Supplementary Materials:

On Aerosol Liquid Water and Sulfate Associations: The Potential for Fine Particulate Matter Biases

Jonathon E. Babila [1], Annmarie G. Carlton [1,*], Christopher J. Hennigan [2] and Virendra P. Ghate [3]

[1] Department of Chemistry, University of California, Irvine, CA 92617, USA; jbabila@uci.edu
[2] Department of Chemical, Biochemical, and Environmental Engineering, University of Maryland, Baltimore County, 1000 Hilltop Circle, Baltimore, MD 21250, USA; hennigan@umbc.edu
[3] Environmental Science Division, Argonne National Laboratory, Argonne, IL 60439, USA; vghate@anl.gov
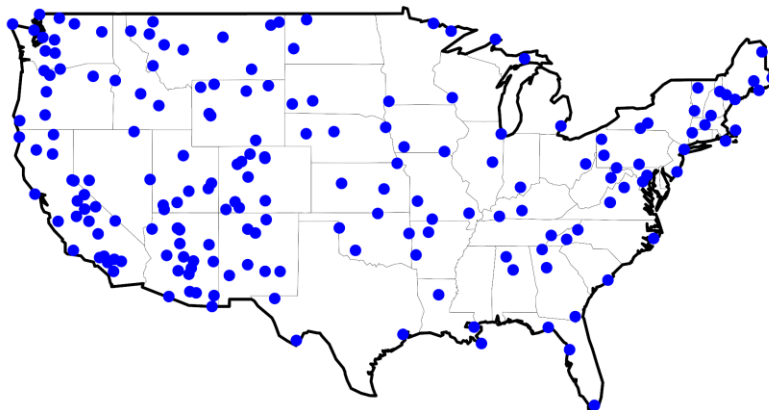***** Correspondence: agcarlto@uci.edu

Figure S1. IMPROVE network monitoring locations.

