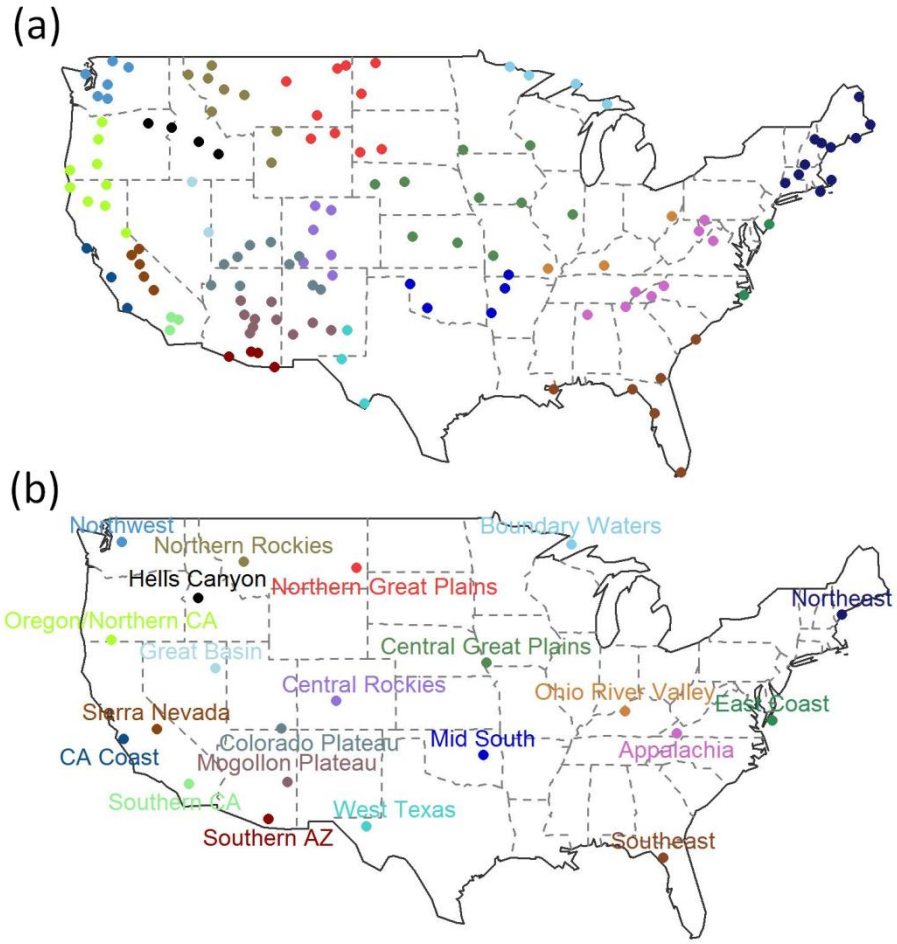

Fig. S1. Locations of all a) IMPROVE network sites and b) chemical climatology regions. Each site is color-coded by region.

