.27)	24.3 (-20.2 to 151.5)	1.90 (1.55 to 5.37)	62.6 (47.5 to 164.9)	0.46 (0.26 to 0.52)
SE	Winter	0.44 (-0.34 to 1.61)	16.6 (-13.1 to 78.1)	2.19 (1.73 to 3.14)	56.4 (43.7 to 93.7)	0.52 (0.37 to 0.64)
	Spring	0.00 (-1.31 to 0.96)	0.2 (-37.4 to 53.0)	1.68 (1.28 to 4.59)	50.9 (44.8 to 69.0)	0.53 (0.36 to 0.79)
	Summer	-0.23 (-1.96 to 1.00)	-7.6 (-56.2 to 53.4)	1.94 (0.89 to 2.54)	52.8 (36.7 to 74.2)	0.50 (0.35 to 0.62)
	Fall	-0.01 (-0.89 to 1.29)	-0.4 (-38.3 to 68.2)	1.93 (1.21 to 2.45)	51.8 (39.0 to 86.7)	0.59 (0.46 to 0.68)
	All	-0.05 (-0.97 to 1.14)	-1.9 (-36.0 to 61.9)	2.12 (1.52 to 3.06)	55.0 (42.5 to 78.9)	0.51 (0.42 to 0.65)
Ohio Valley	Winter	0.99 (0.22 to 2.32)	54.7 (11.7 to 145.4)	1.64 (1.21 to 3.37)	70.1 (38.0 to 152.3)	0.54 (0.41 to 0.63)
	Spring	0.23 (-0.72 to 0.51)	15.4 (-35.6 to 34.7)	1.35 (1.10 to 2.13)	52.0 (39.0 to 60.1)	0.49 (0.40 to 0.69)
	Summer	-0.64 (-1.63 to 0.06)	-30.2 (-57.7 to 3.3)	1.35 (0.88 to 2.07)	40.7 (30.7 to 59.8)	0.48 (0.36 to 0.61)
	Fall	0.21 (-0.59 to 1.06)	9.7 (-29.2 to 57.0)	1.56 (0.89 to 2.03)	43.7 (39.1 to 67.3)	0.63 (0.41 to 0.72)
	All	0.24 (-0.58 to 0.84)	12.5 (-26.0 to 48.6)	1.65 (1.18 to 1.95)	53.9 (39.2 to 69.8)	0.46 (0.42 to 0.54)
MW	Winter	1.37 (0.58 to 2.51)	118.6 (33.9 to 199.3)	2.38 (1.69 to 3.39)	121.9 (59.1 to 203.1)	0.52 (0.25 to 0.66)
	Spring	0.33 (-0.39 to 1.10)	24.5 (-25.1 to 100.1)	1.34 (0.91 to 1.83)	69.0 (43.0 to 112.4)	0.48 (0.33 to 0.74)
	Summer	-0.80 (-1.87 to 0.22)	-33.2 (-67.5 to 11.8)	1.39 (1.10 to 2.60)	43.3 (38.3 to 68.0)	0.48 (0.33 to 0.55)
	Fall	0.05 (-0.42 to 1.18)	3.0 (-23.8 to 85.2)	1.38 (0.88 to 2.05)	54.7 (38.7 to 90.9)	0.61 (0.52 to 0.73)
	All	0.28 (-0.52 to 1.19)	17.8 (-27.5 to 84.7)	1.76 (1.44 to 2.18)	65.4 (45.1 to 101.9)	0.39 (0.23 to 0.44)
South	Winter	0.45 (-0.45 to 1.35)	25.9 (-22.6 to 85.7)	1.85 (1.36 to 4.07)	61.2 (41.1 to 109.3)	0.44 (0.20 to 0.62)
	Spring	-0.10 (-1.23 to 0.09)	-5.0 (-58.4 to 5.3)	1.35 (1.07 to 3.89)	49.7 (42.1 to 62.2)	0.49 (0.18 to 0.63)
	Summer	-0.18 (-1.33 to 0.44)	-11.1 (-56.5 to 29.9)	1.36 (0.79 to 2.27)	47.5 (36.2 to 67.1)	0.58 (0.20 to 0.65)
	Fall	0.44 (-1.15 to 0.72)	21.7 (-39.7 to 41.7)	1.61 (1.26 to 6.24)	55.7 (37.4 to 73.7)	0.53 (0.18 to 0.69)
	All	0.09 (-1.00 to 0.56)	4.1 (-45.3 to 32.1)	1.75 (1.35 to 4.14)	59.2 (40.0 to 64.7)	0.51 (0.23 to 0.63)
SW	Winter	1.26 (-0.84 to 3.73)	56.6 (-24.3 to 202.7)	2.63 (2.00 to 5.39)	77.4 (49.9 to 213.9)	0.48 (0.16 to 0.55)
	Spring	0.71 (-0.47 to 1.66)	93.8 (-26.0 to 256.5)	1.21 (0.95 to 2.24)	112.1 (46.8 to 261.0)	0.46 (0.33 to 0.60)
	Summer	0.08 (-0.87 to 0.30)	5.9 (-44.2 to 26.8)	1.54 (0.65 to 1.74)	51.9 (42.2 to 61.5)	0.50 (0.28 to 0.72)
	Fall	0.72 (-0.35 to 1.87)	57.7 (-14.3 to 140.2)	1.46 (1.16 to 3.03)	68.4 (39.1 to 146.6)	0.54 (0.38 to 0.63)
	All	0.76 (-0.49 to 1.83)	59.2 (-20.2 to 135.0)	1.73 (1.35 to 3.34)	77.0 (46.7 to 151.2)	0.55 (0.30 to 0.63)
NRP	Winter	-0.40 (-1.79 to 0.63)	-24.1 (-61.9 to 61.4)	4.22 (1.95 to 4.51)	94.3 (70.2 to 130.6)	-0.00 (-0.16 to 0.22)
	Spring	-0.32 (-1.01 to 0.15)	-22.8 (-61.3 to 17.3)	1.23 (0.66 to 2.12)	64.3 (56.8 to 76.6)	0.39 (-0.05 to 0.80)
	Summer	-0.86 (-1.77 to 0.17)	-49.5 (-69.3 to 4.6)	1.84 (0.84 to 6.89)	58.1 (36.2 to 76.0)	0.60 (0.21 to 0.94)
	Fall	-0.46 (-1.13 to 0.23)	-29.7 (-55.8 to 14.9)	1.64 (1.22 to 3.54)	60.3 (55.0 to 67.7)	0.36 (0.20 to 0.76)
	All	-0.48 (-1.25 to 0.02)	-31.4 (-58.8 to 1.7)	2.24 (1.53 to 4.56)	70.2 (61.2 to 75.0)	0.25 (0.06 to 0.80)
NW	Winter	2.26 (-1.69 to 6.05)	71.6 (-40.0 to 173.6)	5.22 (3.52 to 8.83)	101.5 (71.5 to 186.7)	0.41 (0.17 to 0.57)
	Spring	1.88 (-0.53 to 3.98)	142.1 (-26.9 to 505.3)	3.99 (1.85 to 5.34)	165.0 (63.1 to 505.8)	0.41 (0.23 to 0.67)
	Summer	1.21 (-0.63 to 2.61)	66.4 (-23.7 to 257.2)	3.53 (0.68 to 5.40)	102.6 (41.1 to 268.3)	0.51 (0.16 to 0.68)
	Fall	2.05 (-0.31 to 4.23)	77.4 (-10.7 to 167.4)	4.97 (2.53 to 7.70)	104.5 (63.7 to 173.6)	0.45 (0.21 to 0.55)
	All	1.58 (-0.78 to 4.25)	76.5 (-23.4 to 195.6)	4.56 (2.39 to 6.48)	116.7 (66.9 to 206.3)	0.41 (0.35 to 0.54)
West	Winter	-0.08 (-1.51 to 1.34)	-2.1 (-30.2 to 41.3)	3.29 (1.90 to 4.89)	56.6 (32.1 to 64.8)	0.54 (0.13 to 0.69)
	Spring	-0.03 (-0.42 to 1.05)	-1.7 (-18.8 to 80.2)	1.38 (0.82 to 1.72)	51.6 (31.1 to 89.9)	0.48 (0.24 to 0.78)
	Summer	-0.07 (-0.94 to 0.28)	-4.2 (-34.0 to 17.6)	1.47 (0.85 to 4.49)	44.1 (37.6 to 52.6)	0.51 (0.26 to 0.90)
	Fall	0.10 (-0.75 to 1.04)	4.1 (-18.7 to 44.0)	2.20 (1.46 to 3.56)	46.8 (33.5 to 61.6)	0.59 (0.34 to 0.89)
	All	-0.10 (-0.90 to 0.91)	-2.6 (-24.1 to 42.6)	2.14 (1.62 to 3.39)	52.1 (33.6 to 61.6)	0.67 (0.30 to 0.74)
All	Winter	0.89 (0.02 to 2.56)	43.3 (0.7 to 130.7)	2.77 (2.13 to 4.28)	79.0 (51.6 to 144.4)	0.46 (0.32 to 0.57)
	Spring	0.40 (-0.62 to 0.95)	25.3 (-31.2 to 68.7)	1.66 (1.51 to 2.71)	59.8 (46.6 to 91.6)	0.43 (0.23 to 0.64)
	Summer	-0.43 (-1.53 to 0.53)	-23.6 (-50.5 to 28.9)	1.90 (1.13 to 2.72)	47.5 (41.2 to 66.8)	0.49 (0.26 to 0.64)
	Fall	0.16 (-0.51 to 1.23)	8.1 (-24.0 to 70.6)	2.20 (1.57 to 3.27)	56.0 (41.8 to 87.6)	0.56 (0.36 to 0.62)
	All	0.24 (-0.64 to 1.13)	13.2 (-25.3 to 64.5)	2.34 (1.74 to 3.05)	58.4 (45.4 to 88.8)	0.47 (0.35 to 0.55)

Table S7. Median (range) of performance statistics for PM_{2.5} OC at IMPROVE sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	0.75 (0.24 to 1.16)	76.2 (23.1 to 130.3)	1.54 (1.11 to 1.96)	89.4 (61.6 to 137.5)	0.62 (0.47 to 0.78)
	Spring	0.18 (-0.25 to 0.37)	21.2 (-28.1 to 58.7)	0.86 (0.67 to 1.35)	59.3 (53.4 to 81.2)	0.54 (0.26 to 0.65)
	Summer	-0.60 (-1.03 to -0.30)	-41.0 (-61.9 to -22.3)	0.96 (0.55 to 1.61)	48.3 (36.3 to 67.4)	0.56 (0.33 to 0.74)
	Fall	0.11 (-0.18 to 0.36)	11.3 (-19.2 to 47.0)	0.84 (0.51 to 1.05)	54.0 (41.6 to 71.3)	0.67 (0.52 to 0.73)
	All	0.15 (-0.25 to 0.35)	13.6 (-22.1 to 39.2)	1.13 (0.91 to 1.35)	62.9 (55.4 to 75.0)	0.49 (0.32 to 0.61)
SE	Winter	0.55 (-0.06 to 0.99)	40.2 (-4.3 to 77.9)	2.18 (1.50 to 2.93)	72.4 (66.3 to 97.4)	0.44 (0.36 to 0.62)
	Spring	-0.18 (-1.17 to 0.74)	-12.6 (-45.0 to 54.3)	2.24 (0.96 to 5.73)	56.9 (50.4 to 83.1)	0.46 (0.07 to 0.73)
	Summer	-0.63 (-1.23 to 0.36)	-33.1 (-58.7 to 24.5)	1.46 (0.87 to 2.33)	55.1 (45.6 to 71.2)	0.60 (0.49 to 0.73)
	Fall	-0.00 (-0.50 to 0.64)	-0.3 (-40.6 to 47.2)	1.30 (0.85 to 2.27)	55.3 (41.5 to 76.6)	0.68 (0.33 to 0.73)
	All	-0.14 (-0.56 to 0.59)	-7.8 (-32.6 to 44.1)	2.03 (1.16 to 3.26)	59.8 (51.7 to 76.9)	0.44 (0.19 to 0.64)
Ohio Valley	Winter	0.39 (-0.16 to 0.69)	41.6 (-15.3 to 64.4)	1.05 (0.61 to 1.43)	65.6 (36.5 to 84.7)	0.54 (0.43 to 0.73)
	Spring	-0.28 (-0.60 to 0.44)	-19.8 (-47.5 to 37.1)	1.07 (0.61 to 6.33)	53.0 (42.0 to 77.4)	0.58 (0.12 to 0.69)
	Summer	-0.56 (-1.05 to -0.01)	-29.9 (-56.3 to -1.0)	0.99 (0.74 to 1.76)	44.3 (35.5 to 58.3)	0.59 (0.34 to 0.75)
	Fall	-0.01 (-0.56 to 0.43)	-0.6 (-43.5 to 36.3)	1.10 (0.69 to 1.24)	45.5 (40.2 to 58.0)	0.68 (0.55 to 0.80)
	All	-0.05 (-0.51 to 0.28)	-3.4 (-37.8 to 24.3)	1.12 (0.78 to 3.38)	50.4 (45.7 to 57.6)	0.54 (0.21 to 0.58)
MW	Winter	0.44 (-0.07 to 0.86)	66.2 (-9.0 to 135.8)	0.75 (0.33 to 1.35)	73.6 (35.3 to 141.0)	0.76 (0.50 to 0.85)
	Spring	0.04 (-0.50 to 0.29)	4.5 (-53.8 to 34.3)	0.97 (0.56 to 3.14)	58.8 (51.1 to 70.7)	0.53 (0.11 to 0.67)
	Summer	-0.67 (-1.16 to -0.39)	-42.9 (-75.0 to -29.1)	1.10 (0.84 to 1.68)	53.5 (43.7 to 75.5)	0.43 (0.16 to 0.65)
	Fall	-0.03 (-0.45 to 0.40)	-4.3 (-52.1 to 50.3)	0.63 (0.52 to 5.14)	46.2 (42.3 to 76.0)	0.71 (0.60 to 0.82)
	All	-0.04 (-0.45 to 0.19)	-3.9 (-46.6 to 21.3)	1.02 (0.95 to 2.72)	57.4 (50.9 to 70.4)	0.38 (0.18 to 0.68)
South	Winter	0.24 (-0.24 to 1.46)	32.8 (-33.2 to 192.7)	1.26 (0.61 to 16.61)	63.9 (46.7 to 227.6)	0.57 (0.29 to 0.72)
	Spring	-0.40 (-0.81 to -0.22)	-31.2 (-66.6 to -20.2)	1.21 (0.69 to 5.52)	57.1 (47.2 to 69.8)	0.66 (0.22 to 0.74)
	Summer	-0.47 (-0.76 to -0.15)	-31.8 (-58.2 to -12.8)	0.84 (0.61 to 1.01)	49.2 (41.7 to 60.8)	0.65 (0.57 to 0.69)
	Fall	-0.08 (-0.51 to 0.24)	-8.1 (-50.5 to 18.3)	0.91 (0.54 to 2.27)	49.5 (37.5 to 65.9)	0.66 (0.32 to 0.79)
	All	-0.19 (-0.51 to 0.18)	-15.5 (-47.9 to 17.3)	1.03 (0.81 to 7.89)	52.0 (47.4 to 79.6)	0.53 (0.18 to 0.64)
SW	Winter	-0.04 (-0.21 to 0.13)	-8.0 (-34.6 to 25.5)	0.64 (0.50 to 0.97)	54.9 (52.0 to 72.9)	0.80 (0.62 to 0.87)
	Spring	-0.11 (-0.37 to 0.03)	-25.8 (-59.7 to 8.3)	0.55 (0.32 to 1.56)	55.3 (43.3 to 71.2)	0.62 (0.19 to 0.73)
	Summer	-0.33 (-0.65 to -0.10)	-36.6 (-58.9 to -17.6)	0.85 (0.44 to 5.10)	59.8 (48.7 to 73.1)	0.44 (0.16 to 0.76)
	Fall	-0.06 (-0.29 to 0.04)	-9.7 (-36.7 to 6.2)	0.99 (0.66 to 3.90)	57.3 (51.2 to 73.1)	0.47 (0.10 to 0.72)
	All	-0.12 (-0.34 to -0.05)	-19.4 (-45.1 to -8.3)	1.18 (0.54 to 2.71)	57.2 (50.4 to 69.0)	0.55 (0.21 to 0.72)
NRP	Winter	-0.02 (-0.16 to 0.09)	-6.4 (-40.9 to 29.0)	0.39 (0.26 to 0.82)	61.8 (52.8 to 88.2)	0.26 (0.13 to 0.48)
	Spring	-0.12 (-0.47 to -0.07)	-30.6 (-76.9 to -15.6)	0.59 (0.33 to 2.21)	64.1 (50.5 to 88.5)	0.57 (0.07 to 0.97)
	Summer	-0.51 (-0.72 to 0.69)	-37.9 (-68.5 to 25.5)	1.80 (1.17 to 13.66)	64.8 (58.0 to 91.7)	0.49 (0.24 to 0.77)
	Fall	-0.16 (-0.44 to 0.32)	-14.6 (-52.5 to 27.7)	1.87 (0.68 to 5.45)	62.5 (53.3 to 88.0)	0.62 (0.26 to 0.78)
	All	-0.24 (-0.38 to 0.19)	-33.4 (-56.1 to 15.4)	1.46 (1.00 to 7.16)	64.6 (55.5 to 85.1)	0.60 (0.23 to 0.77)
NW	Winter	0.31 (0.10 to 0.40)	61.7 (17.5 to 98.5)	1.68 (1.42 to 2.51)	127.3 (99.9 to 162.7)	0.61 (0.18 to 0.69)
	Spring	0.23 (-0.21 to 0.45)	39.0 (-32.6 to 141.7)	1.92 (0.79 to 2.29)	119.5 (75.3 to 186.1)	0.53 (0.31 to 0.60)
	Summer	0.10 (-0.65 to 0.58)	7.2 (-45.9 to 60.7)	2.95 (1.17 to 10.21)	89.3 (61.3 to 137.4)	0.44 (0.31 to 0.71)
	Fall	0.67 (0.01 to 1.62)	46.2 (1.8 to 152.4)	3.65 (1.34 to 23.81)	103.5 (73.6 to 224.7)	0.49 (0.14 to 0.58)
	All	0.32 (-0.08 to 0.71)	31.8 (-9.4 to 65.4)	2.67 (1.42 to 13.08)	107.0 (78.8 to 127.7)	0.47 (0.30 to 0.56)
West	Winter	0.01 (-0.10 to 0.31)	1.7 (-15.4 to 35.1)	0.71 (0.54 to 2.63)	59.2 (48.5 to 81.5)	0.83 (0.56 to 0.90)
	Spring	-0.24 (-0.42 to 0.04)	-32.3 (-50.3 to 8.0)	0.55 (0.45 to 1.53)	54.8 (52.2 to 58.4)	0.59 (0.20 to 0.65)
	Summer	-0.27 (-0.87 to 0.60)	-21.4 (-47.8 to 21.3)	2.32 (0.64 to 4.82)	64.7 (42.2 to 75.1)	0.65 (0.23 to 0.84)
	Fall	0.12 (-0.29 to 0.32)	10.2 (-24.0 to 28.1)	3.54 (1.86 to 4.78)	71.7 (62.2 to 90.5)	0.37 (0.20 to 0.75)
	All	-0.07 (-0.36 to 0.18)	-6.6 (-33.1 to 12.7)	2.45 (1.38 to 3.29)	63.7 (54.9 to 74.9)	0.55 (0.28 to 0.73)
All	Winter	0.27 (0.01 to 0.37)	44.0 (1.2 to 54.9)	1.30 (1.05 to 4.21)	82.0 (62.8 to 88.9)	0.58 (0.28 to 0.68)
	Spring	-0.03 (-0.45 to 0.11)	-4.4 (-49.3 to 16.4)	1.23 (0.96 to 2.62)	61.7 (59.6 to 72.0)	0.45 (0.13 to 0.52)
	Summer	-0.30 (-0.71 to -0.09)	-23.3 (-55.1 to -5.4)	2.17 (0.98 to 6.37)	63.3 (52.7 to 69.7)	0.55 (0.21 to 0.70)
	Fall	0.10 (-0.30 to 0.23)	11.2 (-31.3 to 22.0)	2.49 (1.34 to 8.14)	65.3 (52.2 to 79.3)	0.50 (0.17 to 0.67)
	All	-0.05 (-0.31 to 0.09)	-4.7 (-32.6 to 9.8)	1.93 (1.25 to 5.74)	66.3 (56.7 to 72.1)	0.48 (0.28 to 0.60)

