Supplement of:

Long-term Regional Trends of Nitrogen and Sulfur Deposition in the United States from 2002 to 2017

Sarah E. Benish et al.
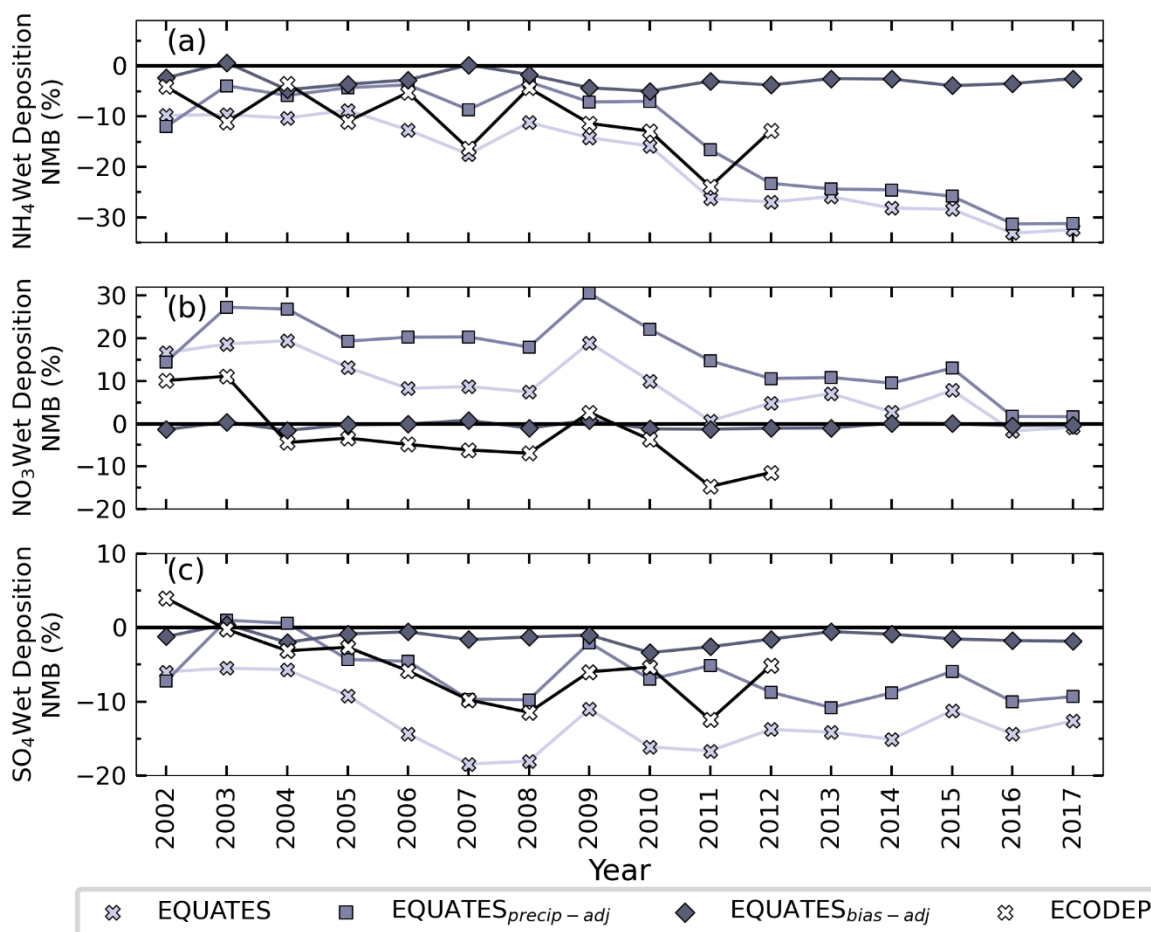
Figure S1. Annual normalized mean bias (NMB, %) for unadjusted, precipitation-adjusted, and bias-adjusted modeled NH₄ (panel a), NO₃ (panel b), and SO₄ (panel c) wet deposition compared to NTN measurements.