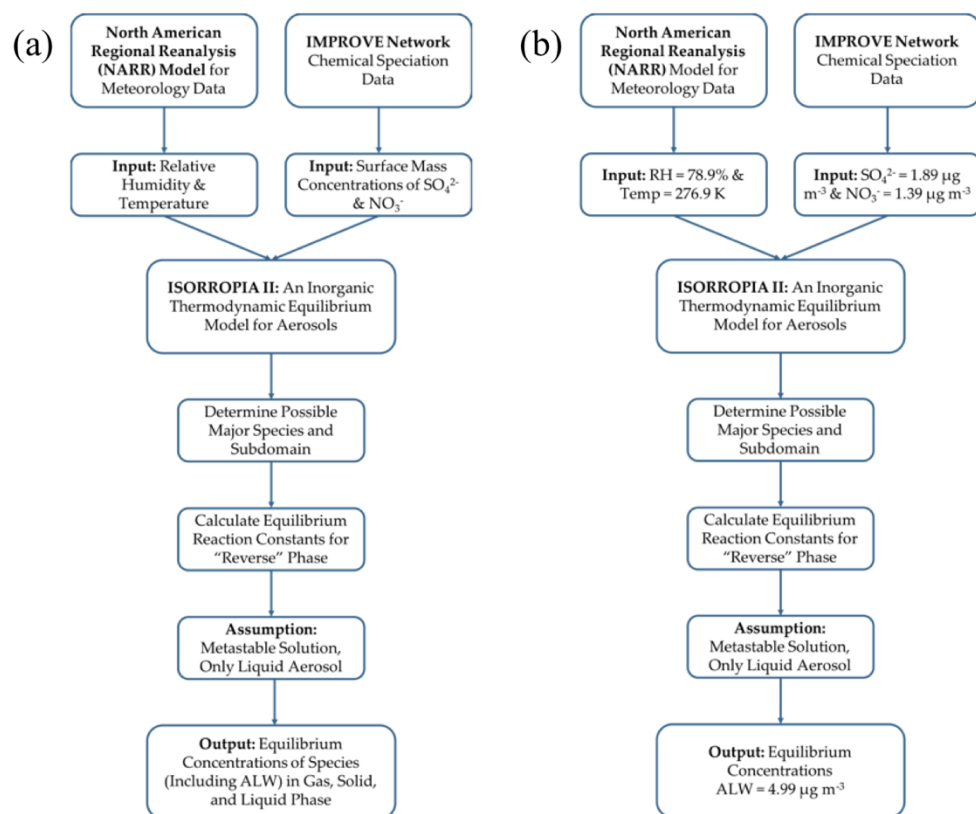**Figure S2.** (**a**) Flow chart summarizing the method for calculating aerosol liquid water from IMPROVE chemical speciation data. (**b**) Example inputs and output for the Washington D.C. IMPROVE site on 1/1/2004.