Table S8. Median (range) of performance statistics for PM_{2.5} EC at CSN sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
NE	Winter	0.32 (0.25 to 0.78)	55.3 (33.0 to 110.4)	0.78 (0.61 to 1.24)	71.8 (61.2 to 121.3)	0.63 (0.52 to 0.69)
	Spring	0.15 (0.08 to 0.36)	27.4 (12.2 to 75.9)	0.60 (0.46 to 0.71)	59.1 (49.9 to 94.4)	0.61 (0.48 to 0.66)
	Summer	0.13 (-0.07 to 0.25)	21.3 (-10.5 to 37.4)	0.60 (0.46 to 0.76)	51.4 (40.1 to 64.8)	0.53 (0.40 to 0.58)
	Fall	0.21 (-0.07 to 0.44)	29.4 (-7.1 to 61.9)	0.80 (0.56 to 1.03)	57.1 (47.9 to 78.2)	0.58 (0.35 to 0.76)
	All	0.19 (0.09 to 0.44)	28.4 (11.6 to 66.4)	0.71 (0.53 to 0.90)	58.3 (53.1 to 84.8)	0.56 (0.50 to 0.65)
SE	Winter	0.26 (0.10 to 0.52)	42.3 (13.2 to 73.0)	0.60 (0.50 to 0.90)	64.6 (50.6 to 90.3)	0.50 (0.41 to 0.61)
	Spring	0.16 (-0.04 to 0.34)	33.5 (-6.0 to 60.6)	0.49 (0.36 to 0.59)	63.3 (41.8 to 79.2)	0.54 (0.44 to 0.73)
	Summer	0.14 (-0.02 to 0.47)	26.9 (-3.0 to 104.3)	0.45 (0.33 to 0.74)	60.6 (48.9 to 121.1)	0.41 (0.17 to 0.53)
	Fall	0.23 (-0.14 to 0.50)	36.0 (-16.3 to 82.3)	0.57 (0.45 to 0.87)	57.4 (45.8 to 95.6)	0.58 (0.33 to 0.65)
	All	0.19 (-0.02 to 0.44)	32.6 (-2.9 to 70.2)	0.53 (0.44 to 0.77)	57.9 (47.6 to 88.2)	0.52 (0.43 to 0.62)
Ohio Valley	Winter	0.35 (0.12 to 0.71)	67.7 (18.5 to 126.1)	0.67 (0.45 to 0.99)	87.5 (56.1 to 134.2)	0.48 (0.38 to 0.56)
	Spring	0.13 (-0.09 to 0.37)	25.9 (-13.0 to 65.2)	0.44 (0.40 to 0.64)	55.7 (41.8 to 81.6)	0.51 (0.38 to 0.67)
	Summer	0.07 (-0.07 to 0.38)	11.0 (-9.9 to 49.3)	0.42 (0.37 to 0.74)	48.5 (39.8 to 68.6)	0.52 (0.44 to 0.63)
	Fall	0.18 (-0.12 to 0.47)	27.8 (-13.6 to 70.3)	0.63 (0.41 to 0.82)	52.7 (45.0 to 83.1)	0.61 (0.32 to 0.66)
	All	0.19 (-0.05 to 0.48)	34.6 (-6.2 to 72.4)	0.54 (0.43 to 0.79)	61.5 (45.4 to 86.7)	0.54 (0.43 to 0.57)
MW	Winter	0.38 (0.21 to 0.70)	92.8 (45.5 to 173.8)	0.62 (0.48 to 0.99)	103.5 (71.3 to 176.3)	0.56 (0.44 to 0.71)
	Spring	0.20 (-0.02 to 0.46)	48.9 (-3.3 to 112.0)	0.41 (0.32 to 0.76)	67.1 (48.6 to 119.0)	0.53 (0.42 to 0.75)
	Summer	0.09 (-0.01 to 0.31)	16.6 (-1.7 to 60.0)	0.39 (0.30 to 0.62)	49.8 (40.1 to 72.1)	0.58 (0.53 to 0.67)
	Fall	0.18 (-0.04 to 0.51)	34.2 (-6.2 to 96.9)	0.46 (0.34 to 0.75)	54.9 (45.8 to 99.7)	0.72 (0.44 to 0.76)
	All	0.22 (0.03 to 0.49)	45.3 (6.4 to 101.4)	0.46 (0.40 to 0.79)	65.9 (51.9 to 107.8)	0.56 (0.45 to 0.66)
South	Winter	0.25 (0.13 to 0.52)	41.9 (24.4 to 83.2)	0.77 (0.47 to 1.38)	77.9 (57.9 to 137.5)	0.39 (0.10 to 0.67)
	Spring	0.21 (0.11 to 0.34)	49.2 (27.1 to 68.9)	0.46 (0.32 to 1.02)	69.2 (48.8 to 109.5)	0.53 (0.17 to 0.65)
	Summer	0.28 (0.12 to 0.50)	66.3 (29.4 to 106.6)	0.59 (0.27 to 1.08)	90.9 (52.7 to 136.7)	0.43 (0.28 to 0.56)
	Fall	0.22 (0.16 to 0.52)	40.5 (27.4 to 79.1)	0.66 (0.33 to 1.66)	69.4 (49.3 to 123.0)	0.60 (0.12 to 0.64)
	All	0.27 (0.15 to 0.46)	45.6 (29.8 to 82.0)	0.64 (0.35 to 1.31)	76.6 (54.1 to 126.0)	0.45 (0.16 to 0.65)
SW	Winter	0.30 (-0.41 to 0.74)	31.8 (-32.0 to 70.4)	1.03 (0.69 to 1.34)	65.8 (50.2 to 84.5)	0.49 (0.17 to 0.64)
	Spring	0.29 (-0.08 to 0.63)	73.2 (-12.5 to 179.4)	0.58 (0.42 to 1.03)	89.7 (52.7 to 180.7)	0.56 (0.02 to 0.72)
	Summer	0.25 (-0.06 to 0.48)	64.6 (-9.6 to 104.7)	0.45 (0.42 to 0.74)	80.2 (56.8 to 120.4)	0.36 (0.30 to 0.69)
	Fall	0.25 (-0.53 to 0.68)	37.1 (-41.8 to 93.3)	0.71 (0.46 to 1.26)	61.3 (53.9 to 100.8)	0.57 (0.14 to 0.66)
	All	0.28 (-0.28 to 0.63)	52.8 (-29.0 to 98.0)	0.82 (0.52 to 0.97)	71.4 (57.6 to 108.2)	0.58 (0.23 to 0.70)
NRP	Winter	0.02 (-0.36 to 0.18)	4.2 (-48.5 to 59.4)	0.74 (0.56 to 1.15)	108.6 (59.9 to 124.4)	0.01 (-0.14 to 0.21)
	Spring	0.06 (-0.15 to 0.11)	21.8 (-34.5 to 47.0)	0.32 (0.28 to 0.49)	80.4 (51.9 to 89.7)	0.45 (0.05 to 0.73)
	Summer	0.04 (-0.02 to 0.25)	11.8 (-6.1 to 61.2)	0.35 (0.25 to 1.43)	69.8 (44.7 to 85.9)	0.62 (0.29 to 0.92)
	Fall	-0.01 (-0.28 to 0.24)	-2.7 (-33.4 to 58.8)	0.49 (0.38 to 0.91)	74.2 (59.7 to 101.3)	0.24 (-0.02 to 0.45)
	All	0.02 (-0.12 to 0.16)	5.3 (-19.1 to 49.2)	0.53 (0.42 to 0.95)	82.5 (62.9 to 94.2)	0.22 (0.03 to 0.65)
NW	Winter	0.76 (0.16 to 1.24)	74.0 (17.0 to 123.9)	1.45 (0.95 to 2.19)	102.4 (69.0 to 137.4)	0.51 (0.39 to 0.58)
	Spring	0.77 (0.28 to 1.28)	165.3 (57.1 to 360.8)	1.50 (0.82 to 2.14)	184.7 (104.7 to 361.3)	0.46 (0.35 to 0.57)
	Summer	1.02 (0.57 to 1.71)	280.1 (131.0 to 433.7)	2.15 (0.79 to 3.33)	290.2 (143.2 to 435.2)	0.44 (0.30 to 0.55)
	Fall	0.93 (0.47 to 1.58)	104.9 (49.7 to 171.8)	1.85 (1.24 to 2.55)	125.5 (95.6 to 179.5)	0.34 (0.26 to 0.51)
	All	0.78 (0.41 to 1.44)	124.4 (57.2 to 180.5)	1.69 (1.15 to 2.46)	150.5 (100.7 to 193.3)	0.39 (0.28 to 0.52)
West	Winter	0.11 (-0.27 to 0.40)	9.2 (-20.3 to 28.4)	0.88 (0.58 to 1.20)	47.1 (39.2 to 64.0)	0.50 (0.32 to 0.63)
	Spring	0.32 (0.10 to 0.52)	58.2 (20.8 to 109.3)	0.57 (0.43 to 0.83)	80.7 (58.2 to 114.2)	0.57 (0.30 to 0.69)
	Summer	0.32 (0.19 to 0.89)	66.8 (45.1 to 119.2)	0.52 (0.35 to 1.60)	79.6 (66.8 to 130.3)	0.65 (0.48 to 0.84)
	Fall	0.29 (0.02 to 0.50)	34.6 (2.5 to 53.6)	0.71 (0.48 to 1.25)	57.2 (45.2 to 68.0)	0.53 (0.42 to 0.79)
	All	0.27 (0.04 to 0.49)	37.5 (4.6 to 52.4)	0.66 (0.50 to 1.17)	64.4 (48.8 to 79.2)	0.56 (0.45 to 0.72)
All	Winter	0.27 (0.18 to 0.65)	43.0 (25.3 to 87.0)	0.80 (0.66 to 1.13)	72.0 (68.0 to 104.6)	0.48 (0.46 to 0.59)
	Spring	0.20 (0.10 to 0.43)	38.8 (15.2 to 89.1)	0.59 (0.47 to 0.79)	68.3 (60.1 to 104.5)	0.48 (0.41 to 0.61)
	Summer	0.19 (0.10 to 0.41)	35.9 (18.9 to 71.7)	0.71 (0.51 to 0.94)	65.6 (55.2 to 90.2)	0.43 (0.32 to 0.51)
	Fall	0.23 (0.03 to 0.53)	34.1 (3.6 to 78.3)	0.69 (0.63 to 1.08)	62.5 (54.8 to 90.6)	0.56 (0.28 to 0.61)
	All	0.22 (0.12 to 0.50)	33.6 (16.4 to 78.9)	0.68 (0.61 to 0.96)	65.6 (61.2 to 95.0)	0.51 (0.40 to 0.54)

Table S9. Median (range) of performance statistics for PM_{2.5} EC at IMPROVE sites for years 2007-2015.

Region	Season	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	r
NE	Winter	0.12 (0.09 to 0.22)	48.5 (26.2 to 117.8)	0.37 (0.26 to 0.50)	69.3 (51.1 to 125.3)	0.78 (0.63 to 0.85)
	Spring	0.09 (-0.00 to 0.11)	44.5 (-0.3 to 71.4)	0.21 (0.16 to 0.40)	68.6 (49.8 to 84.5)	0.76 (0.53 to 0.88)
	Summer	0.00 (-0.08 to 0.09)	1.0 (-22.0 to 44.8)	0.18 (0.13 to 0.36)	45.9 (37.9 to 66.4)	0.76 (0.65 to 0.87)
	Fall	0.09 (0.01 to 0.12)	40.1 (4.1 to 57.6)	0.21 (0.13 to 0.44)	63.3 (40.3 to 73.4)	0.83 (0.65 to 0.86)
	All	0.07 (0.02 to 0.12)	36.9 (5.6 to 60.1)	0.24 (0.20 to 0.42)	61.6 (51.7 to 75.8)	0.77 (0.64 to 0.83)
SE	Winter	0.17 (0.04 to 0.31)	48.1 (6.9 to 75.3)	0.46 (0.39 to 0.70)	73.0 (50.6 to 89.1)	0.65 (0.63 to 0.74)
	Spring	0.06 (-0.22 to 0.19)	15.1 (-32.0 to 58.5)	0.49 (0.24 to 0.73)	54.8 (44.2 to 79.4)	0.61 (0.48 to 0.84)
	Summer	0.01 (-0.17 to 0.17)	4.6 (-34.4 to 64.1)	0.32 (0.19 to 0.52)	49.8 (41.2 to 88.0)	0.79 (0.72 to 0.82)
	Fall	0.10 (-0.03 to 0.25)	32.0 (-5.5 to 67.3)	0.35 (0.21 to 0.56)	54.3 (38.2 to 78.9)	0.77 (0.75 to 0.87)
	All	0.09 (-0.10 to 0.17)	31.5 (-17.9 to 52.2)	0.40 (0.30 to 0.54)	60.1 (48.2 to 75.0)	0.69 (0.64 to 0.75)
Ohio Valley	Winter	0.13 (-0.03 to 0.22)	50.6 (-7.3 to 68.1)	0.25 (0.16 to 0.34)	64.4 (35.8 to 76.4)	0.62 (0.43 to 0.69)
	Spring	0.04 (-0.13 to 0.12)	19.0 (-26.9 to 53.6)	0.21 (0.15 to 1.28)	46.8 (32.7 to 83.4)	0.56 (0.12 to 0.66)
	Summer	-0.06 (-0.16 to 0.04)	-18.3 (-37.2 to 15.4)	0.15 (0.13 to 0.30)	37.2 (32.0 to 50.3)	0.55 (0.29 to 0.75)
	Fall	0.08 (-0.02 to 0.19)	29.0 (-5.2 to 64.2)	0.23 (0.17 to 0.31)	50.2 (35.5 to 73.6)	0.68 (0.48 to 0.75)
	All	0.05 (-0.08 to 0.10)	16.8 (-20.2 to 37.6)	0.25 (0.18 to 0.68)	50.1 (38.0 to 63.1)	0.54 (0.20 to 0.64)
MW	Winter	0.13 (0.02 to 0.16)	78.1 (9.0 to 101.4)	0.19 (0.11 to 0.27)	84.7 (36.5 to 113.0)	0.77 (0.52 to 0.85)
	Spring	0.04 (-0.10 to 0.08)	18.1 (-34.8 to 50.8)	0.21 (0.09 to 0.63)	59.9 (36.5 to 70.8)	0.68 (0.27 to 0.76)
	Summer	-0.04 (-0.10 to 0.07)	-24.0 (-30.8 to 26.7)	0.14 (0.10 to 0.27)	41.3 (38.0 to 52.1)	0.66 (0.52 to 0.81)
	Fall	0.04 (-0.02 to 0.23)	21.3 (-10.5 to 94.1)	0.16 (0.11 to 1.22)	45.3 (34.1 to 108.6)	0.74 (0.71 to 0.84)
	All	0.04 (-0.05 to 0.12)	19.1 (-18.8 to 63.4)	0.21 (0.13 to 0.63)	57.9 (39.3 to 84.2)	0.64 (0.46 to 0.73)
South	Winter	0.08 (0.02 to 0.36)	49.7 (9.1 to 241.2)	0.30 (0.12 to 3.54)	77.4 (38.3 to 257.2)	0.55 (0.41 to 0.75)
	Spring	-0.01 (-0.07 to 0.03)	-4.1 (-32.1 to 10.3)	0.18 (0.12 to 0.81)	46.5 (40.6 to 63.8)	0.68 (0.18 to 0.82)
	Summer	-0.02 (-0.03 to 0.03)	-11.1 (-15.8 to 18.5)	0.11 (0.09 to 0.18)	45.2 (38.4 to 50.5)	0.60 (0.44 to 0.75)
	Fall	0.06 (-0.01 to 0.10)	27.2 (-6.3 to 47.5)	0.21 (0.13 to 0.48)	52.0 (36.7 to 65.4)	0.69 (0.21 to 0.82)
	All	0.02 (-0.01 to 0.10)	11.2 (-6.7 to 68.0)	0.19 (0.16 to 1.68)	52.7 (41.1 to 97.4)	0.53 (0.26 to 0.74)
SW	Winter	0.02 (-0.07 to 0.08)	14.9 (-32.5 to 49.7)	0.28 (0.17 to 0.39)	58.7 (50.0 to 81.5)	0.81 (0.78 to 0.87)
	Spring	0.04 (-0.03 to 0.07)	52.6 (-23.2 to 77.7)	0.24 (0.10 to 0.33)	83.3 (61.7 to 109.8)	0.73 (0.43 to 0.80)
	Summer	0.02 (-0.04 to 0.08)	20.2 (-30.0 to 65.4)	0.21 (0.14 to 0.53)	82.5 (58.4 to 111.3)	0.48 (0.16 to 0.78)
	Fall	0.04 (-0.03 to 0.10)	29.4 (-15.7 to 62.5)	0.37 (0.21 to 0.45)	79.6 (63.5 to 90.3)	0.68 (0.36 to 0.76)
	All	0.03 (-0.04 to 0.07)	29.1 (-25.5 to 52.0)	0.26 (0.18 to 0.38)	71.1 (60.8 to 93.2)	0.66 (0.53 to 0.78)
NRP	Winter	0.01 (0.00 to 0.05)	14.1 (0.6 to 92.3)	0.09 (0.08 to 0.19)	67.2 (54.1 to 131.1)	0.42 (0.20 to 0.54)
	Spring	-0.00 (-0.04 to 0.06)	-2.7 (-35.9 to 96.1)	0.07 (0.06 to 0.46)	48.4 (42.8 to 124.2)	0.57 (0.09 to 0.96)
	Summer	0.04 (-0.03 to 0.35)	32.5 (-21.0 to 133.1)	0.26 (0.16 to 2.88)	69.2 (52.6 to 161.8)	0.45 (0.17 to 0.72)
	Fall	0.06 (-0.04 to 0.15)	48.2 (-24.9 to 124.4)	0.42 (0.14 to 1.13)	82.0 (55.5 to 156.1)	0.52 (0.08 to 0.72)
	All	0.02 (-0.01 to 0.15)	17.4 (-14.1 to 120.0)	0.24 (0.14 to 1.51)	75.8 (53.7 to 151.2)	0.54 (0.08 to 0.74)
NW	Winter	0.12 (0.08 to 0.20)	93.8 (46.7 to 231.5)	0.59 (0.41 to 0.84)	156.2 (96.1 to 252.8)	0.77 (0.60 to 0.84)
	Spring	0.20 (0.08 to 0.26)	260.0 (64.6 to 294.8)	0.80 (0.42 to 1.06)	291.8 (114.0 to 328.5)	0.72 (0.48 to 0.77)
	Summer	0.36 (0.05 to 0.66)	205.4 (40.1 to 388.8)	1.34 (0.29 to 2.57)	234.4 (89.5 to 410.7)	0.61 (0.31 to 0.77)
	Fall	0.35 (0.11 to 0.57)	171.4 (70.7 to 416.3)	1.14 (0.51 to 4.93)	194.7 (110.1 to 442.1)	0.60 (0.18 to 0.78)
	All	0.24 (0.09 to 0.43)	189.8 (68.4 to 309.2)	1.01 (0.50 to 2.87)	222.8 (117.0 to 327.2)	0.58 (0.37 to 0.75)
West	Winter	0.02 (-0.00 to 0.12)	16.9 (-1.9 to 54.1)	0.20 (0.16 to 0.54)	61.8 (50.6 to 86.8)	0.82 (0.73 to 0.91)
	Spring	0.03 (0.01 to 0.06)	32.4 (6.3 to 49.0)	0.13 (0.11 to 0.25)	68.6 (62.9 to 78.0)	0.71 (0.44 to 0.73)
	Summer	0.06 (0.01 to 0.42)	37.2 (3.7 to 137.3)	0.38 (0.13 to 1.08)	74.1 (50.1 to 161.3)	0.63 (0.39 to 0.82)
	Fall	0.12 (0.03 to 0.20)	62.8 (14.3 to 98.3)	0.57 (0.36 to 0.87)	97.7 (71.0 to 129.3)	0.51 (0.41 to 0.62)
	All	0.06 (0.02 to 0.18)	39.7 (10.7 to 90.4)	0.43 (0.22 to 0.66)	81.9 (62.3 to 120.9)	0.62 (0.50 to 0.71)
All	Winter	0.07 (0.03 to 0.13)	40.0 (13.1 to 78.0)	0.36 (0.29 to 0.93)	69.1 (58.2 to 102.0)	0.71 (0.37 to 0.76)
	Spring	0.06 (-0.03 to 0.09)	41.8 (-12.0 to 72.5)	0.38 (0.26 to 0.54)	75.4 (57.5 to 97.5)	0.56 (0.40 to 0.67)
	Summer	0.06 (-0.01 to 0.17)	34.3 (-4.2 to 88.9)	0.60 (0.18 to 1.42)	82.3 (51.3 to 116.6)	0.48 (0.35 to 0.66)
	Fall	0.11 (0.03 to 0.16)	57.2 (13.9 to 91.9)	0.57 (0.29 to 1.69)	87.5 (55.6 to 118.9)	0.59 (0.18 to 0.65)
	All	0.07 (0.02 to 0.13)	44.5 (7.9 to 84.0)	0.46 (0.31 to 1.23)	79.1 (59.2 to 110.2)	0.55 (0.33 to 0.66)