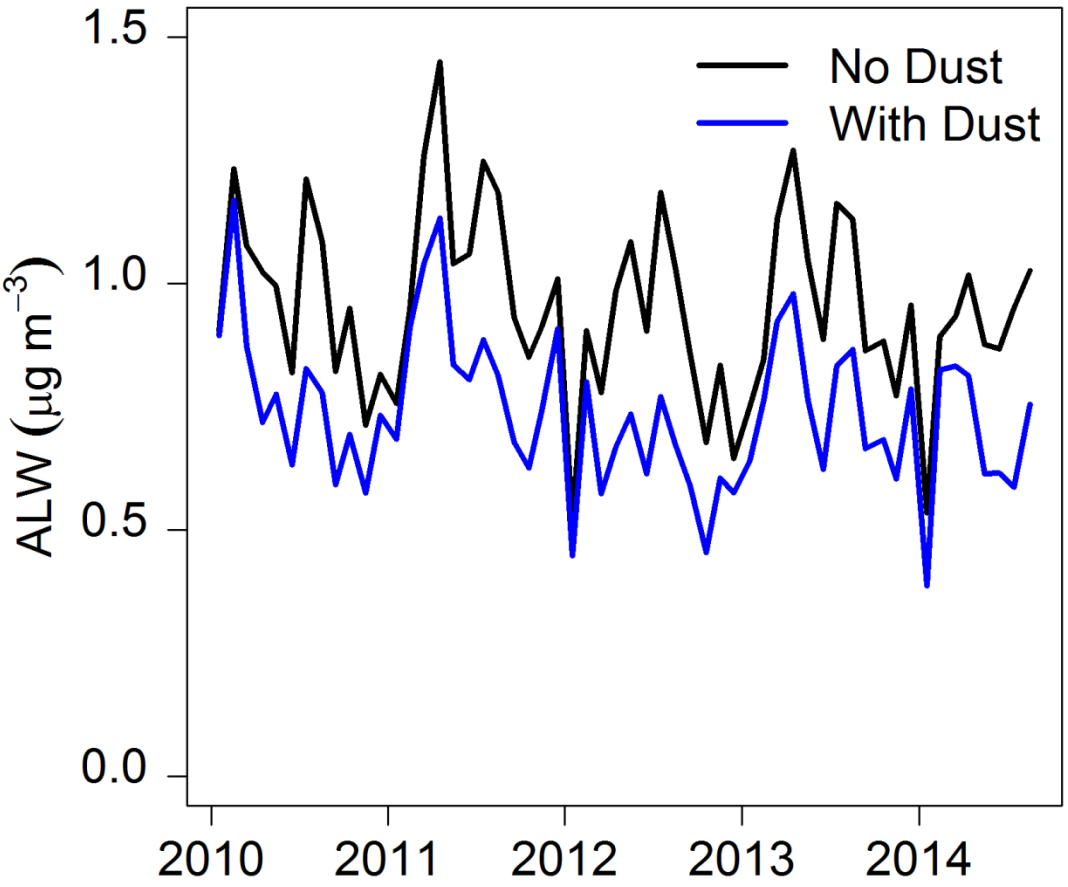

Fig. S2. Median ALW mass concentrations calculated from ISORROPIA with dust (Na^+, Ca^{2+}, K_+, Mg^{2+}, Cl^-) included (blue line) and without dust (black line). In the dust sensitivity, cations form insoluble species with sulfate and precipitate. If ammonium were present, it would compete with dust for sulfate and form more soluble species and increase ALW. Here, the dust sensitivity likely represents a lower bound.

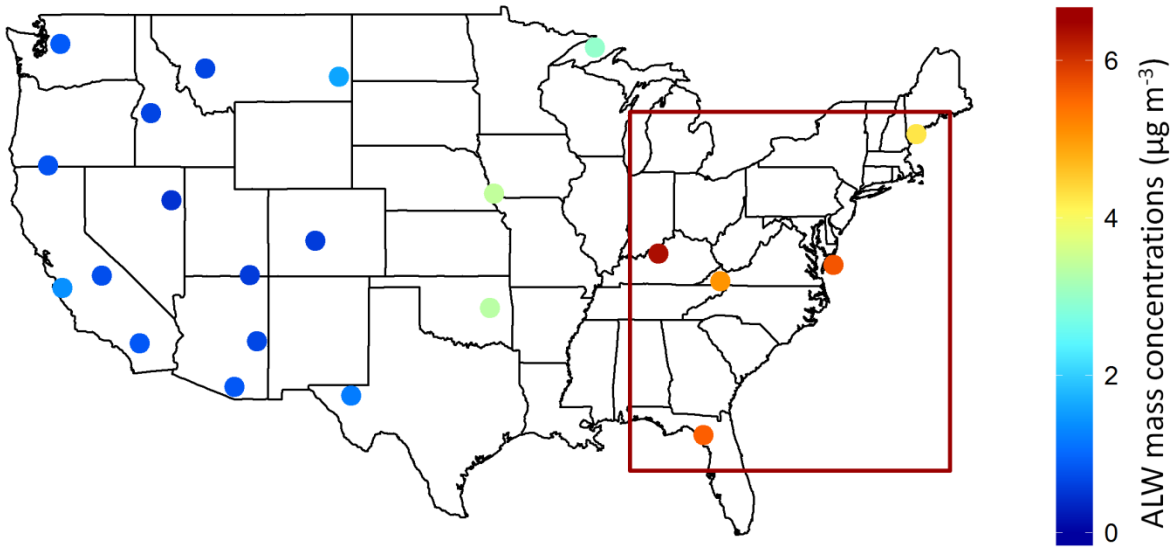

Fig. S3. Average annual ALW mass concentrations across the CONUS. The largest concentrations are located east of the Mississippi River. The boxed area indicates the chemical climatology regions investigated in the eastern US ALW scenarios.