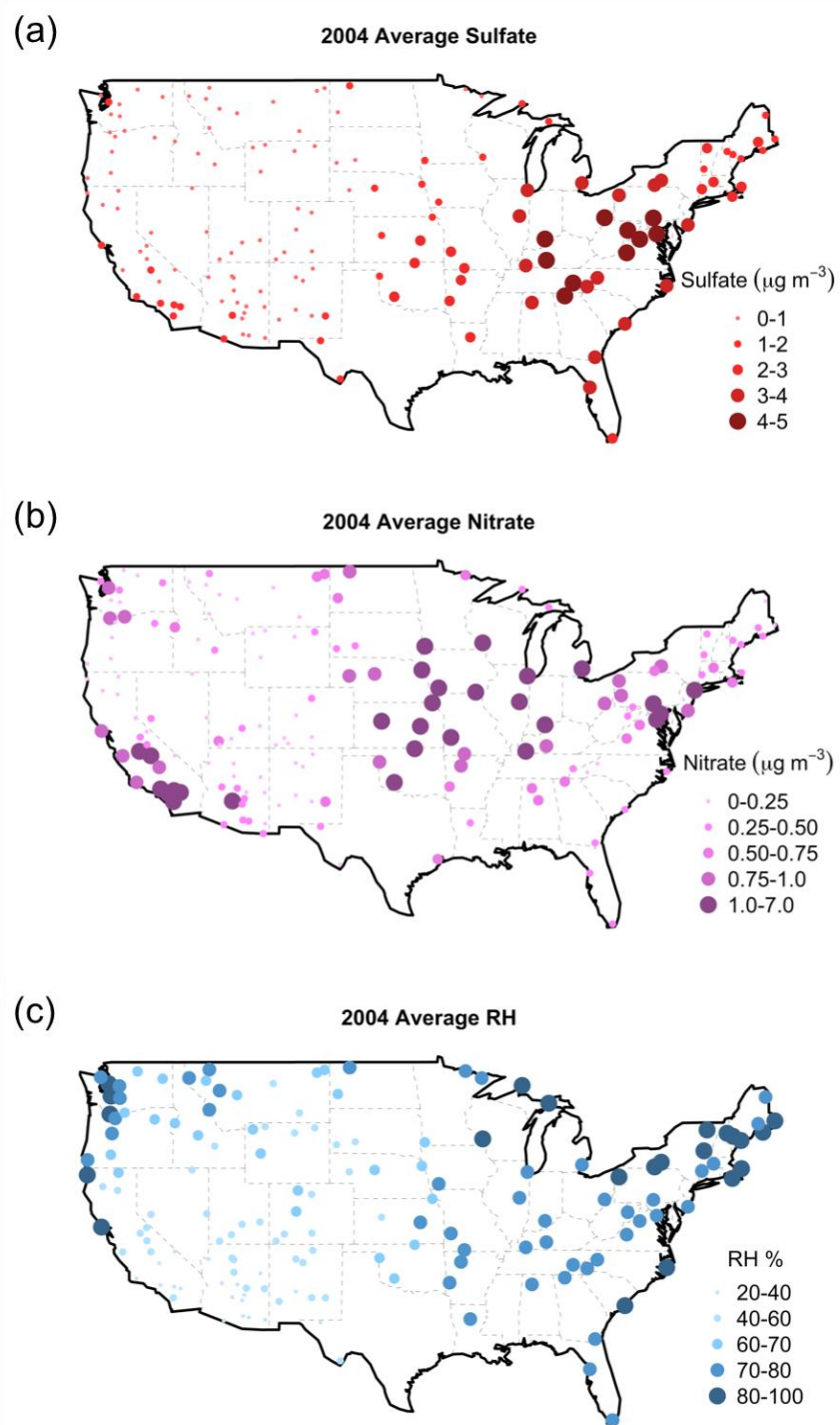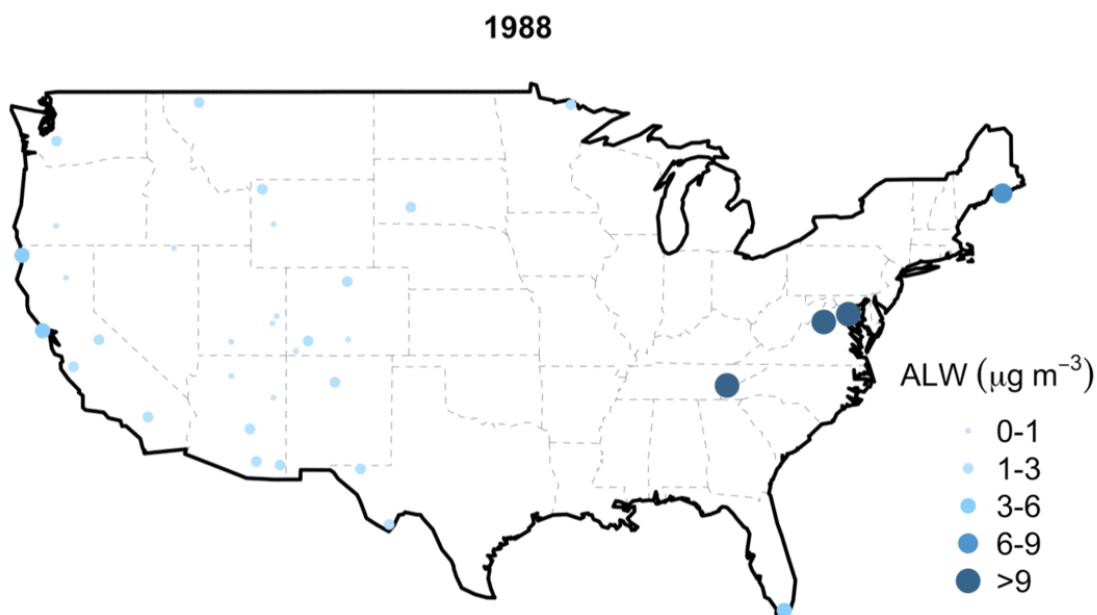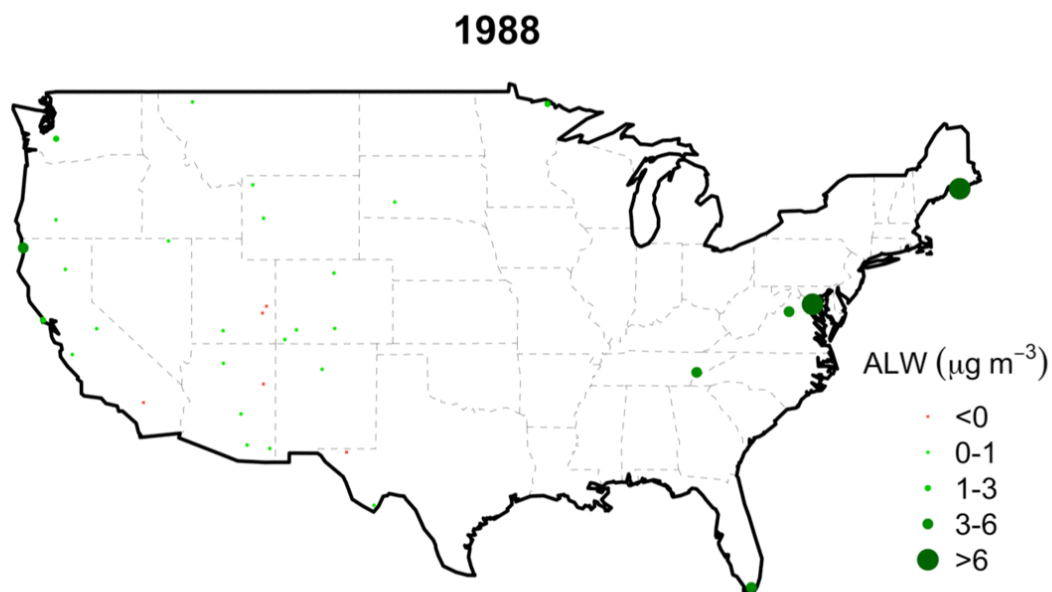
Figure S3. 2004 annual average (**a**) sulfate mass concentrations, (**b**) nitrate mass concentrations, and (**c**) RH % at IMPROVE monitoring sites.