Table S10. Model performance statistics for PM_{2.5} at AQS (FRM/FEM) sites for the 2015 base case.

Region	Season	N	Avg. Obs. (µg m ⁻³)	Avg. Mod. (µg m ⁻³)	MB (µg m ⁻³)	NMB (%)	RMSE (µg m ⁻³)	NME (%)	r
Northeast	Winter	13001	10.04	12.74	2.71	27.0	7.33	48.0	0.68
	Spring	13538	7.97	8.83	0.86	10.8	5.19	44.0	0.59
	Summer	13660	8.38	8.02	-0.36	-4.3	4.06	35.2	0.67
	Fall	13270	7.18	9.08	1.90	26.5	5.40	50.0	0.73
	Annual	53469	8.38	9.64	1.26	15.0	5.60	44.2	0.67
Southeast	Winter	11190	8.07	10.28	2.21	27.4	5.65	47.4	0.58
	Spring	11961	8.06	8.25	0.18	2.3	4.08	33.6	0.55
	Summer	11641	9.78	8.45	-1.33	-13.6	4.86	35.3	0.47
	Fall	11365	6.93	8.13	1.20	17.3	4.32	41.7	0.70
	Annual	46157	8.22	8.76	0.54	6.6	4.75	39.1	0.55
Ohio Valley	Winter	10323	9.49	11.60	2.10	22.1	5.75	43.2	0.63
	Spring	10867	8.90	9.85	0.95	10.6	4.60	36.3	0.65
	Summer	10714	10.95	10.56	-0.39	-3.6	5.55	34.3	0.55
	Fall	10568	8.41	10.96	2.54	30.2	6.23	47.1	0.65
	Annual	42472	9.44	10.73	1.29	13.6	5.56	39.8	0.59
Upper Midwest	Winter	6478	8.79	9.72	0.92	10.5	4.75	38.2	0.70
	Spring	6643	7.32	8.27	0.96	13.1	4.30	41.9	0.67
	Summer	6718	7.88	7.85	-0.03	-0.4	5.26	40.8	0.56
	Fall	6664	6.81	9.14	2.33	34.2	4.92	49.3	0.75
	Annual	26503	7.69	8.74	1.04	13.6	4.82	42.2	0.64
South	Winter	8041	7.53	10.13	2.60	34.5	11.81	56.6	0.36
	Spring	8369	8.08	7.12	-0.96	-11.9	4.24	36.3	0.51
	Summer	8440	10.80	8.31	-2.49	-23.0	6.04	40.3	0.34
	Fall	8340	7.55	7.99	0.44	5.9	3.76	35.5	0.63
	Annual	33190	8.50	8.37	-0.13	-1.6	7.15	41.8	0.34
Southwest	Winter	4911	7.46	7.90	0.45	6.0	6.50	55.9	0.52
	Spring	4998	4.88	5.88	1.00	20.6	3.60	48.4	0.44
	Summer	5069	6.12	4.85	-1.27	-20.8	4.15	43.1	0.59
	Fall	5091	5.31	5.90	0.59	11.1	4.35	52.2	0.49
	Annual	20069	5.93	6.12	0.19	3.2	4.77	50.2	0.52
N. Rockies & Plains	Winter	4987	5.57	3.60	-1.98	-35.5	6.80	63.4	0.23
	Spring	5380	4.57	5.00	0.44	9.6	29.58	61.6	0.20
	Summer	5260	9.98	7.68	-2.30	-23.1	17.61	57.4	0.57
	Fall	5010	5.57	5.42	-0.15	-2.7	5.65	56.4	0.44
	Annual	20637	6.43	5.45	-0.99	-15.3	18.06	59.2	0.34
Northwest	Winter	8994	7.90	7.82	-0.08	-1.0	10.20	80.9	0.25
	Spring	9306	5.02	6.84	1.82	36.2	6.65	71.5	0.48
	Summer	9993	9.17	11.12	1.95	21.2	32.40	67.7	0.46
	Fall	9868	7.03	9.39	2.37	33.7	15.33	78.3	0.31
	Annual	38161	7.31	8.85	1.55	21.2	19.26	74.3	0.43
West	Winter	10462	11.67	9.58	-2.08	-17.8	8.09	43.3	0.68
	Spring	10989	7.52	6.95	-0.57	-7.6	4.17	38.3	0.55
	Summer	11065	8.95	8.53	-0.43	-4.8	6.36	43.5	0.51
	Fall	10587	8.61	9.11	0.50	5.8	16.85	46.9	0.37
	Annual	43103	9.16	8.52	-0.64	-7.0	10.02	43.1	0.44

Table S11. Definition of statistics used in the CMAQ model performance evaluation.

Statistic	Description
$MB (\mu\text{g m}^{-3}) = \frac{1}{n} \sum_{i=1}^n (P_i - O_i)$	Mean bias (MB) is defined as the average difference between predicted (P) and observed (O) concentrations for the total number of samples (n)
$RMSE (\mu\text{g m}^{-3}) = \sqrt{\sum_{i=1}^n (P_i - O_i)^2 / n}$	Root mean-squared error (RMSE)
$NMB (\%) = \frac{\sum_{i=1}^n (P_i - O_i)}{\sum_{i=1}^n O_i} \times 100$	The normalized mean bias (NMB) is defined as the sum of the difference between predictions and observations divided by the sum of observed values
$NME (\%) = \frac{\sum_{i=1}^n P_i - O_i }{\sum_{i=1}^n O_i} \times 100$	Normalized mean error (NME) is defined as the sum of the absolute value of the difference between predictions and observations divided by the sum of observed values
$r = \frac{\sum_{i=1}^n (P_i - \bar{P})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^n (P_i - \bar{P})^2} \sqrt{\sum_{i=1}^n (O_i - \bar{O})^2}}$	Pearson correlation coefficient

Table S12. Performance statistics for PM_{2.5} sulfate at CSN and IMPROVE sites for the 2015 base case.

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
Northeast	CSN	Winter	694	1.56	1.56	0.00	0.3	1.45	52.7	0.18
		Spring	764	1.54	1.48	-0.06	-4.0	0.76	30.2	0.64
		Summer	706	1.56	1.60	0.04	2.8	0.56	24.8	0.86
		Fall	703	1.11	1.23	0.12	10.8	0.63	32.9	0.84
		All	2867	1.45	1.47	0.02	1.7	0.91	35.1	0.61
	IMPROVE	Winter	384	1.07	0.85	-0.22	-20.2	0.52	34.6	0.69
		Spring	443	0.99	1.01	0.02	2.1	0.39	28.2	0.72
		Summer	464	0.97	1.05	0.08	8.2	0.42	30.2	0.87
		Fall	440	0.73	0.80	0.07	9.1	0.32	31.1	0.90
		All	1731	0.94	0.93	-0.00	-0.4	0.42	31.0	0.81
Southeast	CSN	Winter	446	1.23	1.31	0.08	6.1	0.74	44.6	0.47
		Spring	490	1.48	1.47	-0.01	-0.4	0.69	31.4	0.54
		Summer	478	1.47	1.47	0.00	0.0	0.52	26.8	0.63
		Fall	470	0.90	1.12	0.22	24.9	0.56	43.7	0.72
		All	1884	1.27	1.34	0.07	5.7	0.64	35.3	0.61
	IMPROVE	Winter	358	1.24	1.11	-0.13	-10.7	0.63	36.0	0.62
		Spring	413	1.54	1.40	-0.14	-9.2	0.62	28.7	0.64
		Summer	391	1.55	1.30	-0.24	-15.8	0.55	28.9	0.72
		Fall	378	0.89	0.99	0.10	11.2	0.45	35.6	0.77
		All	1540	1.31	1.21	-0.11	-8.1	0.57	31.5	0.70
Ohio Valley	CSN	Winter	535	1.77	1.28	-0.49	-27.6	1.24	42.0	0.43
		Spring	601	1.80	1.75	-0.05	-2.7	0.87	31.3	0.66
		Summer	542	2.35	2.33	-0.02	-0.8	1.38	29.5	0.68
		Fall	520	1.31	1.41	0.11	8.2	0.59	28.3	0.83
		All	2198	1.81	1.70	-0.11	-6.2	1.07	32.8	0.66
	IMPROVE	Winter	215	1.32	0.99	-0.33	-25.3	0.66	35.3	0.73
		Spring	245	1.48	1.34	-0.14	-9.3	0.68	32.6	0.59
		Summer	245	1.89	1.73	-0.17	-8.7	0.78	29.2	0.77
		Fall	233	1.08	1.16	0.08	7.2	0.45	31.0	0.87
		All	938	1.45	1.32	-0.14	-9.4	0.66	31.7	0.76
Upper Midwest	CSN	Winter	270	1.43	1.12	-0.31	-21.6	0.76	38.6	0.65
		Spring	291	1.36	1.41	0.05	3.5	0.55	29.0	0.84
		Summer	285	1.59	1.67	0.08	4.8	0.70	26.9	0.84
		Fall	275	1.08	1.23	0.15	14.3	0.62	34.3	0.81
		All	1121	1.37	1.36	-0.00	-0.4	0.66	31.8	0.79
	IMPROVE	Winter	216	0.96	0.70	-0.27	-27.6	0.50	36.9	0.67
		Spring	244	0.83	0.90	0.07	8.0	0.37	33.3	0.80
		Summer	242	0.96	1.07	0.11	11.7	0.41	31.5	0.87
		Fall	238	0.73	0.84	0.11	15.7	0.32	33.6	0.89
		All	940	0.87	0.88	0.01	1.6	0.40	33.8	0.80
South	CSN	Winter	258	1.28	1.13	-0.15	-11.8	0.73	37.7	0.64
		Spring	291	1.47	1.42	-0.05	-3.2	0.72	34.9	0.55
		Summer	288	1.90	1.49	-0.41	-21.6	0.80	32.9	0.61
		Fall	274	1.29	1.38	0.10	7.7	0.65	33.0	0.63
		All	1111	1.49	1.36	-0.13	-8.7	0.73	34.4	0.60
	IMPROVE	Winter	226	0.85	0.79	-0.06	-6.8	0.46	37.4	0.73
		Spring	271	1.16	1.06	-0.10	-8.9	0.60	37.4	0.60
		Summer	273	1.77	1.20	-0.56	-31.8	0.87	39.3	0.58
		Fall	245	1.12	1.02	-0.10	-8.9	0.60	36.2	0.65
		All	1015	1.24	1.03	-0.22	-17.3	0.66	37.9	0.65
Southwest	CSN	Winter	183	0.49	0.62	0.13	26.5	0.34	53.5	0.44
		Spring	202	0.61	0.96	0.35	57.1	0.47	65.3	0.44

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	r
		Summer	191	0.85	0.71	-0.14	-16.1	0.47	40.6	0.18
		Fall	184	0.58	0.68	0.10	17.2	0.43	52.7	0.41
		All	760	0.63	0.75	0.11	17.9	0.43	52.0	0.31
	IMPROVE	Winter	840	0.30	0.43	0.13	45.5	0.24	65.4	0.57
		Spring	959	0.50	0.87	0.37	73.9	0.48	83.4	0.51
		Summer	956	0.77	0.58	-0.19	-24.3	0.46	38.9	0.47
		Fall	916	0.48	0.53	0.05	10.6	0.31	49.4	0.63
All	3671	0.52	0.61	0.09	17.6	0.39	56.0	0.46		
N. Rockies & Plains	CSN	Winter	140	0.72	0.51	-0.20	-28.3	0.48	45.3	0.67
		Spring	149	0.77	0.86	0.09	11.2	0.35	35.5	0.68
		Summer	155	0.78	0.91	0.14	17.6	0.41	37.6	0.78
		Fall	141	0.63	0.69	0.06	9.2	0.35	38.1	0.78
		All	585	0.73	0.75	0.02	3.3	0.40	39.0	0.70
	IMPROVE	Winter	516	0.38	0.34	-0.03	-8.4	0.35	52.6	0.70
		Spring	589	0.52	0.75	0.23	43.8	0.42	61.4	0.60
		Summer	608	0.51	0.74	0.22	43.8	0.58	65.6	0.47
		Fall	571	0.33	0.53	0.20	62.4	0.35	79.9	0.47
		All	2284	0.44	0.60	0.16	37.2	0.44	64.4	0.51
Northwest	CSN	Winter	123	0.42	0.80	0.38	90.8	0.90	127.8	0.15
		Spring	135	0.54	1.07	0.53	99.2	0.70	103.8	0.63
		Summer	130	0.67	1.09	0.42	62.3	0.69	81.4	0.25
		Fall	132	0.44	0.91	0.47	108.3	0.73	113.3	0.32
		All	520	0.52	0.97	0.45	87.6	0.76	103.2	0.32
	IMPROVE	Winter	419	0.13	0.32	0.19	142.5	0.32	162.4	0.38
		Spring	472	0.40	0.82	0.42	104.5	0.57	106.9	0.78
		Summer	470	0.44	0.79	0.35	79.1	0.56	87.2	0.36
		Fall	458	0.25	0.62	0.38	152.4	0.84	161.7	0.29
		All	1819	0.31	0.65	0.34	108.5	0.61	116.1	0.53
West	CSN	Winter	226	0.68	0.81	0.13	19.4	0.64	64.7	0.48
		Spring	251	0.99	1.22	0.24	24.2	0.59	47.5	0.56
		Summer	252	1.29	1.26	-0.03	-2.1	0.61	32.6	0.63
		Fall	233	0.75	0.95	0.20	27.3	0.46	47.5	0.59
		All	962	0.94	1.07	0.14	14.5	0.58	45.1	0.60
	IMPROVE	Winter	492	0.30	0.49	0.20	66.7	0.47	107.1	0.36
		Spring	600	0.63	0.95	0.32	50.4	0.54	68.4	0.47
		Summer	579	0.76	0.86	0.10	12.7	0.48	46.9	0.23
		Fall	539	0.45	0.65	0.19	43.0	0.42	72.2	0.37
		All	2210	0.55	0.75	0.20	37.1	0.48	66.0	0.46
All	CSN	Winter	2875	1.28	1.19	-0.10	-7.5	1.03	47.3	0.42
		Spring	3174	1.38	1.42	0.04	3.1	0.71	34.4	0.68
		Summer	3027	1.58	1.57	-0.01	-0.7	0.80	29.9	0.76
		Fall	2932	1.01	1.16	0.15	15.3	0.58	36.8	0.80
		All	12008	1.32	1.34	0.02	1.8	0.79	36.5	0.67
	IMPROVE	Winter	3666	0.60	0.60	0.00	0.0	0.44	48.7	0.74
		Spring	4236	0.78	0.97	0.18	23.6	0.52	50.1	0.66
		Summer	4228	0.93	0.92	-0.01	-1.2	0.55	40.4	0.70
		Fall	4018	0.58	0.71	0.13	22.6	0.46	50.5	0.70
		All	16148	0.73	0.81	0.08	10.8	0.50	46.7	0.70

Table S13. Performance statistics for PM_{2.5} nitrate at CSN and IMPROVE sites for the 2015 base case.