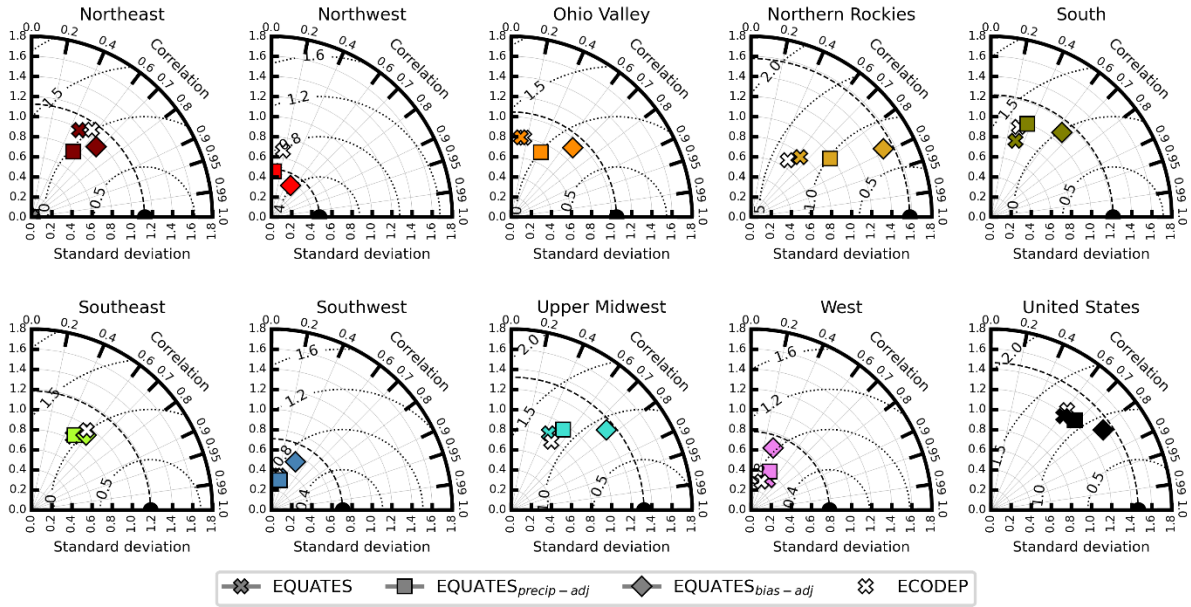
Figure S2. Taylor plot comparing 2002-2017 annual accumulated measured NH₄ wet deposition (kg/ha) with EQUATES and ECODEP (2002-2012) model output across climate regions within the CONUS. The symbols show how the unadjusted (x's), precipitation-adjusted (squares) and bias-adjusted (diamonds) modeled wet deposition compare to the NTN measurements (black circles).
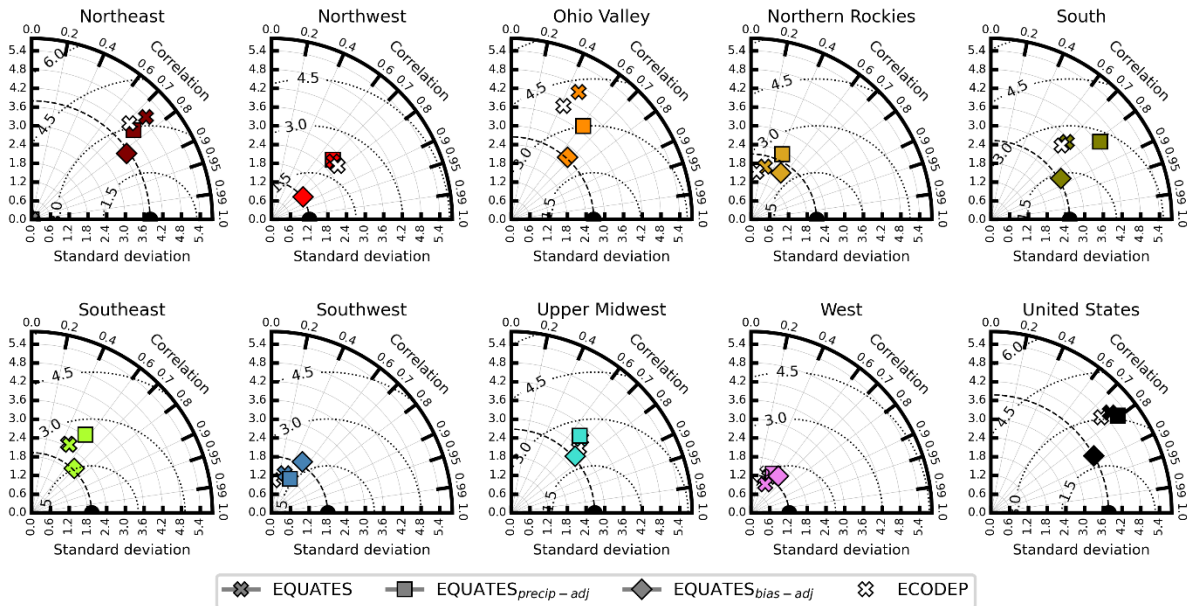
Figure S3. Taylor plot comparing 2002-2017 annual accumulated measured NO₃ wet deposition (kg/ha) with EQUATES and ECODEP (2002-2012) model output across climate regions within the CONUS. The symbols show how the unadjusted (x's), precipitation-adjusted (squares) and bias-adjusted (diamonds) modeled wet deposition compare to the NTN measurements (black circles).