Video S1. GIF of 1988 to 2013 yearly average ALW for IMPROVE monitoring sites. The color and diameter of the points are relative to the ALW mass according to the legend on the right.

Video S2. GIF of 1988 to 2013 yearly average difference between ambient and lab ALW for IMPROVE monitoring sites. The color and diameter of the points are relative to the ALW mass according to the legend on the right.

Figure S4. Eastern and Western U.S. as defined here.

Video S3. GIF of 2004 ALW monthly average for 158 IMPROVE monitoring sites. The color and diameter of the points are relative to the ALW mass according to the legend on the right.

January

ALW (μg m^{-3})

· <0
· 0-1
· 1-3
• 3-6
● >6

Video S4. GIF of 2004 monthly average difference between ambient and lab ALW for 158 IMPROVE monitoring sites. The color and diameter of the points are relative to the ALW mass according to the legend on the right.
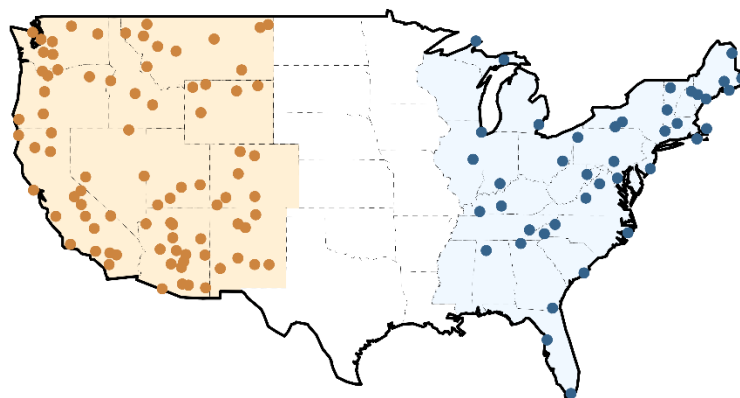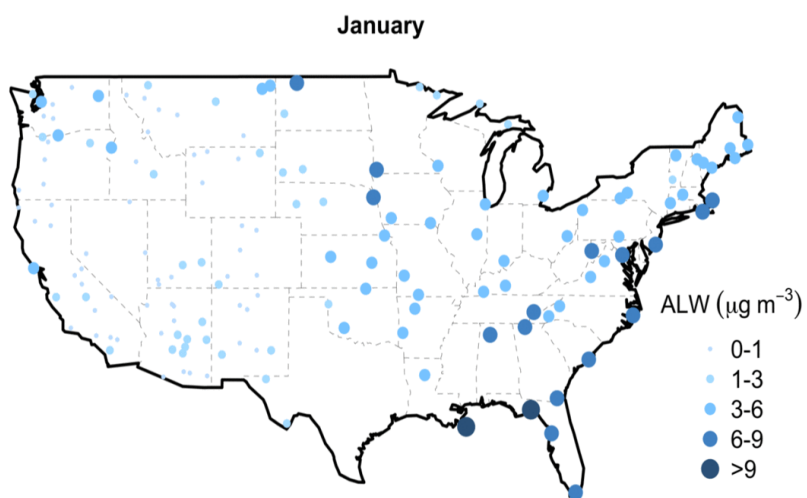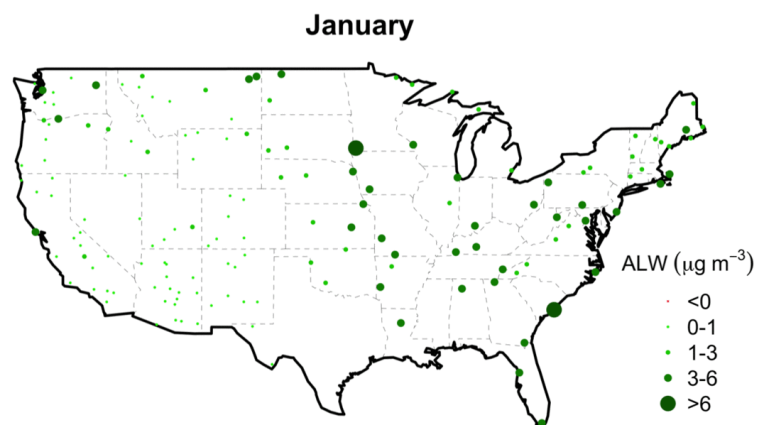
Table S1. Calculations of aerosol liquid water content at ambient and lab conditions for the 25 largest U.S. cities discussed in Reiss et al. Chemical speciation data is directly taken from Reiss et al., while water mass concentrations were calculated via ISORROPIA. "n/a" represents places where data was not available.