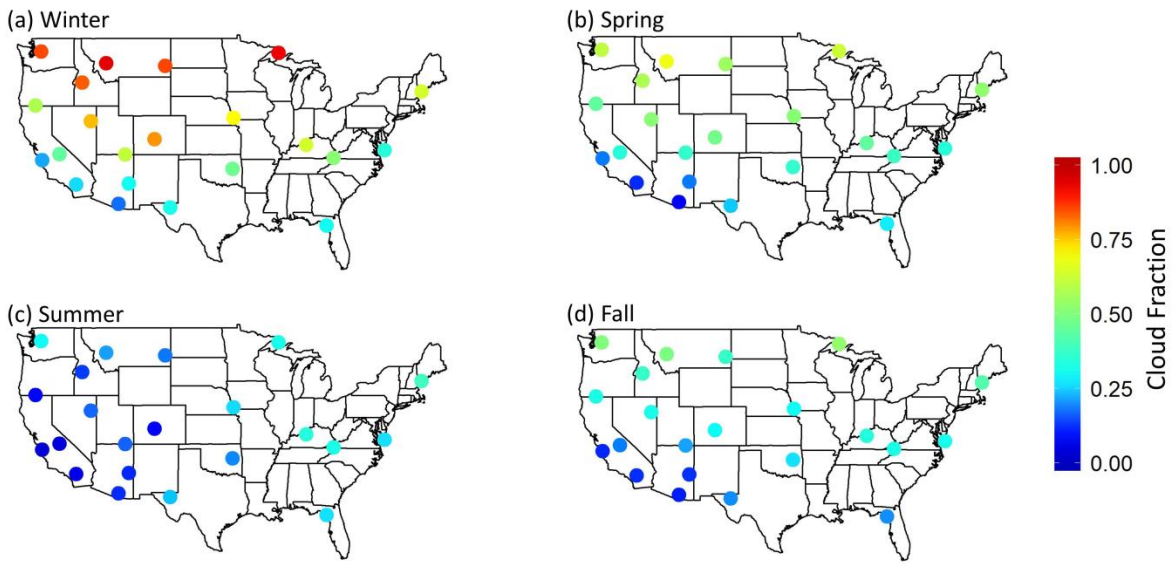

Fig. S4. MODIS cloud fraction across the CONUS from 2010-2014 during a) winter, b) spring, c) summer, and d) fall. A hotter color indicates that a location is in cloud for a greater percentage of time.

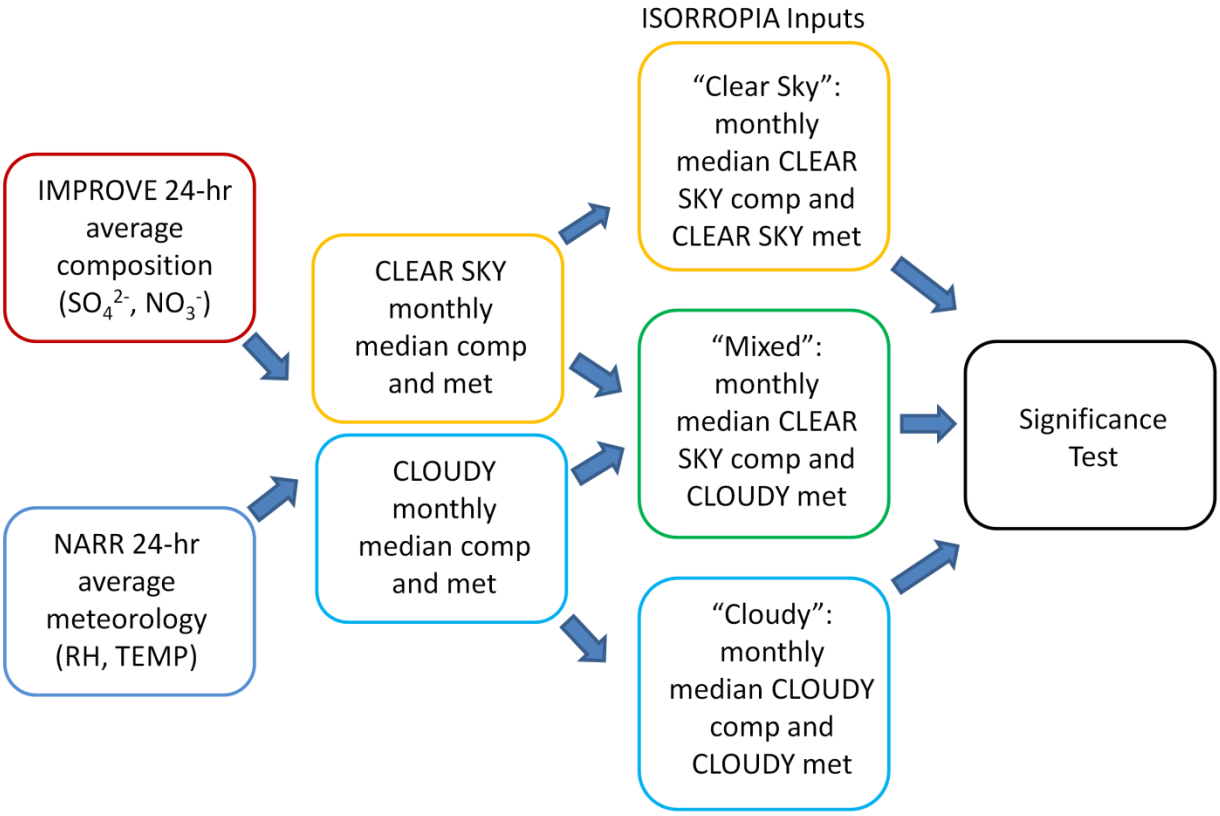

Fig. S5. Flowchart describing how the three scenarios (Cloudy, Clear Sky, and Mixed) for the eastern US are performed and compared.