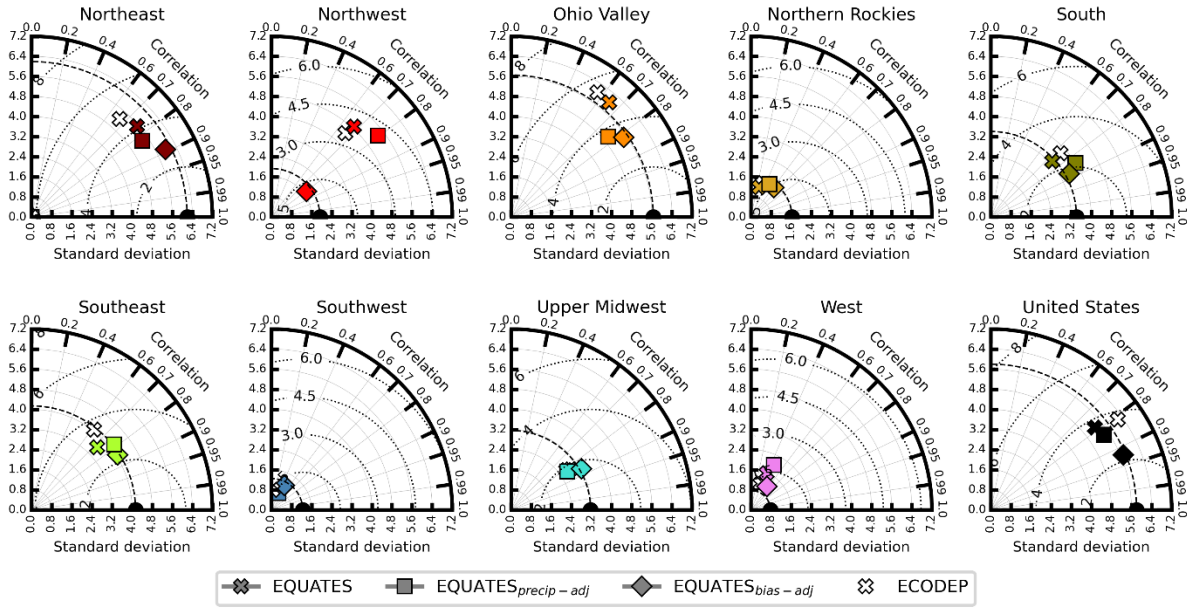
Figure S4. Taylor plot comparing 2002-2017 annual accumulated measured SO$_4$ wet deposition (kg/ha) with EQUATES and ECODEP (2002-2012) model output across climate regions within the CONUS. The symbols show how the unadjusted (x's), precipitation-adjusted (squares) and bias-adjusted (diamonds) modeled wet deposition compare to the NTN measurements (black circles).


Figure S5. Scatter plot comparing the annual accumulated wet deposition trend (kg/ha/yr) from 2002 to 2017 between NTN observations and EQUATES model output for NH$_4$ (a), NO$_3$ (b), and SO$_4$ (c). Each circle denotes a single NTN site, colored by the climate region that meet annual completeness criteria described in the text.