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
Northeast	CSN	Winter	695	2.17	2.53	0.36	16.4	1.42	41.6	0.70
		Spring	764	1.53	1.51	-0.02	-1.3	1.12	47.8	0.77
		Summer	706	0.43	0.28	-0.15	-35.1	0.40	59.3	0.61
		Fall	703	0.67	0.93	0.27	40.2	1.00	74.3	0.68
		All	2868	1.20	1.31	0.11	9.1	1.05	49.7	0.78
	IMPROVE	Winter	384	0.63	1.04	0.42	66.3	0.96	83.8	0.80
		Spring	443	0.43	0.51	0.08	18.1	0.55	70.7	0.64
		Summer	464	0.16	0.16	0.00	0.0	0.23	80.0	0.34
		Fall	440	0.18	0.29	0.11	60.1	0.33	102.8	0.53
		All	1731	0.34	0.48	0.14	41.5	0.57	81.6	0.75
Southeast	CSN	Winter	446	0.93	1.65	0.71	76.7	1.32	93.7	0.63
		Spring	493	0.54	0.58	0.04	7.6	0.51	61.3	0.72
		Summer	478	0.28	0.22	-0.06	-22.5	0.26	62.7	0.32
		Fall	470	0.31	0.61	0.30	98.4	0.77	130.8	0.54
		All	1887	0.51	0.75	0.24	47.0	0.80	86.0	0.67
	IMPROVE	Winter	358	0.62	0.90	0.28	45.0	0.83	80.1	0.67
		Spring	413	0.41	0.43	0.02	4.6	0.45	67.1	0.68
		Summer	391	0.20	0.20	-0.00	-1.0	0.22	63.4	0.42
		Fall	378	0.21	0.32	0.11	52.4	0.37	100.0	0.47
		All	1540	0.36	0.45	0.10	26.9	0.51	76.5	0.68
Ohio Valley	CSN	Winter	496	2.74	2.41	-0.33	-12.1	1.84	40.5	0.63
		Spring	554	1.67	1.35	-0.32	-19.4	1.26	46.9	0.76
		Summer	542	0.64	0.53	-0.11	-17.4	0.75	67.2	0.44
		Fall	520	0.66	0.82	0.15	23.1	0.67	65.6	0.63
		All	2112	1.41	1.26	-0.15	-10.9	1.21	48.5	0.74
	IMPROVE	Winter	215	1.33	1.29	-0.05	-3.5	1.02	54.2	0.62
		Spring	245	0.64	0.66	0.01	2.1	0.85	71.3	0.47
		Summer	245	0.23	0.20	-0.03	-13.3	0.19	53.8	0.64
		Fall	233	0.31	0.35	0.04	13.2	0.36	70.7	0.58
		All	938	0.61	0.60	-0.01	-0.8	0.69	61.0	0.67
Upper Midwest	CSN	Winter	270	2.73	2.33	-0.39	-14.4	1.63	39.0	0.72
		Spring	291	2.26	1.64	-0.62	-27.6	1.83	48.0	0.72
		Summer	285	0.62	0.60	-0.02	-2.7	0.72	63.9	0.61
		Fall	275	0.87	0.96	0.09	10.8	0.81	56.8	0.63
		All	1121	1.61	1.38	-0.24	-14.7	1.34	47.1	0.74
	IMPROVE	Winter	216	1.45	1.31	-0.14	-9.9	1.17	44.3	0.79
		Spring	244	0.91	0.68	-0.23	-25.8	0.84	50.4	0.77
		Summer	242	0.21	0.25	0.04	20.8	0.22	62.5	0.83
		Fall	238	0.46	0.56	0.10	22.6	0.56	66.4	0.64
		All	940	0.74	0.68	-0.06	-7.7	0.77	51.0	0.79
South	CSN	Winter	258	1.36	1.41	0.05	4.0	1.05	55.2	0.61
		Spring	291	0.69	0.65	-0.05	-6.6	0.66	54.8	0.70
		Summer	288	0.39	0.22	-0.17	-43.9	0.29	59.5	0.46
		Fall	274	0.39	0.38	-0.01	-1.8	0.44	71.9	0.51
		All	1111	0.69	0.65	-0.04	-6.5	0.66	58.0	0.70
	IMPROVE	Winter	226	0.88	0.85	-0.03	-2.9	1.07	69.1	0.50
		Spring	271	0.48	0.40	-0.08	-16.3	0.54	61.5	0.66
		Summer	273	0.24	0.19	-0.04	-18.4	0.19	54.8	0.46
		Fall	245	0.21	0.26	0.05	25.6	0.35	86.9	0.35
		All	1015	0.44	0.41	-0.03	-5.8	0.61	66.8	0.59
Southwest	CSN	Winter	183	2.00	1.06	-0.94	-47.1	2.34	62.3	0.50
		Spring	202	0.78	0.45	-0.33	-42.0	0.96	62.6	0.68

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	r
		Summer	191	0.32	0.22	-0.10	-31.2	0.49	104.1	-0.04
		Fall	184	0.63	0.52	-0.12	-18.3	0.97	79.0	0.55
		All	760	0.92	0.56	-0.37	-39.8	1.36	68.8	0.59
	IMPROVE	Winter	840	0.23	0.20	-0.03	-13.6	0.34	74.4	0.64
		Spring	959	0.19	0.13	-0.06	-31.0	0.22	69.5	0.24
		Summer	956	0.14	0.05	-0.08	-62.0	0.13	67.2	0.47
		Fall	916	0.12	0.12	-0.01	-4.4	0.19	73.5	0.41
All	3671	0.17	0.12	-0.05	-27.3	0.23	71.3	0.51		
N. Rockies & Plains	CSN	Winter	141	1.38	0.93	-0.45	-32.8	1.17	54.1	0.75
		Spring	149	0.92	0.69	-0.23	-25.5	0.77	47.7	0.72
		Summer	155	0.32	0.36	0.04	14.2	0.87	93.1	0.42
		Fall	141	0.53	0.62	0.09	16.9	0.67	70.0	0.66
		All	586	0.78	0.64	-0.14	-17.4	0.89	59.0	0.66
	IMPROVE	Winter	516	0.32	0.24	-0.09	-26.6	0.52	74.6	0.47
		Spring	589	0.26	0.24	-0.03	-10.8	0.25	55.9	0.76
		Summer	608	0.16	0.25	0.09	59.5	0.85	152.8	0.46
		Fall	571	0.17	0.32	0.16	95.7	0.48	137.5	0.61
		All	2284	0.22	0.26	0.04	17.0	0.57	95.2	0.37
Northwest	CSN	Winter	123	1.45	1.39	-0.06	-4.2	2.15	77.4	0.33
		Spring	135	0.55	1.14	0.59	106.7	1.45	146.2	0.39
		Summer	130	0.45	1.10	0.65	144.7	1.66	185.4	0.31
		Fall	132	0.77	1.25	0.48	62.4	1.35	106.7	0.36
		All	520	0.79	1.22	0.42	53.2	1.67	112.3	0.28
	IMPROVE	Winter	419	0.26	0.28	0.03	10.3	0.75	109.6	0.39
		Spring	472	0.13	0.29	0.16	117.5	0.67	155.7	0.71
		Summer	470	0.21	0.38	0.17	84.7	0.80	137.2	0.44
		Fall	458	0.19	0.40	0.22	117.0	0.69	160.6	0.52
		All	1819	0.19	0.34	0.15	75.8	0.73	137.8	0.39
West	CSN	Winter	226	4.32	2.98	-1.34	-31.0	3.65	48.4	0.78
		Spring	251	1.53	0.92	-0.61	-40.1	1.41	47.7	0.74
		Summer	252	1.45	1.06	-0.39	-26.6	1.01	42.8	0.82
		Fall	233	1.87	1.31	-0.56	-29.9	2.48	57.8	0.50
		All	962	2.25	1.54	-0.71	-31.7	2.33	49.2	0.77
	IMPROVE	Winter	492	0.83	0.77	-0.06	-7.2	1.53	57.0	0.87
		Spring	600	0.45	0.40	-0.05	-11.6	0.52	56.9	0.63
		Summer	579	0.37	0.46	0.09	24.2	0.50	76.9	0.55
		Fall	539	0.49	0.52	0.02	4.9	0.92	76.1	0.67
		All	2210	0.53	0.53	0.00	0.3	0.93	65.0	0.81
All	CSN	Winter	2838	2.14	2.06	-0.08	-3.9	1.84	49.2	0.68
		Spring	3130	1.27	1.09	-0.17	-13.7	1.15	51.3	0.73
		Summer	3027	0.53	0.44	-0.09	-17.2	0.68	65.4	0.59
		Fall	2932	0.69	0.81	0.12	17.6	1.07	72.8	0.52
		All	11927	1.15	1.09	-0.06	-5.1	1.25	55.2	0.71
	IMPROVE	Winter	3666	0.58	0.62	0.03	5.8	0.90	66.9	0.77
		Spring	4236	0.36	0.35	-0.02	-4.1	0.51	66.1	0.63
		Summer	4228	0.20	0.23	0.02	11.7	0.48	85.7	0.47
		Fall	4018	0.23	0.32	0.08	35.4	0.52	92.9	0.56
		All	16148	0.34	0.37	0.03	9.0	0.62	74.1	0.69

Table S14. Performance statistics for PM_{2.5} OC at CSN and IMPROVE sites for the 2015 base case.

Region	Network	Season	N	Avg. Obs. (µg m ⁻³)	Avg. Mod. (µg m ⁻³)	MB (µg m ⁻³)	NMB (%)	RMSE (µg m ⁻³)	NME (%)	r
Northeast	CSN	Winter	690	2.55	3.74	1.19	46.4	2.27	62.9	0.63
		Spring	729	2.34	2.56	0.22	9.4	1.69	48.4	0.57
		Summer	707	3.30	2.19	-1.11	-33.7	1.96	42.0	0.49
		Fall	696	2.58	2.63	0.06	2.2	1.61	38.7	0.73
		All	2822	2.69	2.77	0.08	3.0	1.90	47.5	0.51
	IMPROVE	Winter	403	0.98	1.72	0.75	76.2	1.33	89.4	0.78
		Spring	462	0.87	1.06	0.18	21.2	0.81	59.3	0.65
		Summer	467	1.45	1.07	-0.38	-26.4	0.89	40.7	0.65
		Fall	472	1.00	1.13	0.12	12.2	0.89	44.7	0.70
		All	1804	1.08	1.23	0.15	13.6	0.99	55.4	0.61
Southeast	CSN	Winter	360	2.66	3.10	0.44	16.6	1.87	43.7	0.59
		Spring	425	2.62	2.28	-0.34	-12.9	1.68	44.9	0.42
		Summer	408	3.07	2.84	-0.23	-7.6	1.57	37.3	0.50
		Fall	402	2.78	2.77	-0.01	-0.4	1.77	44.9	0.68
		All	1595	2.79	2.73	-0.05	-1.9	1.72	42.5	0.56
	IMPROVE	Winter	385	1.26	1.81	0.55	43.6	1.70	73.3	0.60
		Spring	440	1.55	1.56	0.01	0.6	5.73	72.6	0.07
		Summer	423	1.55	1.63	0.08	5.0	1.40	53.0	0.59
		Fall	410	1.29	1.51	0.23	17.5	1.54	58.0	0.73
		All	1658	1.42	1.62	0.21	14.5	3.23	64.0	0.19
Ohio Valley	CSN	Winter	484	2.21	2.59	0.38	17.4	1.21	38.0	0.54
		Spring	558	2.49	2.13	-0.36	-14.4	1.31	39.0	0.49
		Summer	529	3.35	2.28	-1.07	-32.0	1.92	40.2	0.47
		Fall	524	2.92	2.46	-0.46	-15.6	1.56	39.1	0.63
		All	2095	2.75	2.36	-0.39	-14.2	1.53	39.2	0.47
	IMPROVE	Winter	213	0.97	1.47	0.49	50.6	1.40	68.7	0.43
		Spring	239	1.19	1.46	0.27	22.7	6.33	77.4	0.12
		Summer	245	1.51	1.45	-0.06	-4.0	1.05	40.6	0.51
		Fall	233	1.31	1.47	0.16	12.1	1.24	52.2	0.65
		All	930	1.26	1.46	0.21	16.4	3.38	57.6	0.21
Upper Midwest	CSN	Winter	255	1.85	2.63	0.78	42.2	1.69	59.1	0.46
		Spring	289	2.03	2.09	0.06	3.0	1.16	43.0	0.55
		Summer	287	2.99	2.00	-0.99	-33.2	1.61	41.9	0.55
		Fall	278	2.56	2.34	-0.22	-8.6	1.63	41.6	0.61
		All	1109	2.37	2.25	-0.12	-5.0	1.53	45.1	0.43
	IMPROVE	Winter	243	0.67	1.12	0.44	66.2	0.75	73.6	0.85
		Spring	274	0.90	0.97	0.07	7.4	0.83	55.8	0.62
		Summer	271	1.71	0.99	-0.72	-42.0	1.46	51.3	0.48
		Fall	268	1.08	1.20	0.12	11.1	0.85	46.7	0.72
		All	1056	1.10	1.07	-0.03	-3.1	1.02	54.2	0.50
South	CSN	Winter	254	2.59	2.67	0.09	3.3	1.67	41.1	0.62
		Spring	289	2.12	1.55	-0.58	-27.1	1.10	42.1	0.58
		Summer	278	2.62	2.31	-0.31	-11.9	1.47	40.1	0.65
		Fall	274	2.89	2.23	-0.66	-22.7	1.37	37.4	0.63
		All	1095	2.55	2.17	-0.37	-14.7	1.41	40.0	0.63
	IMPROVE	Winter	222	0.76	2.21	1.46	192.7	16.61	227.6	0.29
		Spring	271	0.86	0.56	-0.30	-35.0	0.71	49.7	0.68
		Summer	273	1.32	1.15	-0.17	-12.8	1.01	49.2	0.61
		Fall	245	1.13	1.06	-0.07	-5.9	2.27	54.5	0.32
		All	1011	1.03	1.20	0.18	17.3	7.89	79.6	0.18
Southwest	CSN	Winter	179	2.65	3.33	0.68	25.8	2.08	56.0	0.49
		Spring	193	1.58	1.59	0.01	0.8	0.98	48.0	0.46

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	r
		Summer	189	2.37	1.61	-0.76	-32.0	1.54	42.7	0.72
		Fall	181	2.24	1.93	-0.31	-13.7	1.19	39.1	0.54
		All	742	2.20	2.10	-0.10	-4.5	1.50	46.7	0.58
	IMPROVE	Winter	831	0.48	0.43	-0.06	-11.5	0.52	53.6	0.80
		Spring	959	0.44	0.31	-0.13	-29.6	0.34	43.3	0.55
		Summer	955	0.91	0.68	-0.23	-25.5	0.77	48.7	0.76
		Fall	913	0.57	0.53	-0.04	-7.4	1.58	56.5	0.34
All	3658	0.60	0.49	-0.12	-19.4	0.94	50.4	0.55		
N. Rockies & Plains	CSN	Winter	121	1.43	1.09	-0.34	-24.1	2.37	70.2	0.05
		Spring	148	1.48	0.74	-0.74	-49.9	1.23	63.1	0.39
		Summer	151	3.58	1.81	-1.77	-49.5	3.20	58.1	0.89
		Fall	142	2.05	0.92	-1.13	-55.3	1.64	60.3	0.35
		All	562	2.18	1.15	-1.03	-47.4	2.24	61.2	0.80
	IMPROVE	Winter	536	0.29	0.23	-0.06	-20.2	0.57	75.2	0.13
		Spring	608	0.46	0.39	-0.07	-15.6	2.21	88.5	0.07
		Summer	602	3.11	2.82	-0.29	-9.3	13.66	79.6	0.40
		Fall	570	0.91	0.83	-0.09	-9.7	2.35	74.6	0.48
		All	2316	1.22	1.09	-0.13	-10.6	7.16	79.4	0.42
Northwest	CSN	Winter	122	3.16	5.56	2.40	75.8	4.96	97.2	0.57
		Spring	135	2.32	4.20	1.88	80.8	3.99	105.2	0.41
		Summer	134	3.36	4.33	0.97	28.9	3.69	72.8	0.59
		Fall	129	3.80	4.65	0.85	22.3	5.46	66.7	0.26
		All	520	3.15	4.66	1.51	47.9	4.56	82.9	0.40
	IMPROVE	Winter	419	0.43	0.83	0.40	93.4	2.23	162.7	0.69
		Spring	476	0.59	0.83	0.23	39.0	2.00	119.5	0.57
		Summer	466	2.62	3.20	0.58	22.3	10.21	86.0	0.59
		Fall	455	1.06	2.69	1.62	152.4	23.81	224.7	0.14
		All	1816	1.19	1.90	0.71	59.4	13.08	127.7	0.30
West	CSN	Winter	217	4.10	3.79	-0.31	-7.6	1.90	32.1	0.69
		Spring	249	2.14	1.76	-0.37	-17.5	0.82	31.1	0.78
		Summer	246	2.69	2.07	-0.63	-23.3	1.52	37.6	0.62
		Fall	229	3.62	3.17	-0.46	-12.6	2.05	33.5	0.64
		All	941	3.10	2.65	-0.45	-14.4	1.62	33.6	0.71
	IMPROVE	Winter	497	0.67	0.58	-0.09	-13.0	0.71	58.2	0.83
		Spring	601	0.70	0.41	-0.29	-41.9	0.55	53.9	0.63
		Summer	579	1.77	1.97	0.21	11.7	4.15	72.4	0.52
		Fall	538	1.25	1.32	0.08	6.1	3.21	73.6	0.51
		All	2215	1.11	1.08	-0.03	-2.4	2.68	67.6	0.55
All	CSN	Winter	2682	2.55	3.18	0.63	24.7	2.13	51.6	0.56
		Spring	3015	2.25	2.16	-0.09	-4.1	1.60	46.6	0.50
		Summer	2929	3.09	2.32	-0.77	-24.9	1.97	43.1	0.64
		Fall	2855	2.79	2.56	-0.23	-8.3	1.97	41.8	0.56
		All	11481	2.67	2.54	-0.13	-4.9	1.92	45.4	0.55
	IMPROVE	Winter	3749	0.66	0.96	0.29	44.0	4.21	87.3	0.28
		Spring	4330	0.75	0.72	-0.03	-4.4	2.62	67.7	0.13
		Summer	4281	1.75	1.66	-0.09	-5.4	6.37	65.2	0.48
		Fall	4104	0.99	1.21	0.22	22.0	8.14	79.3	0.17
		All	16464	1.05	1.14	0.09	8.3	5.74	72.1	0.31

Table S15. Performance statistics for PM_{2.5} EC at CSN and IMPROVE sites for the 2015 base case.