State and City	PM$_{2.5}$ (μg m^{-3})	Sulfate (μg m^{-3})	Nitrate (μg m^{-3})	Relative Humidity (%)	Temperature (°F)	Ambient Water (μg m^{-3})	Lab Water (μg m^{-3})
Atlanta, GA	16	4.8	0.8	79.6	62.7	13.13	4.79
Baltimore, MD	15.9	4.8	1.9	71.4	55.4	9.98	4.79
Boston, MA	11.2	3.1	1.3	72.9	50.8	6.80	3.09
Chicago, IL	17.5	3.3	2.6	65.4	49.8	5.93	3.29
Cincinnati, OH	14.7	4.4	2.1	71.7	54.0	9.39	4.39
Cleveland, OH	17.4	4.3	2.7	71.7	50.3	9.18	4.29
Dallas-Fort Worth, TX	12.1	3.2	1.2	62.3	67.2	5.38	3.19
Denver, CO	11.1	1.2	1.9	48.9	48.8	1.56	1.20
Detroit, MI	18.8	3.8	2.6	64.8	50.1	6.83	3.79
Houston, TX	11.7	3.5	0.9	72.2	70.9	7.47	3.49
Los Angeles, CA	19.3	3.6	5.8	60.9	63.3	5.92	3.59
Miami-Fort Laurderdale, FL	11.8	2.6	0.7	77.4	76.8	6.42	2.59
Minneapolis, MN	10.4	1.9	2.1	71.0	46.4	3.95	1.90
New York, NY	15.6	4.1	2.0	68.0	54.5	7.89	4.09
Philadelphia, PA	17.4	4.1	2.1	82.5	55.4	12.64	4.09
Phoenix, AZ	9.5	1.0	1.1	37.5	75.0	1.06	1.00
Pittsburgh, PA	14.9	5.0	1.1	71.0	51.6	10.39	4.99
Portland, OR	10.8	1.4	1.1	71.9	55.7	2.99	1.40
Riverside-San Bernardino, CA	23.8	2.9	8.9	51.8	63.3	3.98	2.89
San Antonio, TX	8.4	n/a	n/a	75.8	69.6	n/a	n/a
San Francisco, CA	14.1	1.4	2.7	80.2	59.2	3.83	1.40
Seattle, WA	12.5	1.8	1.1	77.4	53.4	4.45	1.80
St. Louis, MO-IL	14.5	3.6	2.2	65.9	57.5	6.62	3.59
Tampa, FL	13.3	3.5	0.7	75.5	73.1	8.38	3.49
Washington, DC	15.1	5.2	1.8	68.6	57.9	10.26	5.19