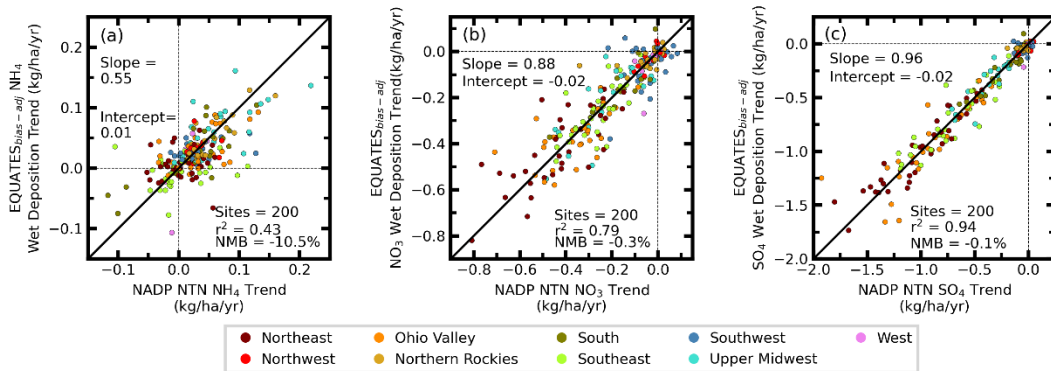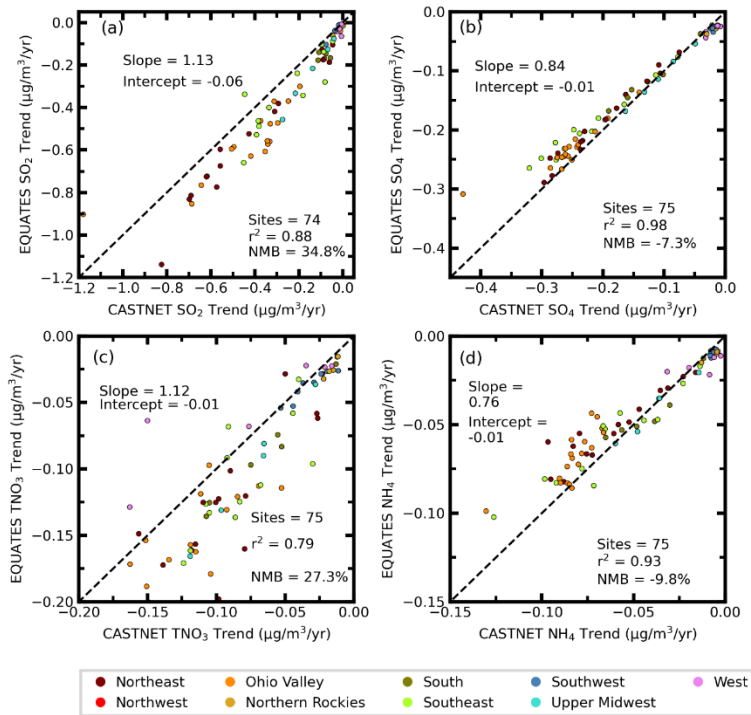**Figure S6. Scatter plot comparing the annual average concentration trend ($\mu g/m^3$/yr) from 2002 to 2017 between CASTNET observations and EQUATES model output of sulfur dioxide (a, SO_2), sulfate (b, SO_4), total oxidized nitrogen (c, TNO_3), and ammonium (d, NH_4). Each circle denotes a single CASTNET site colored by the climate that meet annual completeness criteria.**

Figure S7. Spatial distribution of dry (top) and wet (bottom) oxidized N deposition in 2002 (a and d, kg-N/ha), 2017 (b and e, kg-N/ha), and the 2002-2017 annual trend (c and f, kg-N/ha/yr) with significance at the 95% confidence interval. Grey areas in panels (c) and (f) indicate where the trend is unavailable or not significant (i.e., p-value of the Wald's test is greater than 0.05).