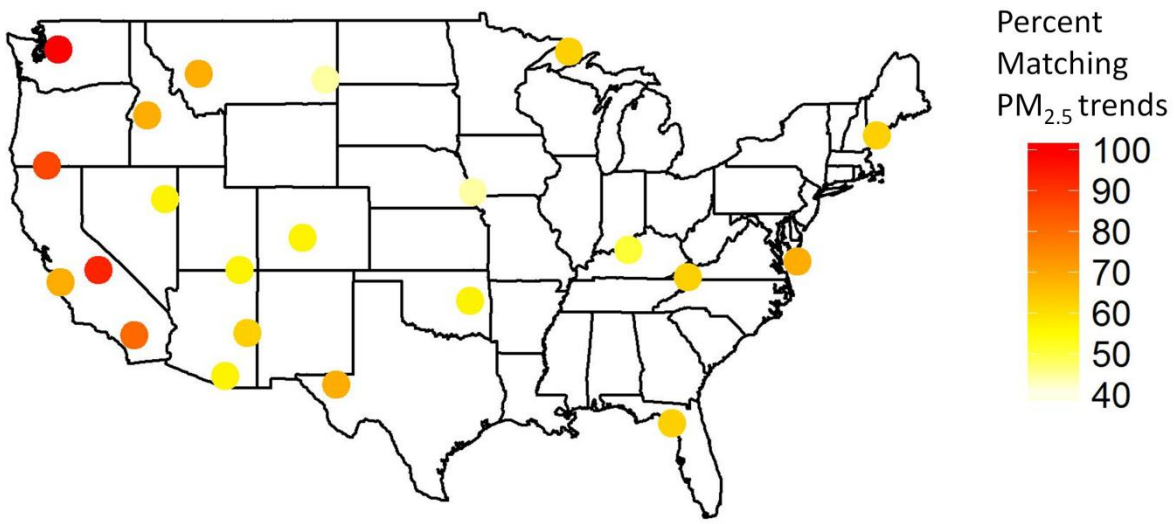

Fig. S6. Percent of $PM_{2.5}$ chemical constituents (SO_4^{2-}, NO_3^-, TOC, ALW) in which cloudy-clear sky directional trends are the same as those of $PM_{2.5}$ in each region across the CONUS. For example, if median daily TOC is greater during Cloudy times, and median daily $PM_{2.5}$ is also greater during Cloudy times, this is counted as a match.

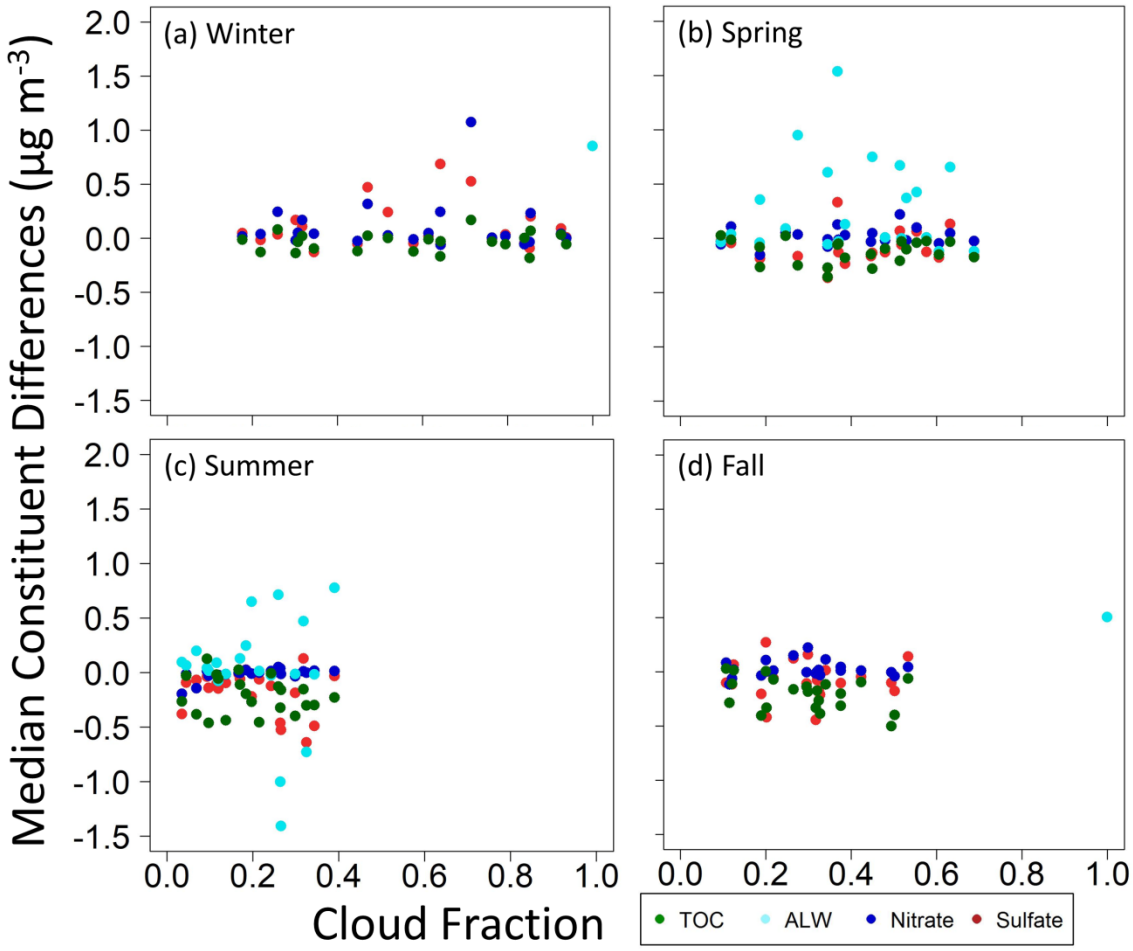

Fig. S7. Plots of median Cloudy – Clear Sky differences by region (dots) in particle chemical constituents (colors) by MODIS median cloud fraction for a) winter, b) spring, c) summer, and d) fall. In each plot, SO_4^{2-} is represented by red, NO_3^- by dark blue, ALW by light blue, and TOC by green.