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	<i>r</i>
Northeast	CSN	Winter	690	0.75	0.99	0.25	33.0	0.67	62.8	0.55
		Spring	729	0.63	0.78	0.15	24.3	0.64	59.1	0.52
		Summer	707	0.62	0.76	0.14	21.7	0.47	49.8	0.52
		Fall	696	0.66	0.88	0.22	33.7	0.73	60.5	0.58
		All	2822	0.66	0.85	0.19	28.4	0.63	58.3	0.55
	IMPROVE	Winter	403	0.22	0.39	0.17	77.4	0.29	88.8	0.81
		Spring	462	0.16	0.27	0.11	68.4	0.21	82.9	0.76
		Summer	467	0.21	0.30	0.09	44.8	0.24	66.4	0.65
		Fall	472	0.19	0.30	0.10	52.7	0.20	67.5	0.82
		All	1804	0.19	0.31	0.12	60.1	0.24	75.8	0.77
Southeast	CSN	Winter	360	0.58	0.84	0.26	45.8	0.56	67.7	0.54
		Spring	425	0.47	0.63	0.16	33.5	0.49	65.9	0.44
		Summer	408	0.47	0.59	0.13	26.9	0.45	62.5	0.32
		Fall	402	0.55	0.78	0.23	42.2	0.53	61.2	0.65
		All	1595	0.51	0.71	0.19	37.4	0.51	64.3	0.52
	IMPROVE	Winter	385	0.29	0.46	0.17	59.3	0.39	76.4	0.74
		Spring	440	0.26	0.39	0.12	47.7	0.57	77.0	0.48
		Summer	423	0.25	0.26	0.01	4.6	0.19	41.2	0.79
		Fall	410	0.26	0.36	0.10	41.0	0.31	59.4	0.77
		All	1658	0.26	0.36	0.10	38.5	0.40	63.9	0.65
Ohio Valley	CSN	Winter	484	0.50	0.74	0.24	49.0	0.45	68.1	0.48
		Spring	558	0.59	0.68	0.08	13.9	0.47	55.7	0.38
		Summer	529	0.72	0.79	0.07	9.6	0.40	40.7	0.54
		Fall	524	0.65	0.79	0.14	22.2	0.41	45.7	0.60
		All	2095	0.62	0.75	0.13	21.3	0.43	51.0	0.50
	IMPROVE	Winter	213	0.21	0.35	0.13	61.6	0.31	76.4	0.43
		Spring	239	0.22	0.33	0.12	53.6	1.28	83.4	0.12
		Summer	245	0.23	0.25	0.02	7.3	0.14	37.2	0.75
		Fall	233	0.24	0.33	0.08	34.3	0.25	59.9	0.48
		All	930	0.23	0.31	0.09	37.6	0.68	63.1	0.20
Upper Midwest	CSN	Winter	255	0.35	0.67	0.32	91.0	0.48	99.1	0.53
		Spring	289	0.38	0.57	0.19	48.9	0.36	67.1	0.56
		Summer	287	0.52	0.71	0.19	35.6	0.39	53.5	0.67
		Fall	278	0.49	0.70	0.21	43.2	0.46	56.3	0.64
		All	1109	0.44	0.66	0.22	50.9	0.42	65.8	0.61
	IMPROVE	Winter	243	0.16	0.29	0.13	80.4	0.19	84.7	0.85
		Spring	274	0.18	0.26	0.08	41.4	0.19	62.8	0.73
		Summer	271	0.27	0.34	0.07	26.7	0.25	52.1	0.71
		Fall	268	0.23	0.35	0.12	53.9	0.25	70.6	0.71
		All	1056	0.21	0.31	0.10	46.8	0.22	65.2	0.73
South	CSN	Winter	254	0.57	0.81	0.24	41.9	0.47	57.9	0.67
		Spring	289	0.40	0.55	0.15	37.1	0.32	56.7	0.57
		Summer	278	0.40	0.51	0.12	29.4	0.27	52.7	0.56
		Fall	274	0.51	0.67	0.16	31.2	0.33	49.3	0.64
		All	1095	0.47	0.63	0.16	35.2	0.35	54.1	0.65
	IMPROVE	Winter	222	0.15	0.51	0.36	241.2	3.54	257.2	0.41
		Spring	271	0.15	0.16	0.01	7.1	0.13	48.2	0.69
		Summer	273	0.15	0.17	0.03	18.5	0.14	45.7	0.75
		Fall	245	0.17	0.22	0.06	34.1	0.48	65.4	0.21
		All	1011	0.15	0.26	0.10	68.0	1.68	97.4	0.27
Southwest	CSN	Winter	179	0.79	1.14	0.35	45.1	0.69	68.5	0.51
		Spring	193	0.37	0.64	0.27	73.2	0.48	93.9	0.43

Region	Network	Season	N	Avg. Obs. ($\mu\text{g m}^{-3}$)	Avg. Mod. ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	NMB (%)	RMSE ($\mu\text{g m}^{-3}$)	NME (%)	r	
		Summer	189	0.36	0.59	0.23	65.0	0.43	81.6	0.52	
		Fall	181	0.51	0.76	0.25	49.2	0.46	72.8	0.55	
		All	742	0.50	0.78	0.28	55.1	0.52	76.8	0.58	
	IMPROVE	Winter	831	0.12	0.14	0.02	19.7	0.17	60.4	0.83	
		Spring	959	0.07	0.11	0.04	64.9	0.10	83.3	0.68	
		Summer	955	0.11	0.13	0.02	15.8	0.14	58.4	0.78	
		Fall	913	0.10	0.13	0.03	29.2	0.37	77.4	0.36	
		All	3658	0.10	0.13	0.03	29.1	0.22	68.4	0.59	
N. Rockies & Plains	CSN	Winter	121	0.22	0.35	0.13	59.4	0.56	124.4	0.09	
		Spring	148	0.19	0.28	0.09	47.0	0.29	89.7	0.51	
		Summer	151	0.35	0.56	0.21	61.2	0.76	80.6	0.92	
		Fall	142	0.28	0.36	0.08	28.2	0.38	80.3	0.24	
		All	562	0.26	0.39	0.13	49.2	0.53	90.2	0.65	
	IMPROVE	Winter	536	0.05	0.09	0.04	70.1	0.13	117.9	0.23	
		Spring	608	0.06	0.12	0.06	96.1	0.46	124.2	0.09	
		Summer	602	0.26	0.61	0.35	133.1	2.88	161.8	0.40	
		Fall	570	0.11	0.24	0.13	124.4	0.55	156.1	0.37	
		All	2316	0.12	0.27	0.15	120.0	1.51	151.2	0.40	
	Northwest	CSN	Winter	122	0.78	1.75	0.97	123.9	1.88	137.4	0.54
			Spring	135	0.53	1.66	1.13	211.8	2.14	218.1	0.35
Summer			134	0.56	2.12	1.56	280.1	2.61	290.2	0.38	
Fall			129	0.80	1.86	1.07	133.6	2.00	148.5	0.34	
All			520	0.66	1.85	1.19	179.0	2.19	190.7	0.39	
IMPROVE		Winter	419	0.09	0.29	0.20	231.5	0.84	252.8	0.78	
		Spring	476	0.09	0.35	0.26	277.5	1.06	294.1	0.72	
		Summer	466	0.23	0.89	0.66	286.3	2.57	299.1	0.58	
		Fall	455	0.14	0.71	0.57	416.3	4.93	442.1	0.18	
		All	1816	0.14	0.56	0.43	309.2	2.87	327.2	0.37	
West		CSN	Winter	217	1.08	1.11	0.03	2.7	0.58	39.2	0.61
			Spring	249	0.42	0.58	0.16	37.0	0.47	58.2	0.42
	Summer		246	0.40	0.65	0.25	61.1	0.37	68.4	0.65	
	Fall		229	0.82	0.96	0.13	16.4	0.57	45.2	0.51	
	All		941	0.67	0.81	0.15	21.8	0.50	48.8	0.63	
	IMPROVE	Winter	497	0.12	0.17	0.05	40.5	0.23	82.1	0.82	
		Spring	601	0.09	0.13	0.04	49.0	0.12	78.0	0.72	
		Summer	579	0.17	0.41	0.24	137.3	0.90	161.3	0.39	
		Fall	538	0.15	0.30	0.15	98.3	0.67	129.3	0.54	
		All	2215	0.13	0.26	0.12	90.4	0.58	120.9	0.50	
	All	CSN	Winter	2682	0.63	0.90	0.27	43.0	0.69	68.0	0.54
			Spring	3015	0.50	0.69	0.19	38.8	0.66	69.5	0.41
Summer			2929	0.53	0.74	0.21	39.3	0.71	64.8	0.44	
Fall			2855	0.60	0.83	0.23	37.8	0.68	61.1	0.54	
All			11481	0.56	0.79	0.22	39.7	0.68	65.7	0.49	
IMPROVE		Winter	3749	0.14	0.26	0.11	78.0	0.93	102.0	0.37	
		Spring	4330	0.12	0.21	0.09	72.5	0.54	97.5	0.40	
		Summer	4281	0.20	0.37	0.17	88.9	1.42	116.6	0.44	
		Fall	4104	0.16	0.31	0.15	91.9	1.69	118.9	0.18	
		All	16464	0.16	0.29	0.13	84.0	1.23	110.2	0.33	

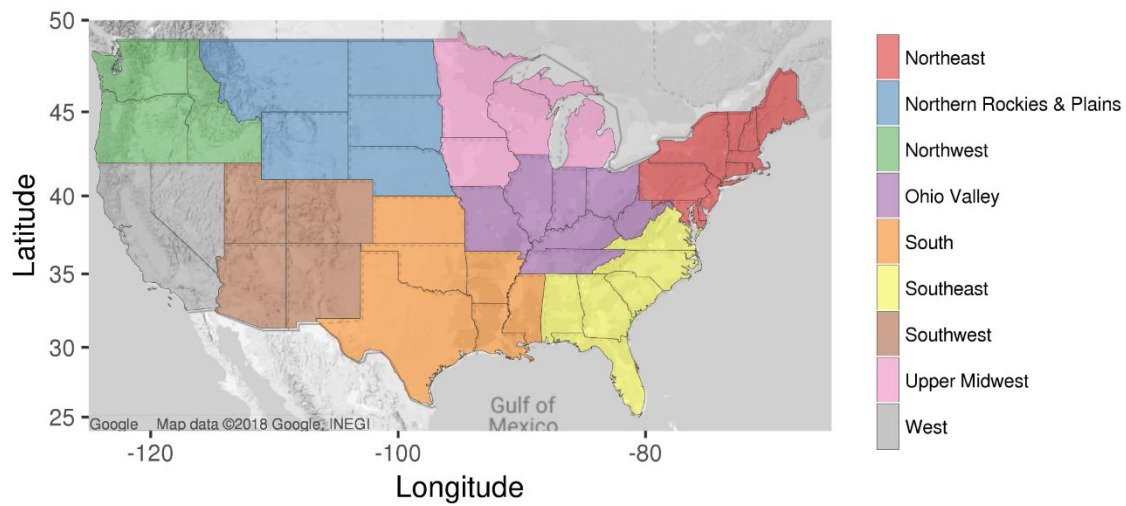


Figure S1. U.S. climate regions (<https://www.ncdc.noaa.gov/monitoring-references/maps/us-climate-regions.php>) used in the CMAQ model performance evaluation.

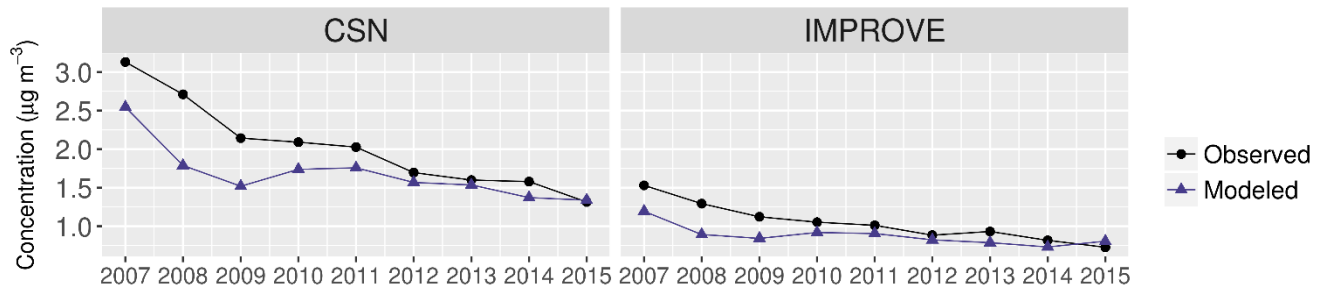


Figure S2. Annual average concentration of sulfate during 2007-2015.

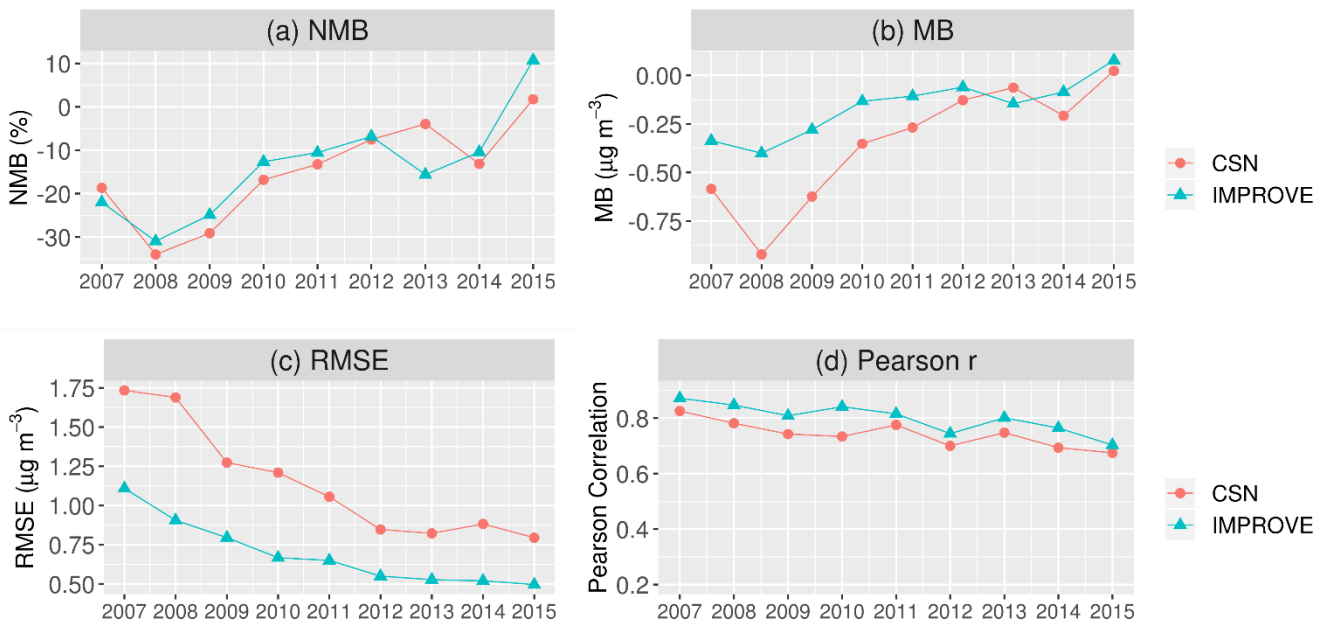


Figure S3. Annual sulfate model performance statistics by year.

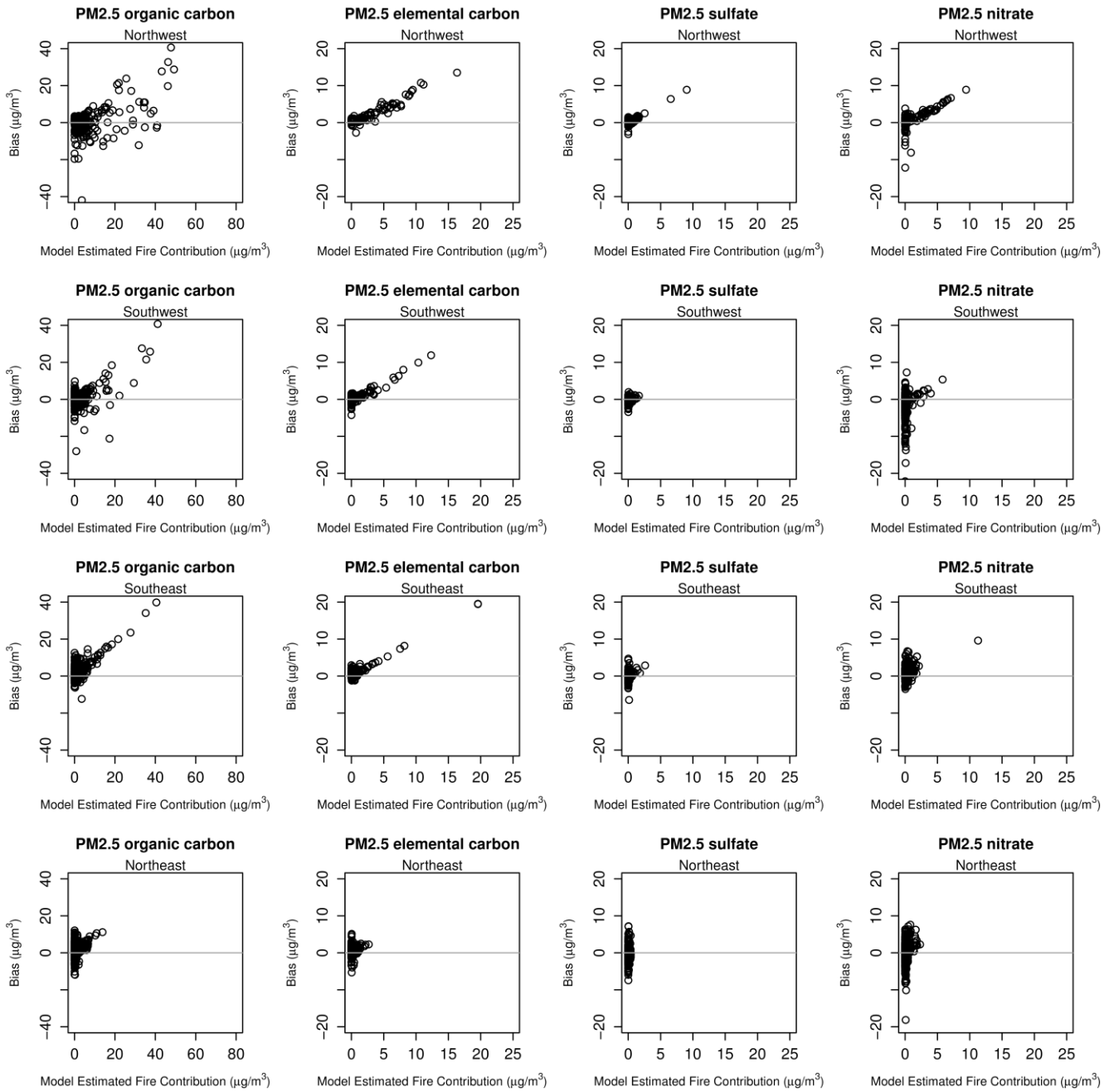


Figure S4. Bias in model predictions of PM_{2.5} OC, EC, SO₄²⁻, and NO₃⁻ in the Northwest, Southwest, Southeast and Northeast regions as a function of the modeled fire contribution to the concentration.

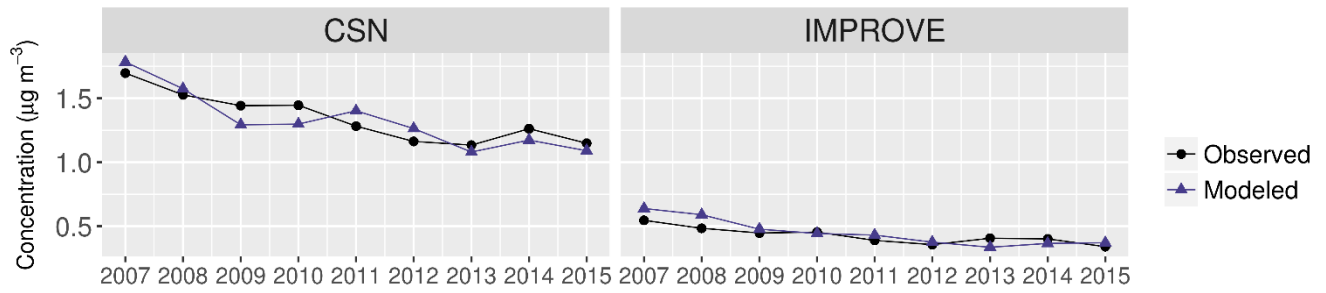


Figure S5. Annual average concentration of nitrate during 2007-2015.