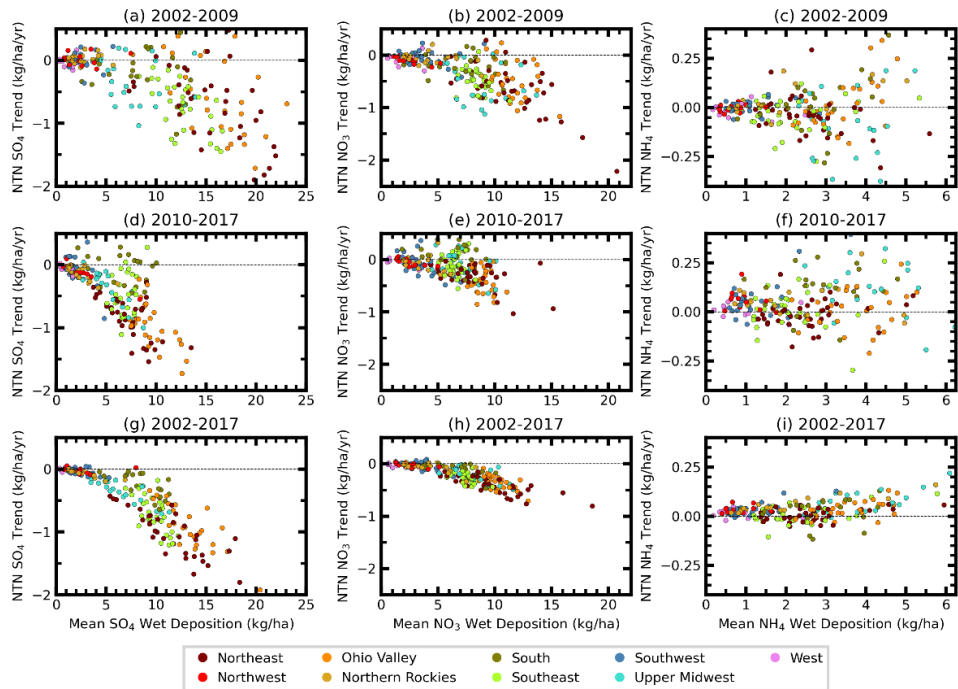
Figure S8. Measured wet deposition trends (kg/ha/yr) of SO₄ (left), NO₃ (middle), and NH₄ (right) as a function of average measured deposition amount from 2002-2009 (top), 2010-2017 (middle), and 2002-2017 (bottom). Each circle is a NTN site colored by the climate region.
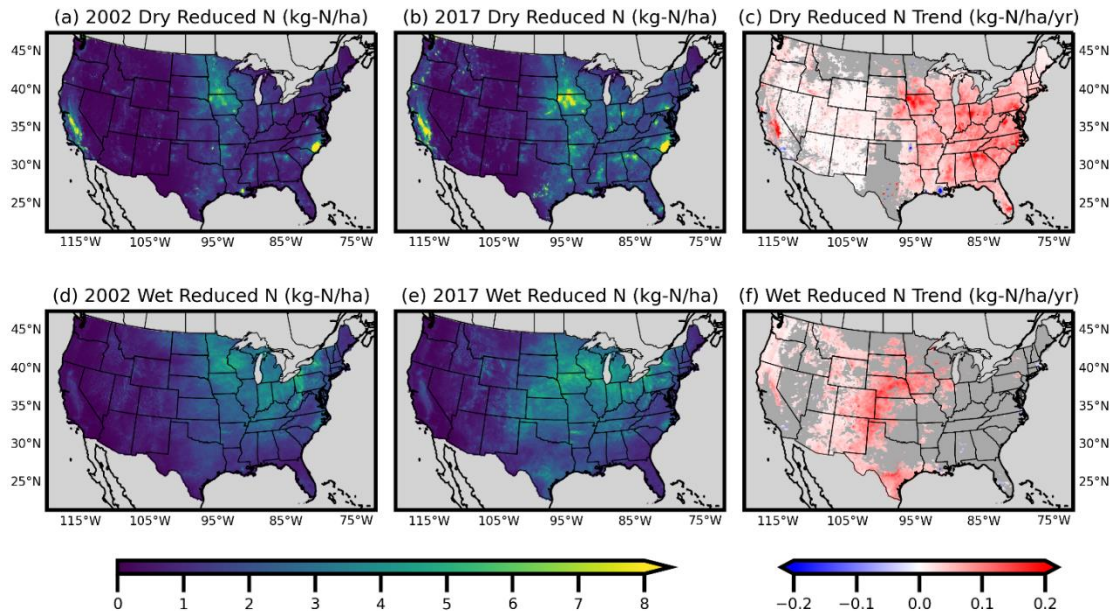
Figure S9. Spatial distribution of dry (top) and wet (bottom) reduced N deposition in 2002 (a and d, kg-N/ha), 2017 (b and e, kg-N/ha), and the 2002-2017 annual trend (c and f, kg-N/ha/yr) with significance at the 95% confidence interval. Grey areas in panels (c) and (f) indicate where the trend is unavailable or not significant (i.e., p-value of the Wald's test is greater than 0.05).