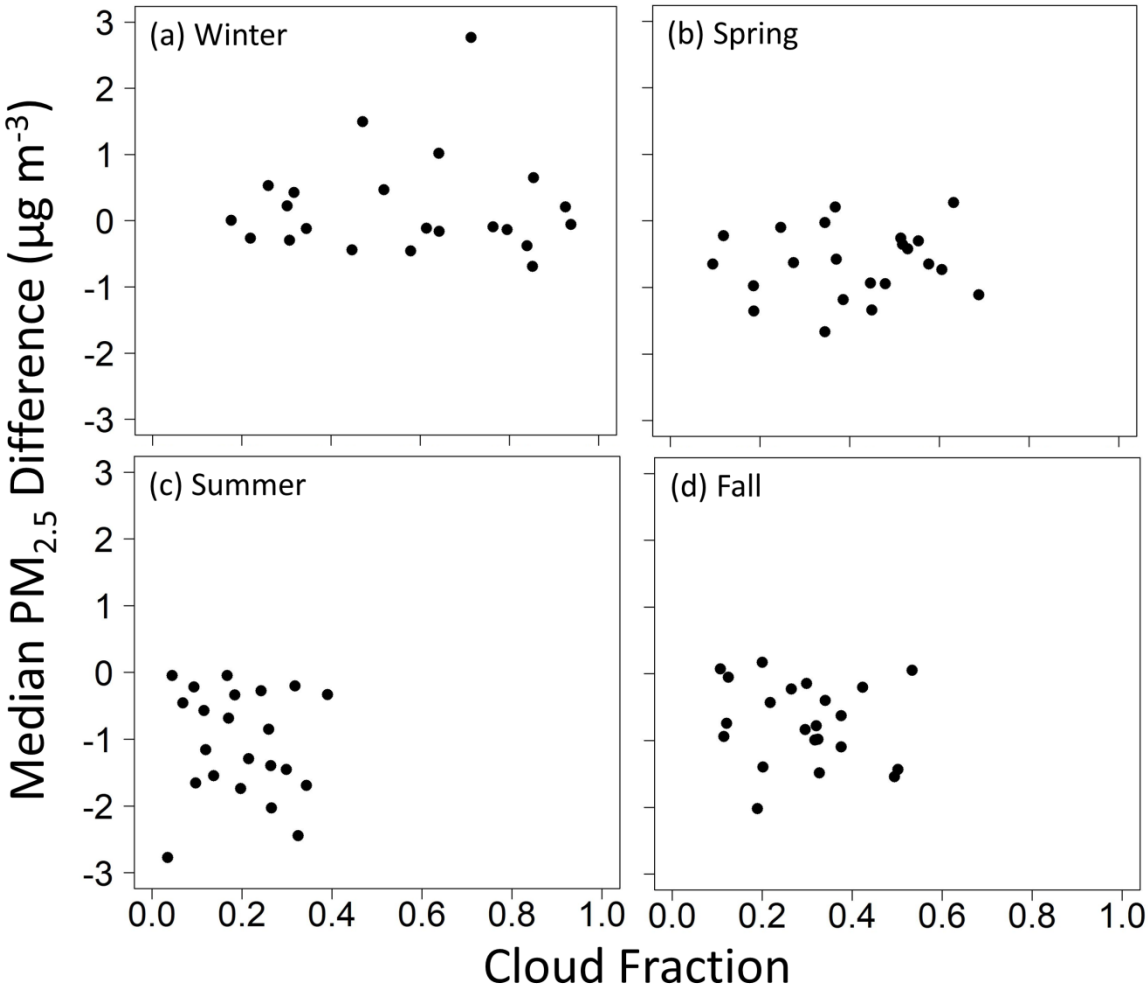

Fig. S8. Plots of median Cloudy – Clear Sky differences by region (dots) in $PM_{2.5}$ by MODIS median cloud fraction for a) winter, b) spring, c) summer, and d) fall.

Table S1. Variable input conditions to ISORROPIA-II for each water uptake scenario.

	"Clear Sky"	**"Cloudy"**	**"Mixed"**
Meteorology **MODIS classification – Clear Sky**	√		
Meteorology **MODIS classification – Cloudy**		√	√
$PM_{2.5}$ Chemical Composition **MODIS classification – Clear Sky**	√		√
$PM_{2.5}$ Chemical Composition **MODIS classification – Cloudy**		√	

Table S2. Cloudy – Clear Sky and Cloudy – Mixed differences in each of the chemical climatology regions that comprise the eastern US. Negative values indicate that Cloudy concentrations are larger.