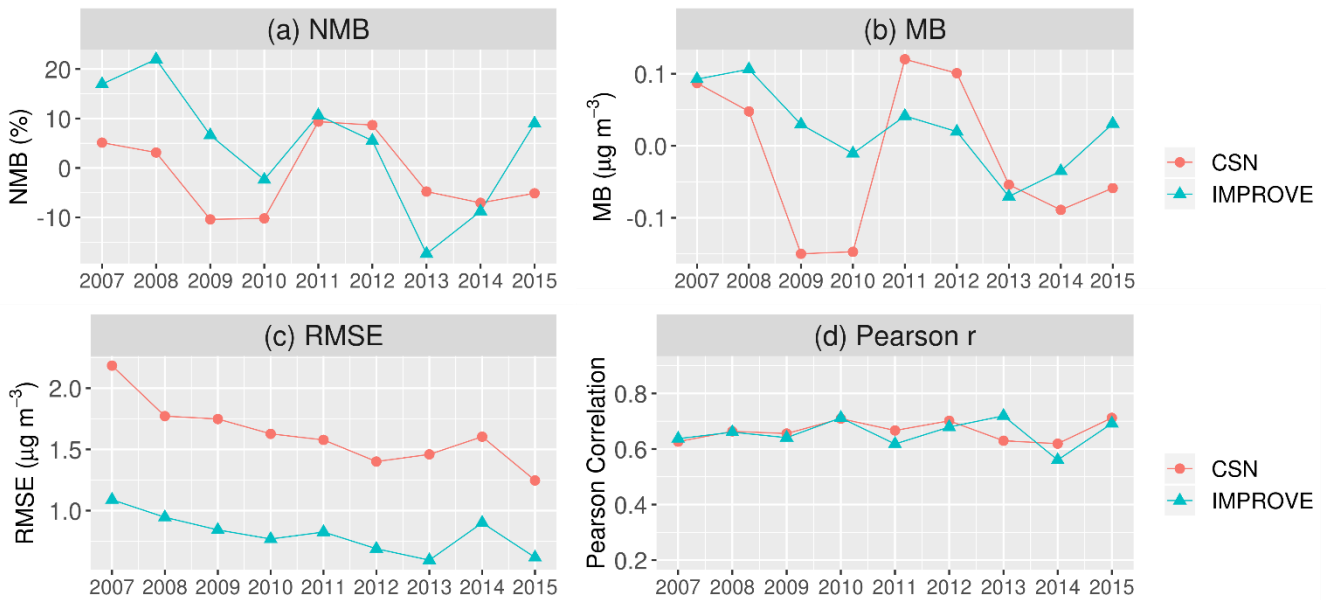


Figure S6. Annual nitrate model performance statistics by year for CSN and IMPROVE sites.

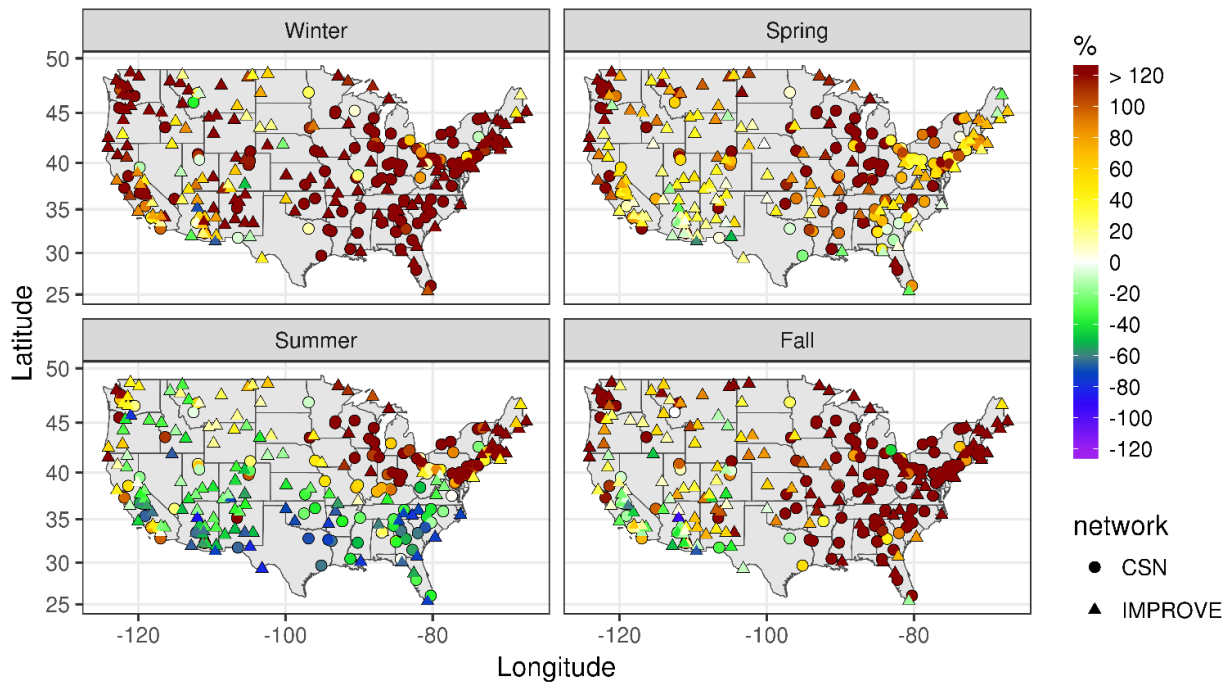


Figure S7. Normalized mean bias (NMB; %) by season for PM_{2.5} soil predictions.

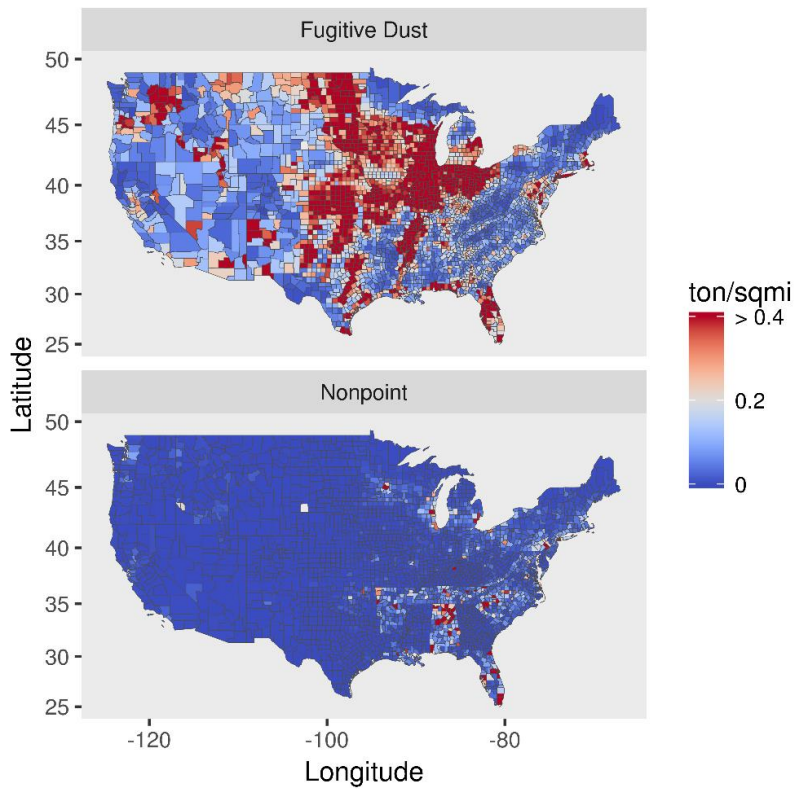


Figure S8. Annual PM_{2.5} soil emissions from the afdust_adj and nonpt sectors.

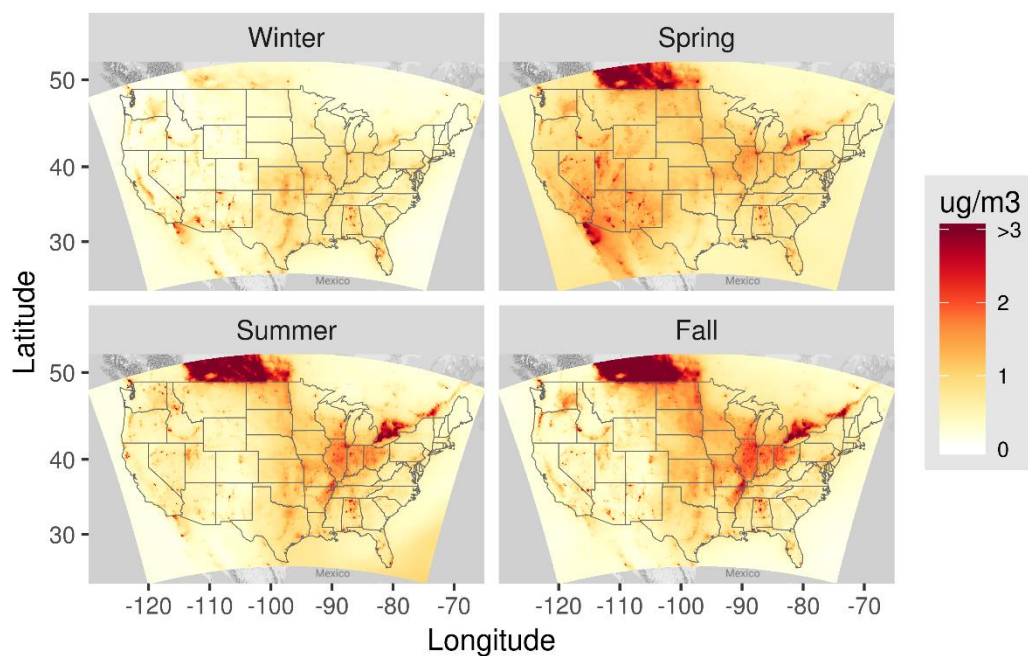


Figure S9. Seasonal average predicted PM_{2.5} soil concentrations ($\mu\text{g m}^{-3}$).

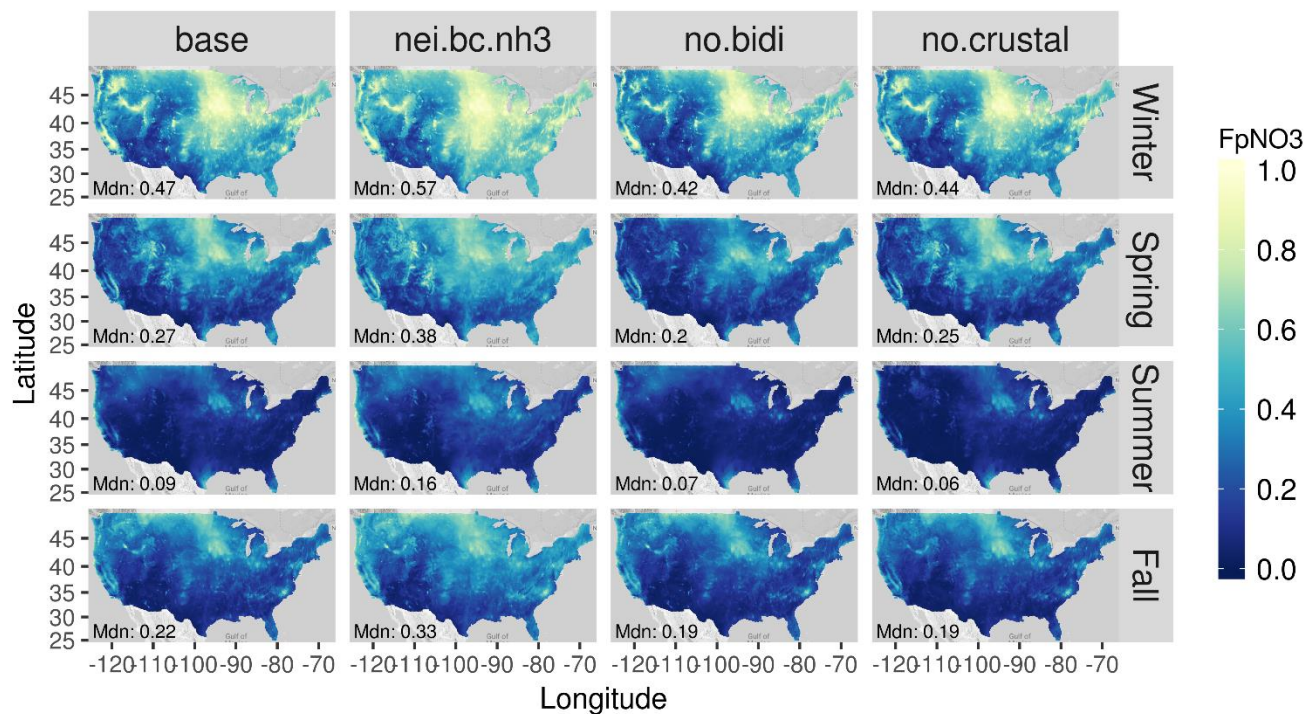


Figure S10. Fraction of total nitrate ($\text{HNO}_3 + \text{NO}_3$) in the particle phase by season for the base, nei.bc.nh3, no.bidi, and no.crustal simulations. Median values across the domain are provided with text.

Table S16. Normalized mean bias (NMB; %) for model predictions of NH₃ at SEARCH and AMoN sites during 2015 in the base, no.bidi, and nei.bc.nh3 simulation cases.

Network	Season	base	no.bidi	nei.bc.nh3
SEARCH	All	-25.8	-37.6	36.5
	Winter	-35.4	-40.9	-10.5
	Spring	-46.5	-53.0	0.6
	Summer	0.1	-26.8	104.8
	Fall	-19.2	-27.9	51.8
AMON	All	-54.0	-66.4	-12.5
	Winter	-70.7	-76.0	-56.4
	Spring	-60.9	-71.6	-29.1
	Summer	-37.3	-59.7	29.5
	Fall	-56.0	-61.7	-18.0

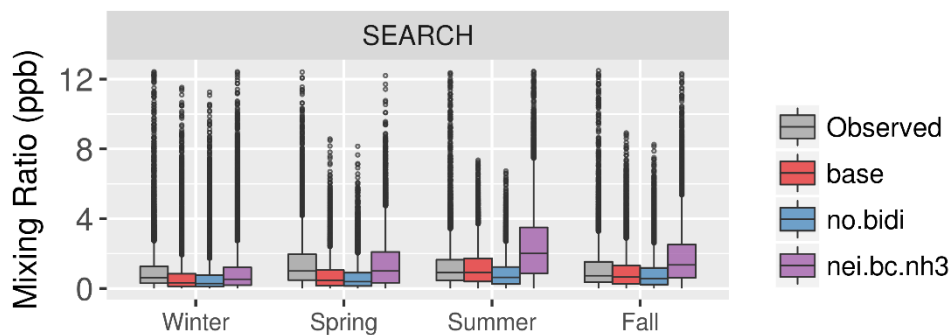


Figure S11. Observed and predicted NH₃ mixing ratios at SEARCH sites; values greater than 12.5 ppb not shown.

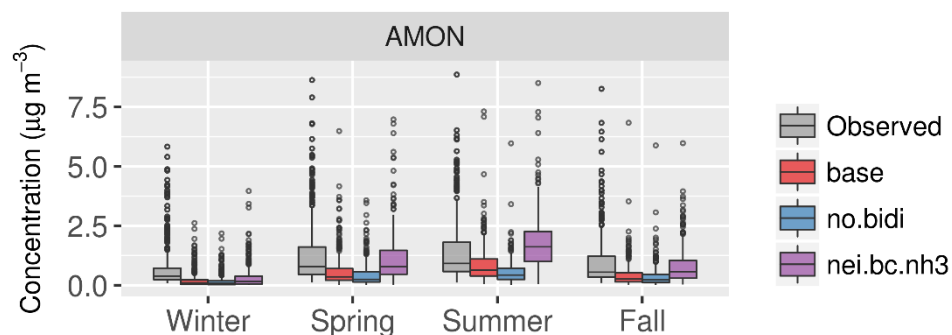


Figure S12. Observed and predicted NH₃ concentrations at AMoN sites; values greater than 9 µg m⁻³ not shown.

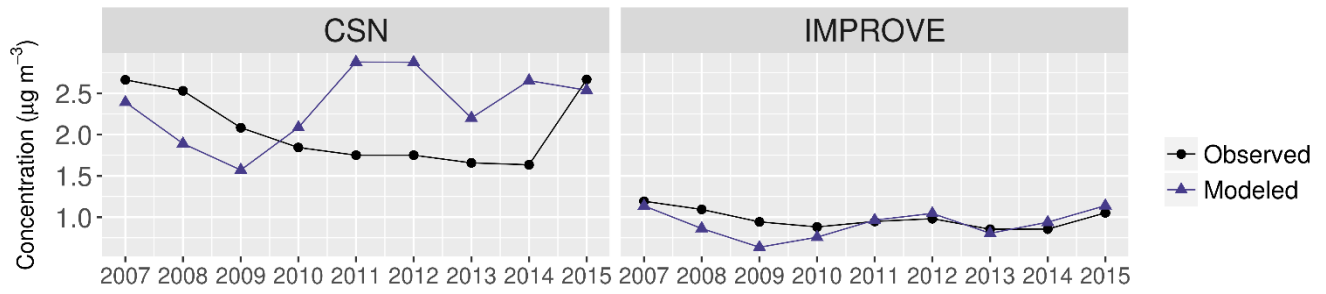


Figure S13. Annual average concentration of OC during 2007-2015.

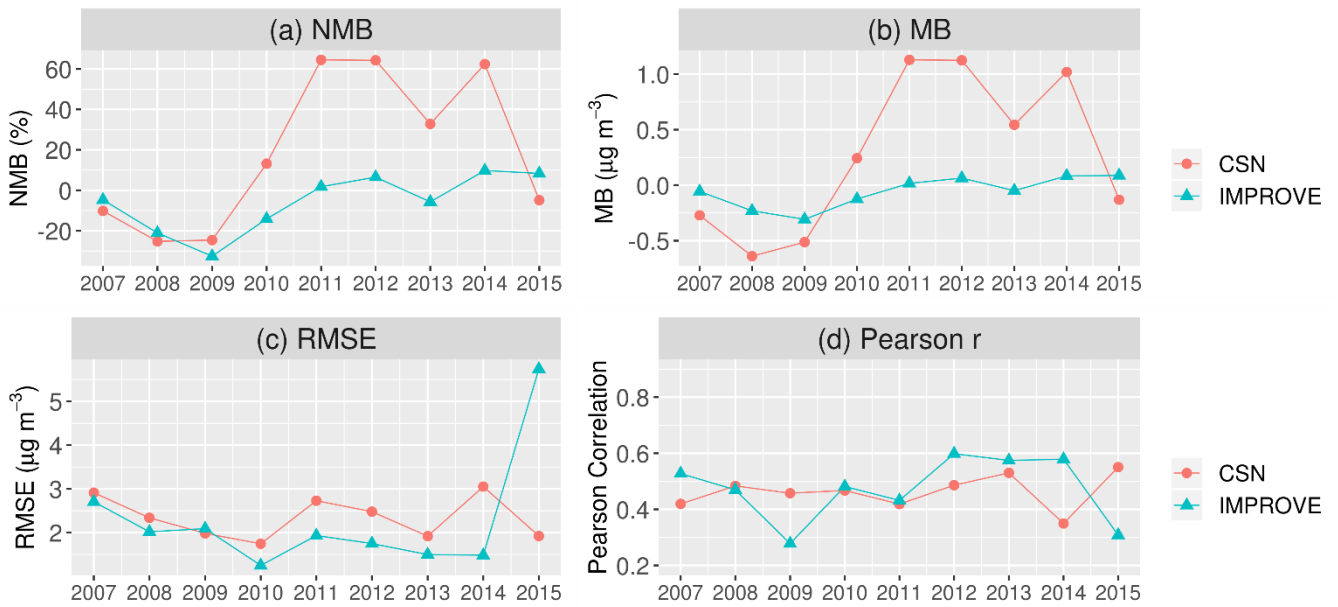


Figure S14. Annual OC model performance statistics by year.

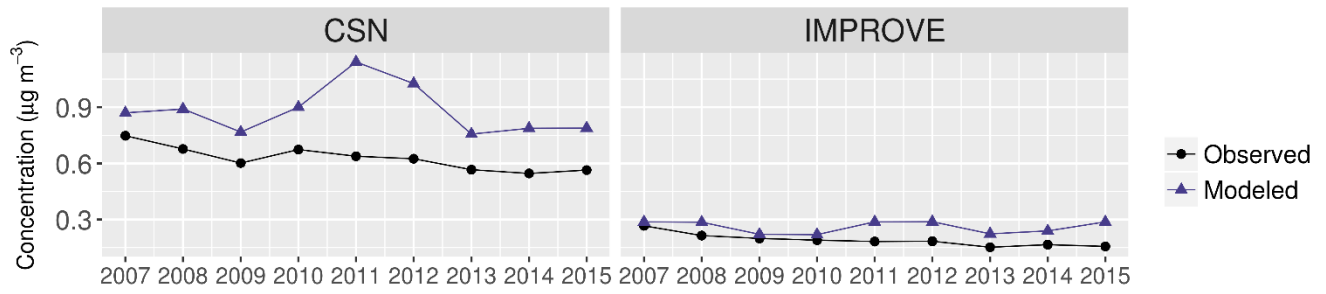


Figure S15. Annual average concentration of EC during 2007-2015.