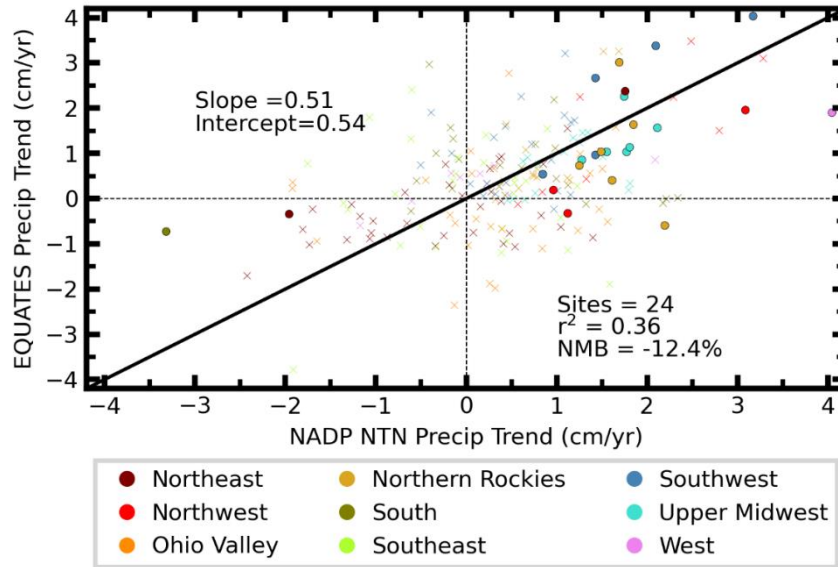
Figure S10. Scatter plot comparing the annual accumulated precipitation trend (cm/yr) from 2002 to 2017 between NTN observations and EQUATES model output. Each marker denotes a single NTN site colored by the climate region that meet annual completeness criteria. Circle markers indicate the observed trend is statistically significant with 95% confidence, while the x's denote unsignificant trends. Summary statistics printed on the figure are computed for sites with statistically significant trends.
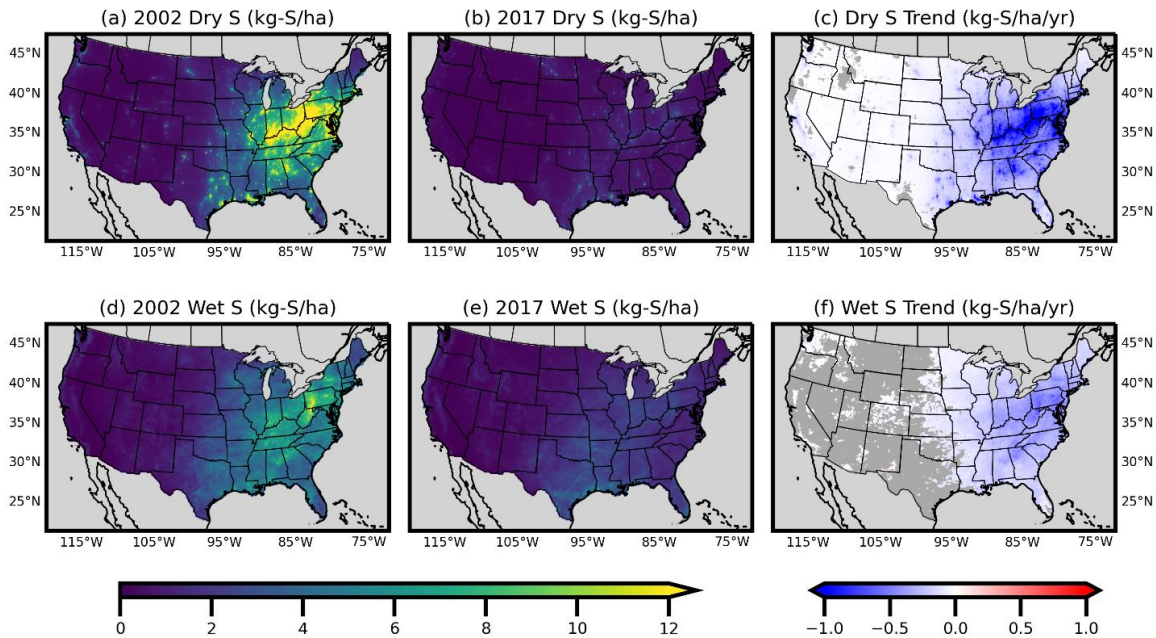
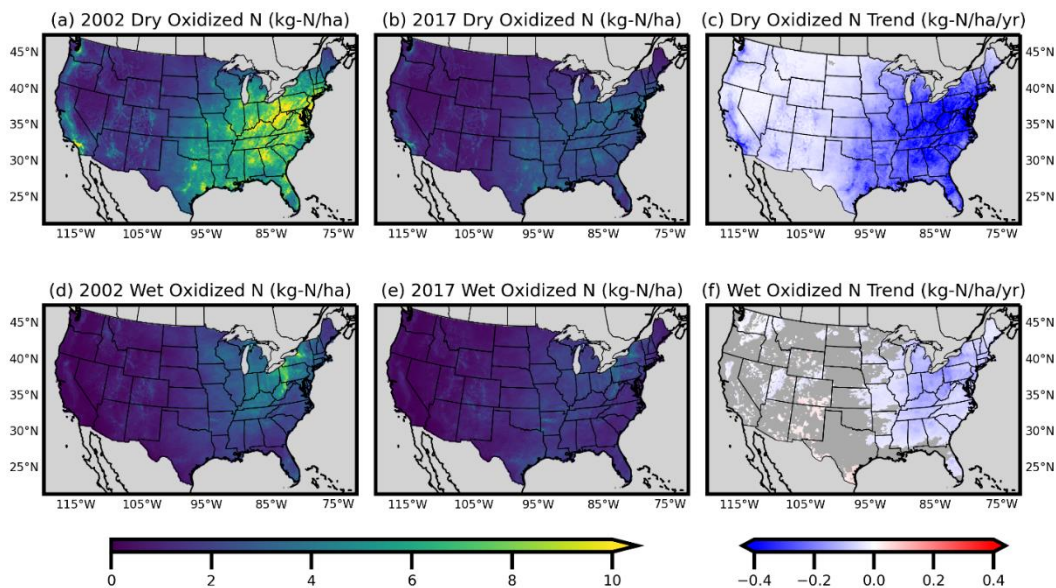**Figure S11. Spatial distribution of dry (top) and wet (bottom) S deposition in 2002 (a and d, kg-S/ha), 2017 (b and e, kg-S/ha), and the 2002-2017 annual trend (c and f, kg-S/ha/yr) with significance at the 95% confidence interval. Grey areas in panels (c) and (f) indicate where the trend is unavailable or not significant (i.e., p-value of the Wald's test is greater than 0.05).**