Season	Cloudy – Clear SO_4^{2-}	Cloudy – Clear NO_3^-	Cloudy – Clear RH	Cloudy – Clear ALW	Cloudy – Mixed ALW
	Ohio River Valley				
Winter	0.57	0.26	0.08	4.08	2.39
Spring	-0.15	0.00	0.04	0.83	-0.44
Summer	-0.28	0.01	0.05	0.75	-0.67
Fall	-0.01	-0.01	0.10	1.17	-0.02
	Appalachia				
Winter	0.19	0.02	0.11	1.36	0.56
Spring	-0.17	-0.01	0.08	0.83	-0.45
Summer	-0.59	0.00	0.04	-0.83	-1.61
Fall	-0.12	0.03	0.09	0.79	-0.35
	Southeast				
Winter	0.16	-0.03	0.06	1.74	0.56
Spring	-0.21	-0.02	0.06	0.89	-0.61
Summer	-0.41	-0.03	0.01	-1.12	-1.20
Fall	-0.18	-0.02	0.04	-0.13	-0.56
	East Coast				
Winter	-0.20	0.06	0.10	2.01	-0.51
Spring	-0.21	-0.07	0.07	0.71	-0.86
Summer	0.07	0.02	0.03	1.23	0.22
Fall	-0.14	0.02	0.08	0.35	-0.50
	Northeast				
Winter	-0.06	-0.07	0.10	0.57	-0.25
Spring	-0.05	-0.06	0.09	0.47	-0.15
Summer	0.07	0.01	0.05	0.90	0.22
Fall	-0.02	0.00	0.08	0.25	-0.10

Table S3. Seasonal differences in ALW and RH medians (cloudy-clear sky) in all chemical climatology regions. Negative values indicate that clear sky temperatures are larger than cloudy. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring		Summer		Fall		Winter	
	ALW	RH	ALW	RH	ALW	RH	ALW	RH
Northwest	***-0.12***	***0.12***	-0.02	***0.13***	***-0.09***	***0.22***	***-0.12***	***0.08***
OR/NorCal	***-0.15***	***0.13***	0.01	***0.26***	***-0.05***	***0.30***	0.00	***0.19***
CA Coast	***0.35***	***0.21***	0.09	***0.39***	***0.66***	***0.28***	***0.24***	***0.21***
Sierra NV	-0.06	***0.23***	0.06	***0.09***	-0.03	***0.24***	-0.01	***0.15***
SoCal	0.04	***0.03***	0.20	***0.16***	-0.09	***0.10***	***0.16***	***0.23***
HellsCyn	0.01	***0.13***	-0.01	***0.20***	***0.07***	***0.25***	-0.02	***0.06***
Great Basin	0.00	***0.12***	***0.13***	***0.11***	0.02	***0.15***	0.01	***0.08***
N Rockies	***-0.12***	***0.13***	0.01	***0.14***	***0.06***	***0.21***	-0.01	0.00
CO Plateau	-0.03	***0.08***	0.02	***0.05***	***0.14***	***0.18***	***0.15***	***0.15***
Mogollon Plateau	-0.04	***0.05***	0.09	***0.08***	***0.15***	***0.11***	***0.15***	***0.15***
Southern AZ	-0.04	0.01	-0.08	***0.11***	0.15	***0.07***	0.05	0.00
N Great Plains	***0.42***	***0.18***	***0.25***	***0.11***	***0.21***	***0.15***	***1.01***	***0.12***
Central Rockies	0.01	***0.15***	0.04	***0.08***	0.04	***0.17***	***0.14***	***0.14***
West TX	0.09	0.03	-0.03	0.01	***0.38***	***0.09***	***0.22***	0.02
Central Great Plains	***0.67***	***0.10***	***0.71***	***0.11***	***0.59***	***0.12***	***2.48***	***0.14***
Mid South	***1.54***	***0.14***	***0.65***	***0.13***	***1.18***	***0.16***	***2.29***	***0.16***
Boundary Waters	***0.65***	***0.12***	***0.47***	***0.06***	***0.66***	***0.07***	0.13	***0.02***
OH River Valley	***0.75***	***0.07***	-0.02	***0.05***	***1.50***	***0.11***	***4.58***	***0.10***
Appalachia	0.13	***0.09***	***-0.73***	***0.05***	***0.81***	***0.12***	***1.51***	***0.12***
Southeast	***0.95***	***0.06***	***-1.41***	***0.01***	***-0.70***	***0.04***	***1.63***	***0.06***
E Coast	0.61	***0.10***	-1.00	***0.04***	0.40	***0.08***	***1.35***	***0.11***
Northeast	***0.37***	***0.10***	***0.78***	***0.06***	***0.43***	***0.09***	***0.75***	***0.10***

Table S4. Seasonal differences in reported $PM_{2.5}$ mass concentration medians (cloudy-clear sky) in all chemical climatology regions. Negative values indicate that clear sky concentrations are larger than cloudy. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring	Summer	Fall	Winter
Northwest	***-0.73***	***-1.46***	***-1.44***	***-0.69***
OR/NorCal	***-0.94***	***-1.66***	***-1.49***	***-0.46***
CA Coast	***-1.36***	***-2.77***	***-0.94***	-0.27
Sierra NV	***-1.67***	-0.05	***-2.02***	***-0.44***
SoCal	-0.23	-0.46	***-0.75***	***0.53***
HellsCyn	***-0.65***	***-1.55***	***-1.10***	***-0.38***
Great Basin	***-0.36***	***-0.69***	***-0.78***	-0.10
N Rockies	***-1.12***	***-1.30***	***-1.55***	-0.06
CO Plateau	***-0.58***	-0.05	***-0.43***	***-0.12***
Mogollon Plateau	***-0.98***	***-0.57***	-0.06	***-0.30***
Southern AZ	-0.65	***-1.16***	0.07	0.00
N Great Plains	***-0.30***	***-0.34***	***-0.63***	***0.65***
Central Rockies	***-0.95***	-0.22	***-0.84***	***-0.14***
West TX	-0.10	-0.28	0.17	***0.42***
Central Great Plains	***-0.26***	***-0.85***	-0.15	***2.76***
Mid South	0.20	***-1.74***	-0.23	***1.49***
Boundary Waters	0.27	-0.20	0.05	0.20
OH River Valley	***-1.34***	***-1.70***	-0.40	***1.02***
Appalachia	***-1.19***	***-2.45***	***-0.99***	***0.46***
Southeast	***-0.63***	***-2.03***	***-1.40***	0.22
E Coast	***-0.03***	***-1.40***	***-0.99***	-0.12
Northeast	***-0.42***	***-0.33***	***-0.21***	-0.16