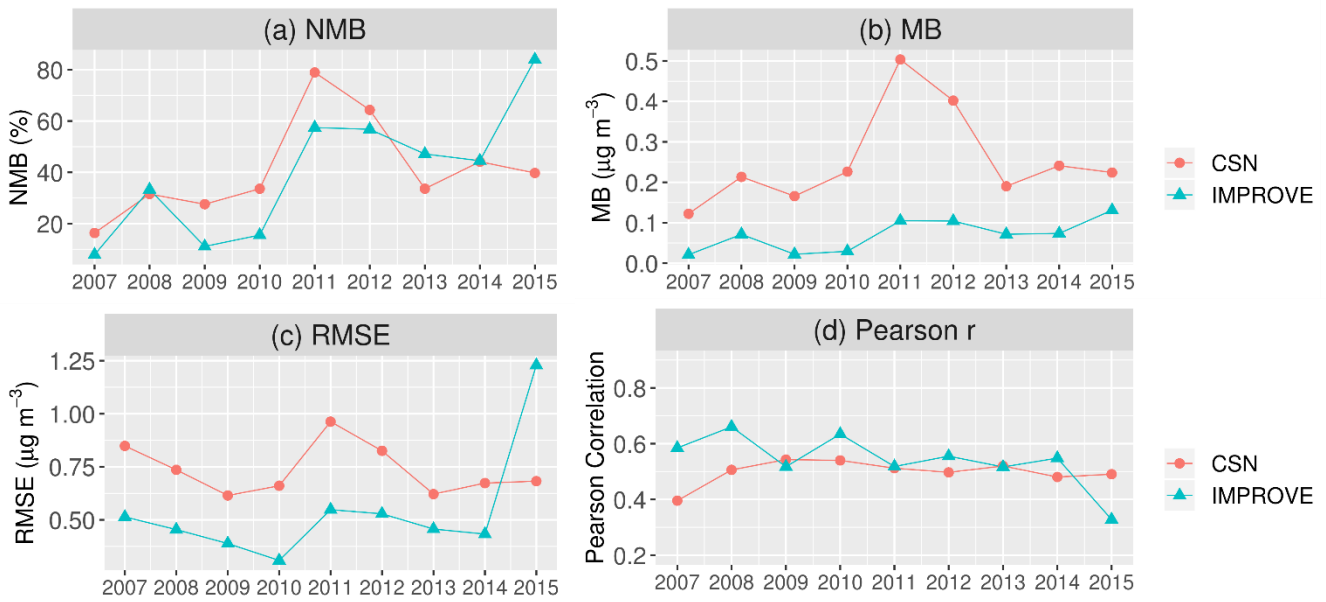


Figure S16. Annual EC model performance statistics by year.

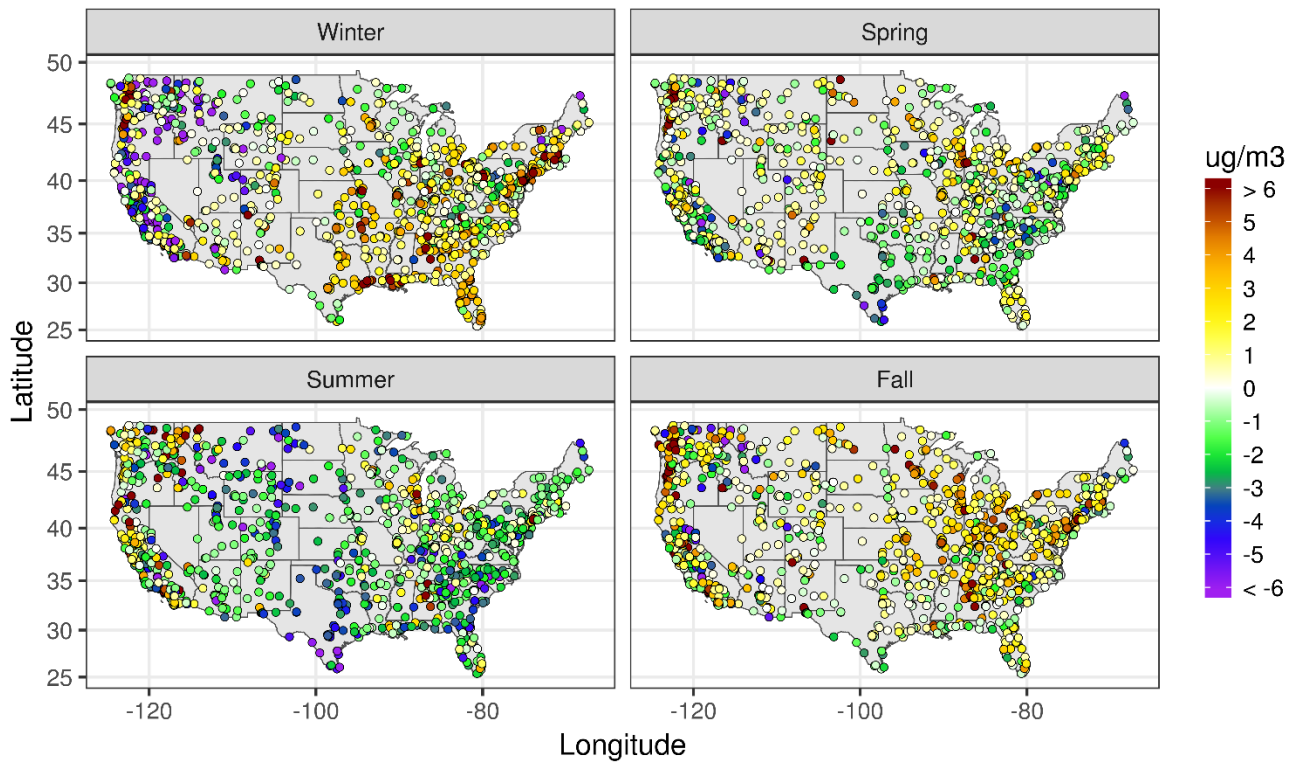


Figure S17. Mean bias (MB; $\mu\text{g m}^{-3}$) by season for PM_{2.5} predictions.

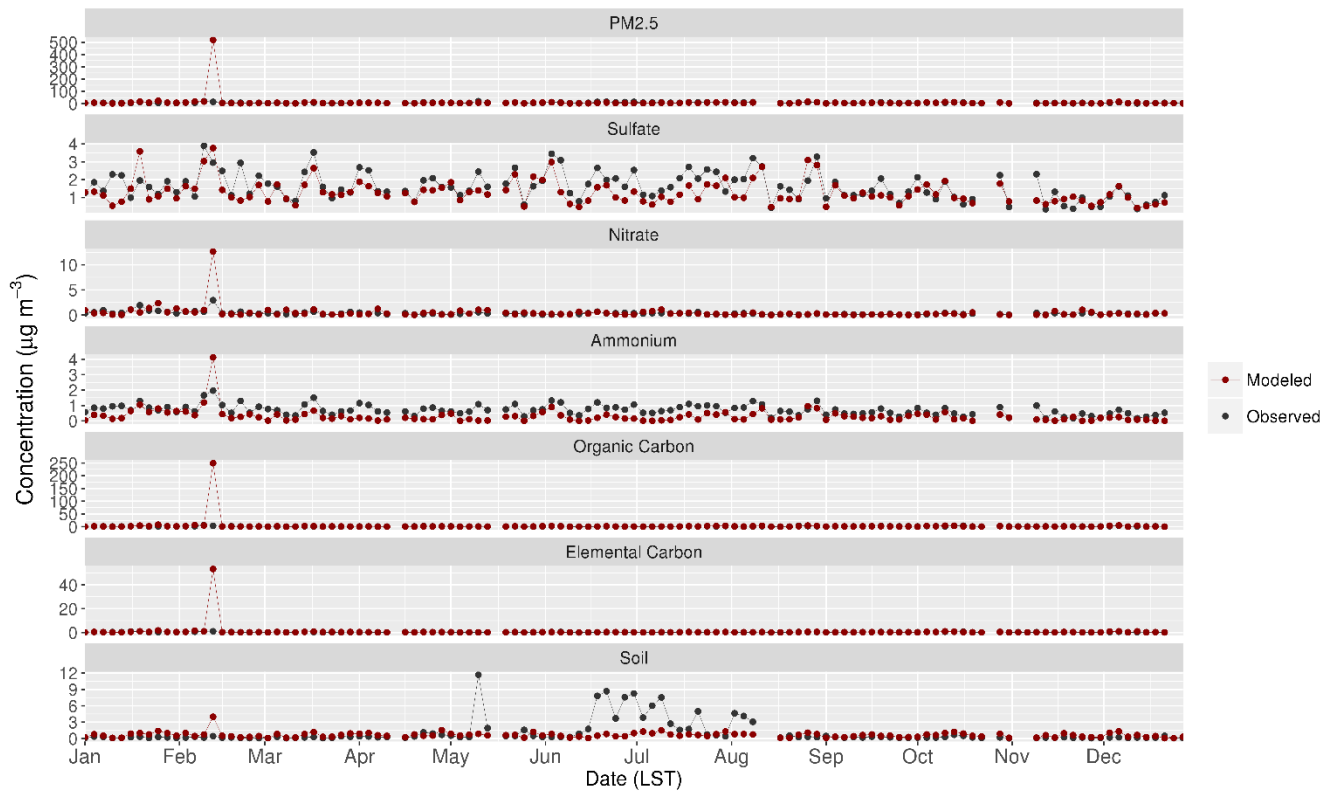


Figure S18. Modeled and observed concentrations of PM^{2.5} and components at the Breton Island IMPROVE site in Louisiana during 2015.

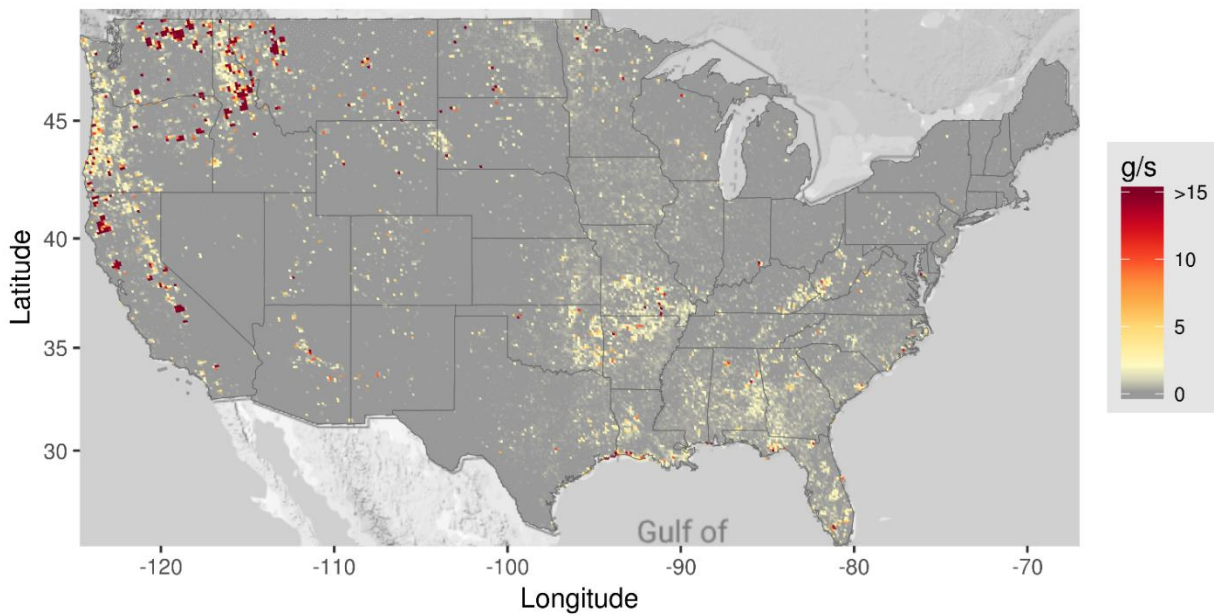


Figure S19. Annual average gridded emission rate (g/s) for primary OC from the ptfire sector.

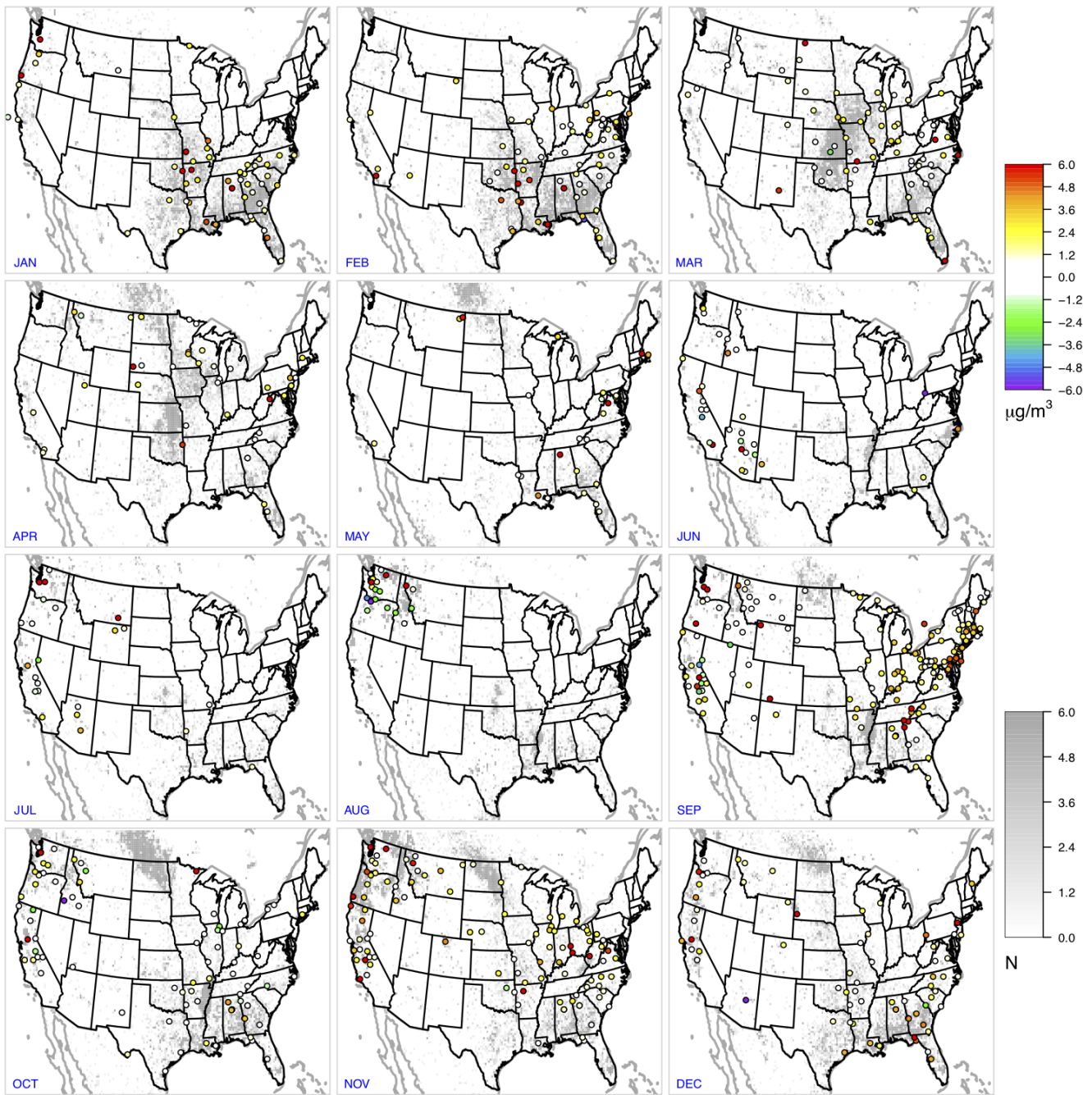


Figure S20. Bias in model predictions of PM_{2.5} OC at IMPROVE and CSN sites on days where the modeled predicted a fire contribution to the concentration. Grey colors indicate the number of Hazard Mapping System fire detections.