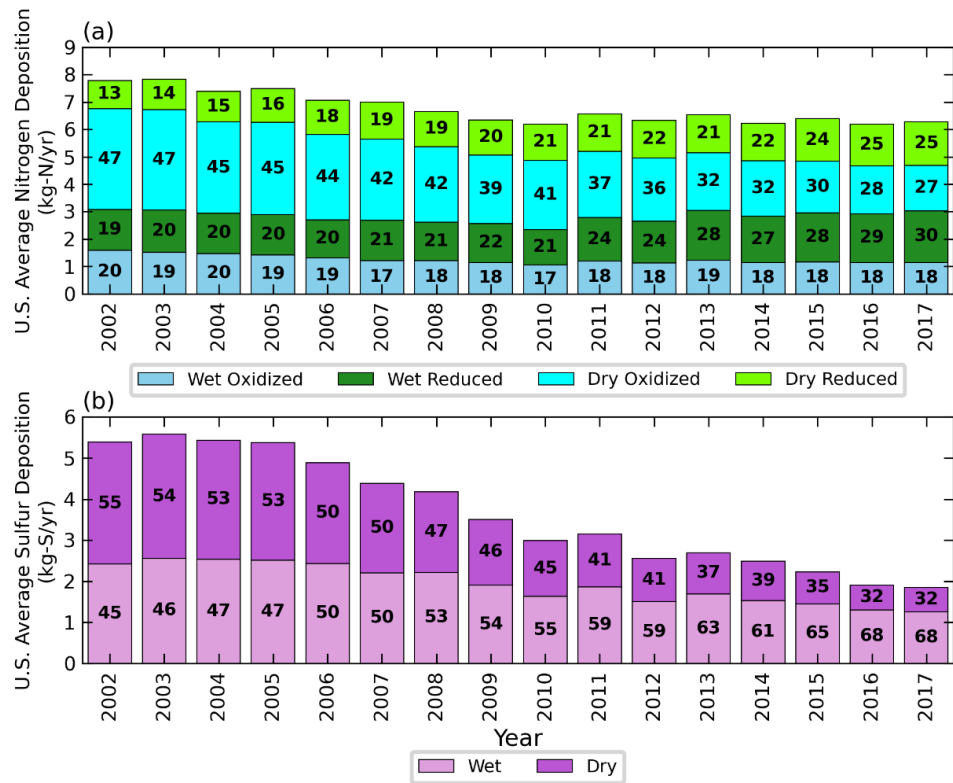
**Figure S12.** Annual average total (wet+dry) nitrogen (panel a, kg-N/ha) and sulfur (panel b, kg-S/ha) deposition from 2002 to 2017 across the CONUS. The bold numbers in each bar denote the percentage contribution to the annual total.

Table S1. List of NTN site locations assessed in this study with years of valid data and climate region designations.

| Site ID	Site Name	Latitude	Longitude	Valid Years	NOAA Region
AL10	Black Belt Research and Extension Center	32.4583	-87.2422	15	Southeast
AL99	Sand Mountain Research and Extension Center	34.2886	-85.9699	16	Southeast
AR02	Warren 2WSW	33.605	-92.0972	16	South
AR03	Caddo Valley	34.1795	-93.0992	15	South
AR16	Buffalo National River-Buffalo Point	36.0842	-92.5868	15	South
AR27	Fayetteville	36.1011	-94.1737	16	South
AZ03	Grand Canyon National Park-Hopi Point	36.0586	-112.184	16	Southwest
AZ06	Organ Pipe Cactus National Monument	31.9492	-112.802	16	Southwest
AZ97	Petrified Forest National Park-Rainbow Forest	34.8224	-109.893	14	Southwest