Table S5. Seasonal differences in SO_4^{2-} mass concentration medians (cloudy-clear sky) in all chemical climatology regions. Negative values indicate that clear sky SO_4^{2-} concentrations are larger than cloudy. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring	Summer	Fall	Winter
Northwest	***-0.18***	***-0.19***	***-0.18***	***-0.09***
OR/NorCal	***-0.17***	***-0.14***	***-0.21***	***-0.05***
CA Coast	***-0.19***	***-0.38***	-0.12	-0.02
Sierra NV	***-0.27***	-0.09	***-0.21***	***-0.05***
SoCal	-0.04	-0.07	-0.12	0.03
HellsCyn	***-0.13***	***-0.10***	***-0.11***	***-0.05***
Great Basin	***-0.06***	-0.06	***-0.08***	-0.01
N Rockies	***-0.16***	***-0.06***	***-0.10***	0.01
CO Plateau	***-0.13***	-0.01	***-0.06***	0.02
Mogollon Plateau	***-0.09***	***-0.11***	0.06	-0.01
Southern AZ	***-0.03***	***-0.15***	-0.10	0.04
N Great Plains	***0.06***	0.02	0.04	***0.20***
Central Rockies	***-0.13***	***-0.04***	***-0.11***	***0.03***
West TX	0.08	-0.12	***0.27***	0.11
Central Great Plains	0.07	0.00	***0.16***	***0.52***
Mid South	***0.33***	***-0.22***	0.12	***0.47***
Boundary Waters	***0.13***	***0.13***	***0.14***	0.09
OH River Valley	-0.14	***-0.49***	0.01	***0.68***
Appalachia	***-0.24***	***-0.64***	***-0.17***	***0.24***
Southeast	***-0.17***	***-0.53***	***-0.42***	***0.17***
E Coast	***-0.37***	-0.46	***-0.44***	-0.13
Northeast	***-0.10***	-0.03	***-0.05***	-0.05

Table S6. Seasonal differences in NO_3^- mass concentration medians (cloudy-clear sky) in all chemical climatology regions. Negative values indicate that clear sky NO_3^- concentrations are larger than cloudy. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring	Summer	Fall	Winter
Northwest	***-0.05***	***-0.03***	***-0.04***	***-0.04***
OR/NorCal	***-0.03***	***-0.03***	***-0.03***	***-0.01***
CA Coast	***-0.15***	***-0.20***	***-0.12***	0.04
Sierra NV	***-0.08***	-0.02	-0.04	***-0.03***
SoCal	0.11	-0.14	-0.06	***0.24***
HellsCyn	***-0.02***	***-0.02***	0.01	***-0.06***
Great Basin	0.00	-0.01	0.00	0.00
N Rockies	***-0.03***	***0.00***	-0.01	0.00
CO Plateau	***-0.02***	0.00	***0.01***	***0.04***
Mogollon Plateau	***-0.04***	***-0.02***	***0.02***	***0.05***
Southern AZ	***-0.06***	***-0.02***	***0.08***	0.01
N Great Plains	***0.10***	***0.02***	***0.04***	***0.23***
Central Rockies	***-0.02***	0.01	***-0.01***	0.02
West TX	***0.05***	0.01	***0.11***	***0.17***
Central Great Plains	***0.22***	***0.05***	***0.22***	***1.07***
Mid South	***0.13***	-0.01	***0.15***	***0.32***
Boundary Waters	***0.05***	***0.01***	***0.04***	0.04
OH River Valley	0.05	0.01	0.11	0.24
Appalachia	0.03	0.00	0.01	0.02
Southeast	0.03	***-0.02***	0.00	-0.02
E Coast	-0.01	***0.04***	-0.02	0.04
Northeast	***-0.02***	0.01	0.01	***-0.06***