1
2

Table S2. 2004 annual average correlations of particulate sulfate and transition metals for a subset of cities.

| City	R² Sulfate and Al	R² Sulfate and Cr	R² Sulfate and Cu	R² Sulfate and Fe	R² Sulfate and Pb	R² Sulfate and Mn	R² Sulfate and Ni
Washington D.C.	6.0E-02	4.6E-01	2.9E-02	4.7E-02	6.6E-02	5.0E-01	2.5E-01
Tallgrass, KS	5.0E-03	1.7E-04	4.7E-02	6.9E-02	7.5E-02	6.9E-02	5.7E-02
Bondville, IL	3.7E-02	3.7E-03	1.1E-02	1.0E-01	1.0E-02	5.6E-02	2.0E-02
Martha's Vineyard, MA	9.3E-02	4.1E-02	2.4E-01	2.3E-01	2.5E-01	2.4E-01	4.0E-01
Quaker City, OH	2.2E-01	2.5E-04	3.1E-02	1.2E-01	4.3E-03	3.7E-02	6.9E-02
Phoenix, AZ	1.4E-01	1.8E-02	5.7E-03	1.1E-02	8.4E-03	4.7E-02	1.4E-01
Seattle, WA	1.4E-01	5.9E-02	1.6E-01	2.6E-01	2.7E-01	4.5E-02	1.7E-01