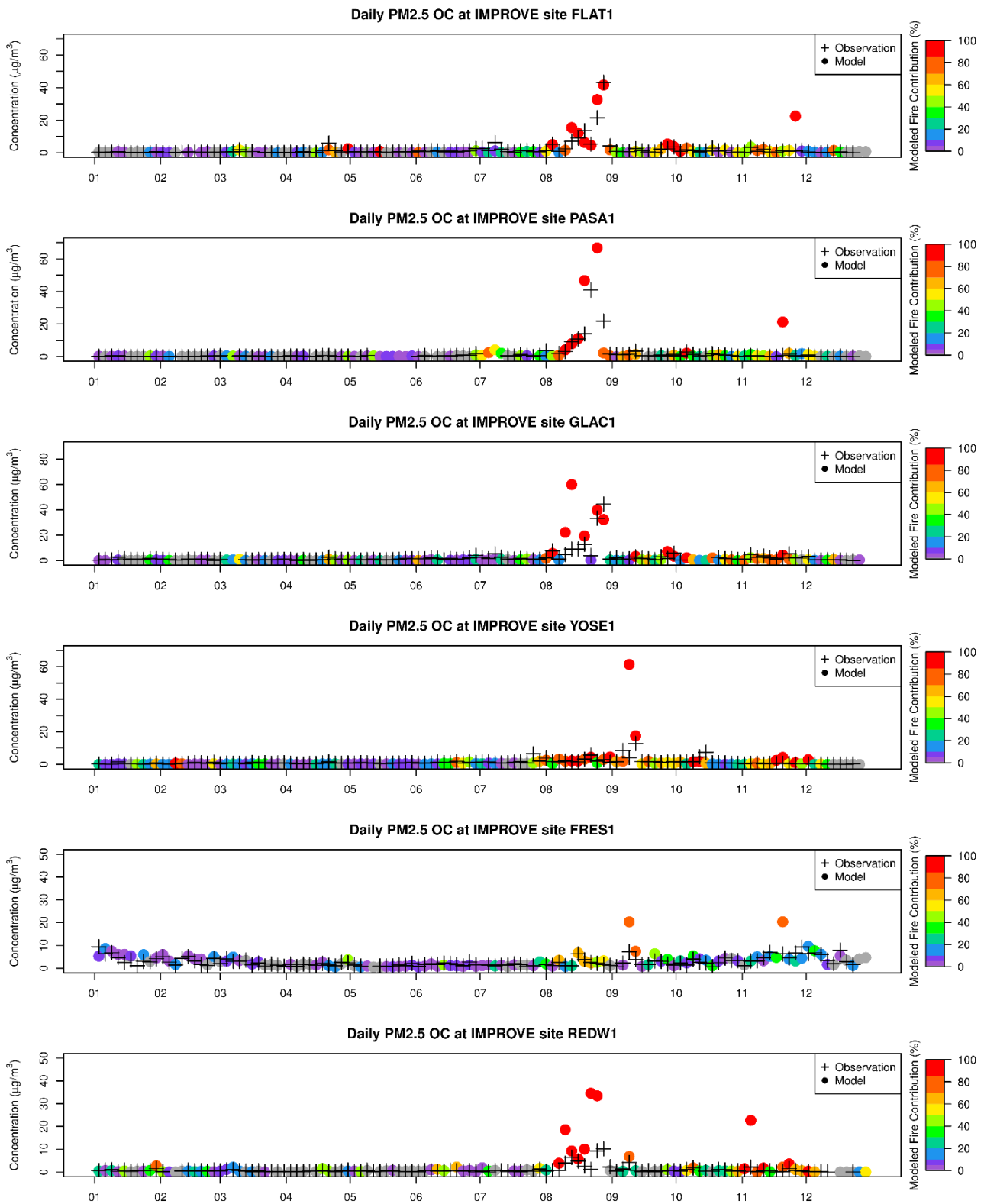


Figure S21. Comparison of PM_{2.5} OC predictions and observations at selected IMPROVE sites colored by the modeled fire contribution. FLAT1: Flathead, MT; PASA1: Pasayten, WA; GLAC1: Glacier National Park, MT; YOSE1: Yosemite National Park, CA; FRES1: Fresno, CA; REDW1: Redwood National Park, CA

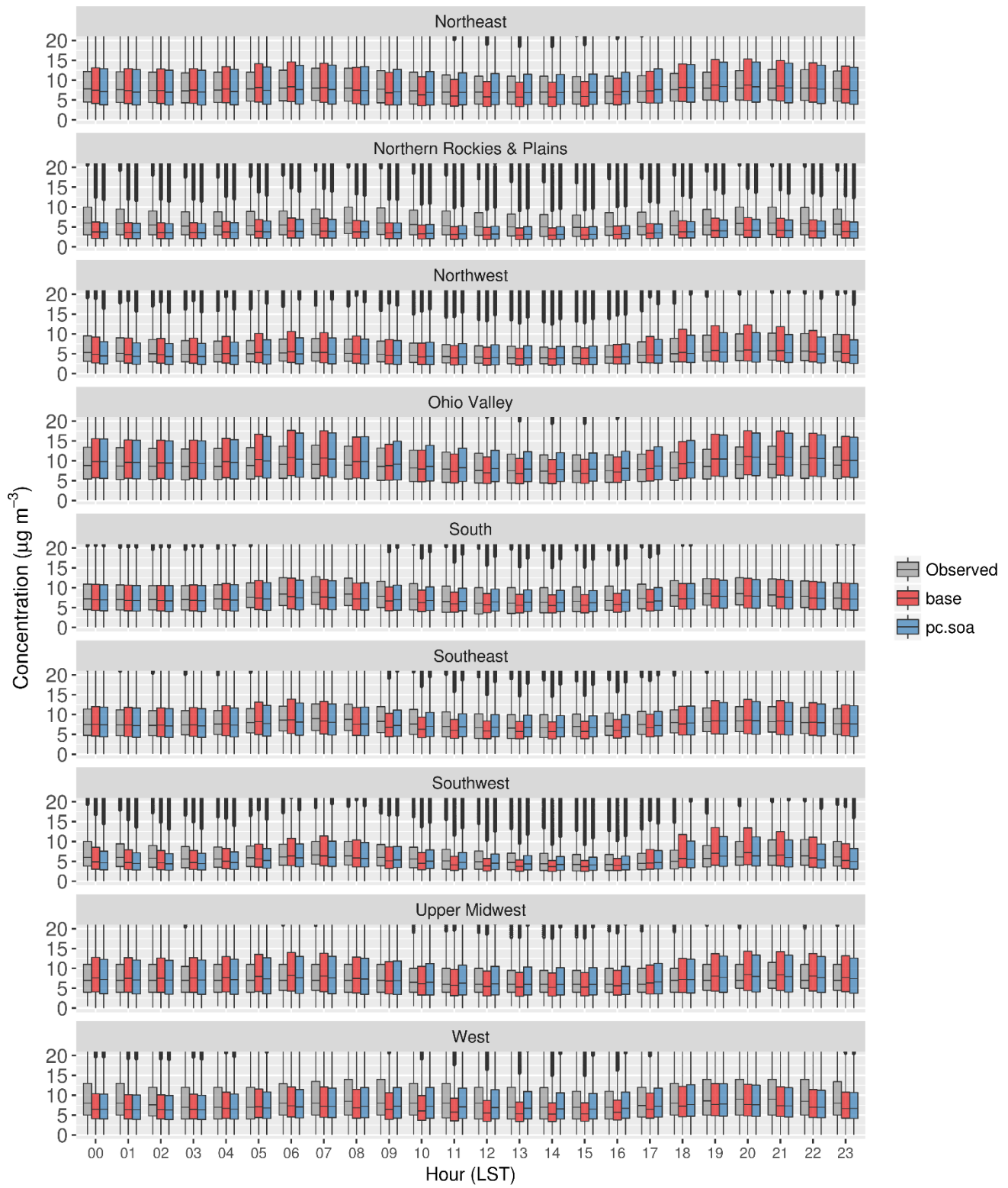


Figure S22. Comparison of hourly modeled and observed PM_{2.5} for the 2015 base case and pc.soa simulations.

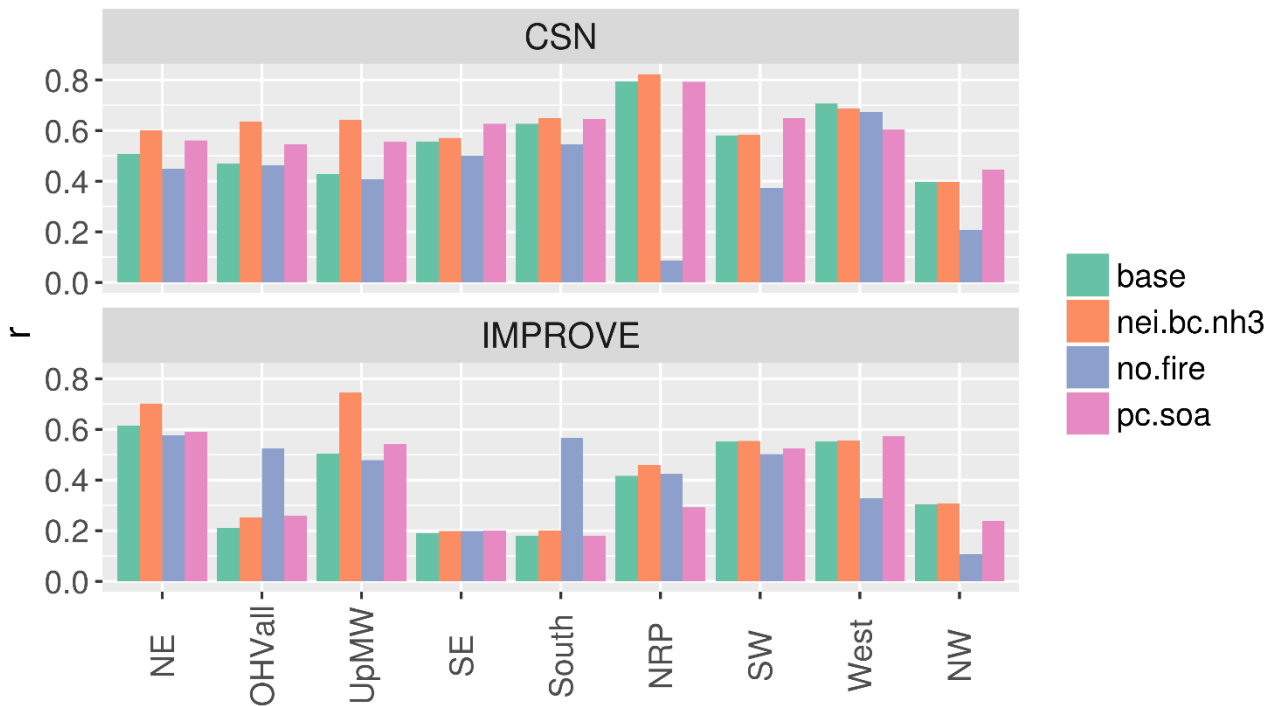


Figure S23. Comparison of annual Pearson correlation coefficient (r) for $PM_{2.5}$ OC in the 2015 base case with that for the wildfire, nei.bc.nh3, no.fire, and pc.soa cases at CSN and IMPROVE sites.

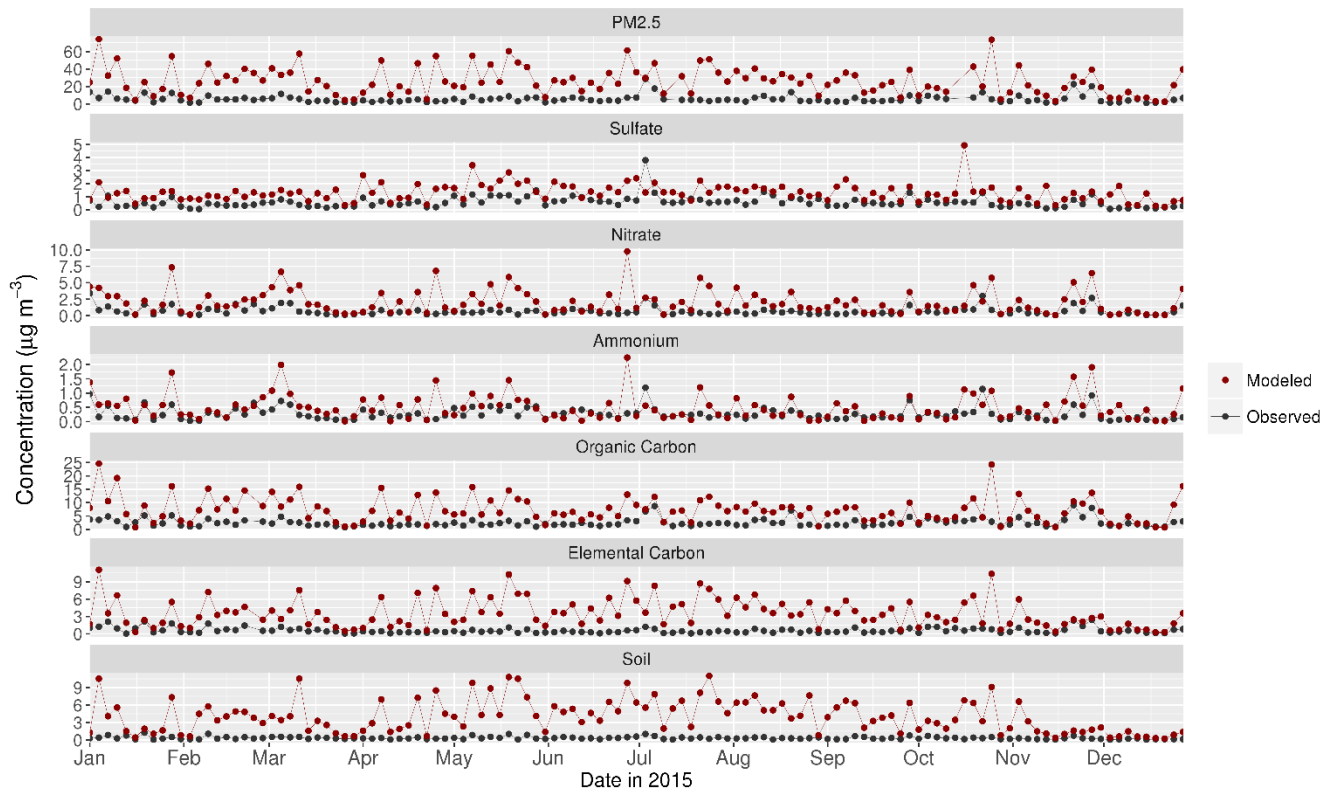


Figure S24. Comparison of 2015 base case model predictions with measurements at the Seattle-Beacon Hill CSN site (530330080). Overpredictions of EC, Soil, and OC appear to be due in part to issues in simulating the planetary boundary layer (PBL) height at this site near the Puget Sound (e.g., see Figure S25).

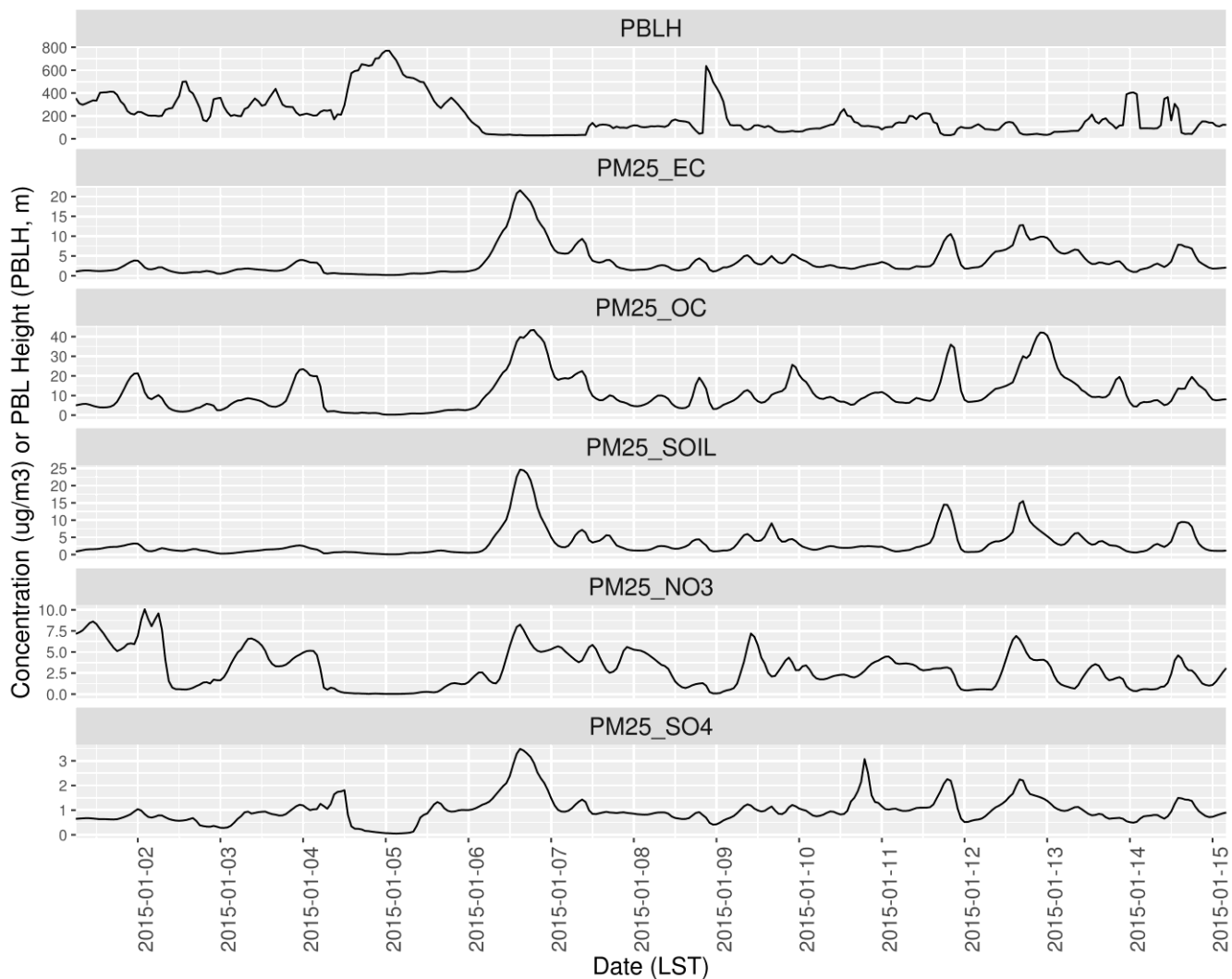


Figure S25. Comparison of 2015 base case PM_{2.5} predictions with modeled PBL height for the grid cell containing the Seattle-Beacon Hill CSN site (530330080). Extremely high concentrations of PM_{2.5} species (e.g., EC and Soil) are predicted on 6 January when the PBL height was generally less than 40 m. Overpredictions of PM_{2.5} species at this site (Figure S24) appear to be due to challenges in simulating PBL height near the Puget Sound coast.

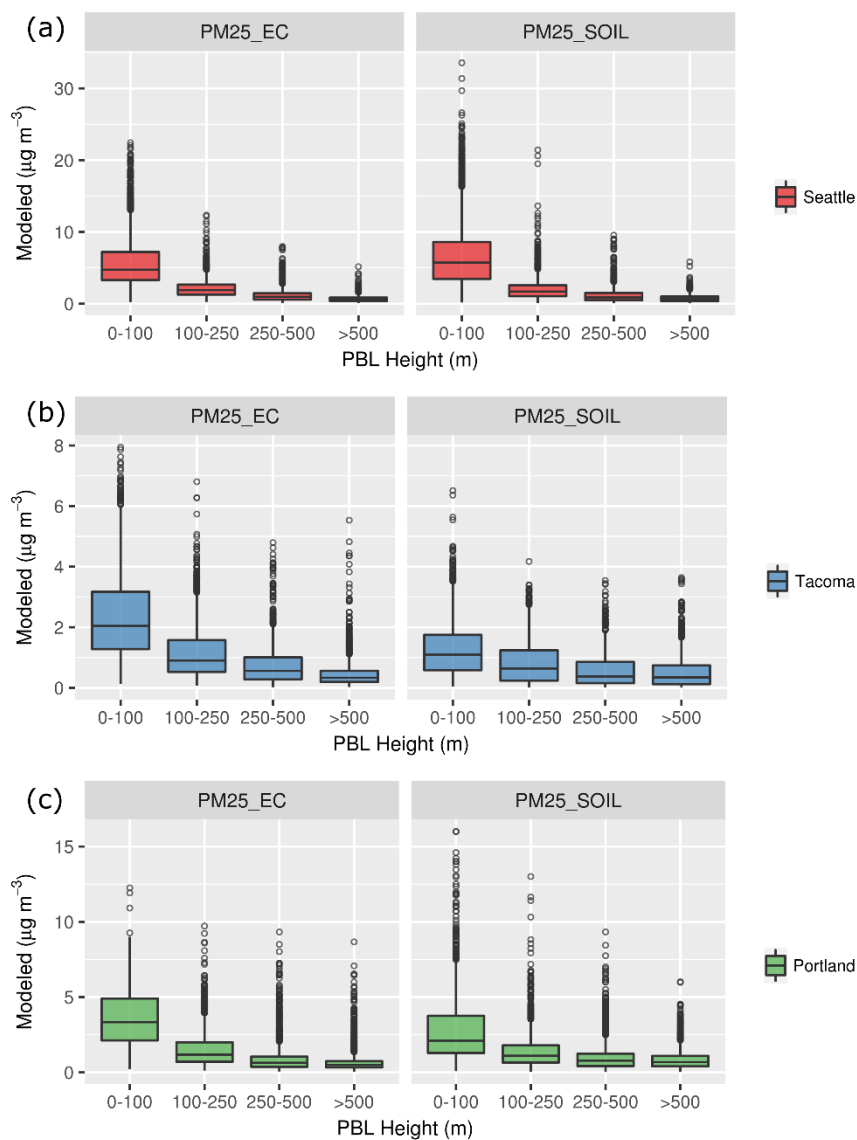


Figure S26. Relationship between simulated PBL height and EC and soil concentration at CSN sites in Seattle, Tacoma, and Portland during 2015. The strong association of concentration and PBL height suggests that overpredictions of EC and soil at these sites is related to the simulation of meteorological mixing.

Disclaimer

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