Table S7. Seasonal differences in TOC mass concentration medians (cloudy-clear sky) in all chemical climatology regions. Negative values indicate that clear sky TOC concentrations are larger than cloudy. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring	Summer	Fall	Winter
Northwest	***-0.15***	***-0.40***	***-0.40***	***-0.18***
OR/NorCal	***-0.15***	***-0.46***	***-0.39***	***-0.12***
CA Coast	***-0.27***	***-0.27***	***-0.29***	-0.13
Sierra NV	***-0.36***	-0.03	***-0.41***	***-0.12***
SoCal	-0.01	***-0.38***	***-0.11***	***0.08***
HellsCyn	-0.03	***-0.44***	***-0.32***	0.00
Great Basin	***-0.03***	-0.11	***-0.17***	-0.03
N Rockies	***-0.18***	***-0.46***	***-0.50***	-0.06
CO Plateau	***-0.05***	0.03	***-0.07***	-0.01
Mogollon Plateau	***-0.08***	-0.02	0.01	***-0.04***
Southern AZ	0.02	-0.06	0.03	-0.01
N Great Plains	***-0.04***	***-0.20***	***-0.20***	***0.07***
Central Rockies	***-0.10***	0.13	***-0.14***	***-0.06***
West TX	0.02	-0.01	0.00	0.02
Central Great Plains	***-0.21***	***-0.13***	***-0.18***	***0.17***
Mid South	-0.06	***-0.27***	***-0.16***	0.02
Boundary Waters	***-0.03***	***-0.16***	***-0.06***	0.03
OH River Valley	***-0.28***	***-0.30***	***-0.12***	***-0.17***
Appalachia	***-0.18***	***-0.30***	***-0.26***	0.00
Southeast	***-0.25***	***-0.16***	***-0.33***	***-0.14***
E Coast	***-0.27***	***-0.32***	***-0.33***	***-0.10***
Northeast	***-0.10***	***-0.23***	***-0.10***	-0.03

Table S8. Seasonal differences in temperature medians (cloudy-clear sky) in all chemical climatology regions. Negative values indicate that clear sky temperatures are larger than cloudy. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring	Summer	Fall	Winter
Northwest	***-1.65***	***-3.69***	***-4.60***	-0.41
OR/NorCal	***-1.89***	***-3.67***	***-6.26***	***-1.02***
CA Coast	***-1.42***	***-4.41***	***-3.40***	0.54
Sierra NV	***-5.91***	0.44	***-7.64***	***-1.65***
SoCal	-2.40	2.60	-2.85	-0.37
HellsCyn	***-3.10***	***-4.87***	***-7.93***	***-1.47***
Great Basin	***-2.50***	-0.13	***-4.99***	***-0.65***
N Rockies	***-4.07***	***-4.60***	***-7.10***	***-2.40***
CO Plateau	***-4.31***	***1.26***	***-5.62***	***-4.36***
Mogollon Plateau	***-3.78***	0.56	***-3.34***	***-2.03***
Southern AZ	-0.46	0.43	***-4.54***	-0.21
N Great Plains	***-4.92***	***-2.78***	***-7.49***	***-4.58***
Central Rockies	***-6.33***	***-1.01***	***-7.29***	***-4.11***
West TX	-0.84	-0.33	-0.01	0.22
Central Great Plains	***-3.42***	***-1.82***	***-1.94***	***-4.10***
Mid South	-0.37	***-2.13***	-0.44	-0.21
Boundary Waters	***-5.10***	-0.86	***-4.34***	***-5.71***
OH River Valley	-0.94	***-1.29***	0.45	-1.33
Appalachia	***-2.11***	***-0.93***	0.88	-0.79
Southeast	***-1.43***	***-0.80***	0.84	***0.69***
E Coast	-0.72	***-0.85***	2.06	***1.97***
Northeast	***-3.28***	***-0.80***	***-1.38***	***-1.68***

Table S9. Seasonal all sky (clear and cloudy data) and clear sky differences in $PM_{2.5}$ mass concentration medians (all sky-clear sky) in all chemical climatology regions. Negative values indicate that clear sky $PM_{2.5}$ concentrations are larger than all sky. Bold and italicized values are significantly different by the Mann-Whitney U Test.

Region	Spring	Summer	Fall	Winter
Northwest	***-0.47***	***-0.40***	***-0.68***	***-0.65***
OR/NorCal	***-0.46***	***-0.14***	***-0.52***	***-0.27***
CA Coast	-0.21	-0.07	-0.14	-0.05
Sierra NV	***-0.67***	-0.01	***-0.36***	***-0.21***
SoCal	-0.08	-0.04	-0.03	0.11
HellsCyn	***-0.42***	-0.18	***-0.45***	-0.31
Great Basin	-0.12	-0.13	***-0.22***	-0.07
N Rockies	***-0.82***	***-0.29***	***-0.91***	-0.05
CO Plateau	***-0.22***	-0.01	***-0.08***	-0.06
Mogollon Plateau	***-0.16***	-0.06	-0.01	-0.09
Southern AZ	-0.01	-0.15	0.02	0.00
N Great Plains	***-0.15***	-0.06	***-0.27***	***0.42***
Central Rockies	***-0.36***	-0.03	***-0.24***	-0.11
West TX	-0.02	-0.13	0.02	0.13
Central Great Plains	-0.14	-0.23	-0.05	***1.69***
Mid South	0.12	-0.40	-0.05	***0.58***
Boundary Waters	0.11	-0.05	0.02	0.19
OH River Valley	-0.78	***-0.72***	-0.17	0.67
Appalachia	***-0.42***	***-0.78***	***-0.30***	0.22
Southeast	-0.19	***-0.74***	***-0.34***	0.07
E Coast	-0.01	-0.36	-0.18	-0.05
Northeast	***-0.21***	***-0.12***	-0.11	-0.08