3

4
5
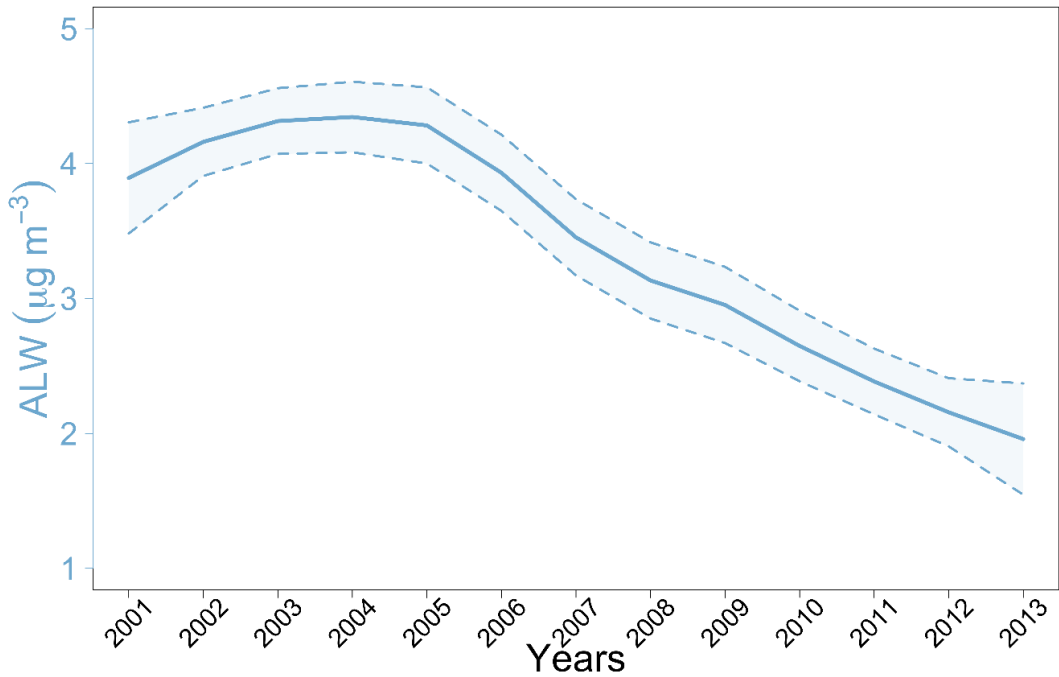
Figure S5. U.S. average ambient aerosol liquid water for 2001 to 2013.
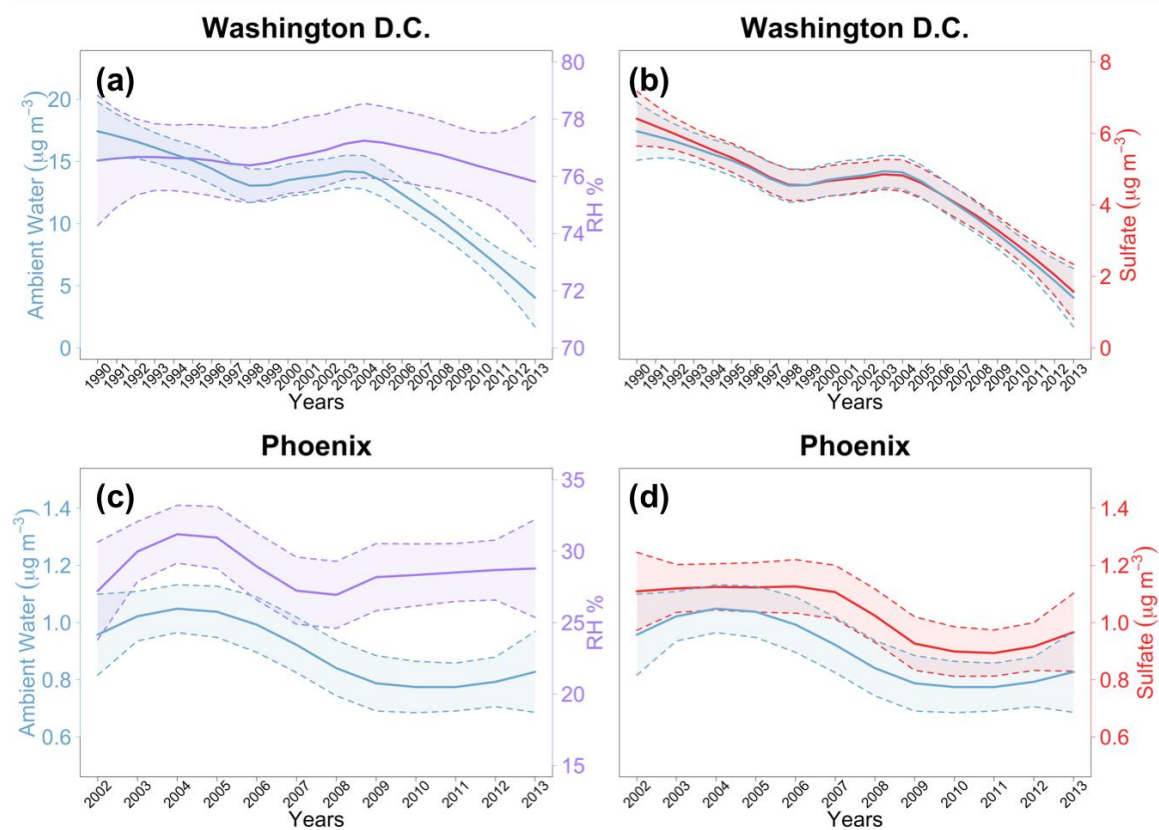
6

Figure S6. Yearly average comparing ambient water content to relative humidity (**Figures a** and **c**), and comparing ambient water content to sulfate mass (**Figures b** and **d**) for cities Washington D.C. (**a** and **b**), and Phoenix (**c** and **d**). Each parameter has a dashed line above and below it representing it's 95% confidence interval. Note different x and y-axis scales between Phoenix and DC.