AZ98	Chiricahua	32.0097	-109.389	16	Southwest
AZ99	Oliver Knoll	33.0708	-109.866	14	Southwest
CA42	Tanbark Flat	34.2071	-117.762	15	West
CA45	Hopland	39.0045	-123.086	15	West
CA66	Pinnacles National Park-Bear Valley	36.4834	-121.157	16	West
CA67	Joshua Tree National Park-Black Rock	34.0695	-116.389	15	West
CA76	Montague	41.7662	-122.48	16	West
CA88	Davis	38.5357	-121.776	15	West
CA96	Lassen Volcanic National Park-Manzanita Lake	40.539	-121.577	13	West
CA99	Yosemite National Park-Hodgdon Meadow	37.7961	-119.858	14	West
CO00	Alamosa	37.4421	-105.868	16	Southwest
CO01	Las Animas Fish Hatchery	38.1177	-103.316	16	Southwest
CO08	Four Mile Park	39.4025	-107.345	16	Southwest
CO10	Gothic	38.9561	-106.986	16	Southwest
CO19	Rocky Meadows National Park-Beaver Meadows	40.3639	-105.581	16	Southwest
CO21	Manitou	39.1008	-105.093	16	Southwest
CO22	Pawnee	40.806	-104.756	14	Southwest
CO91	Wolf Creek Pass	37.4686	-106.787	15	Southwest
CO92	Sunlight Peak	39.4264	-107.38	15	Southwest
CO93	Buffalo Pass-Dry Lake	40.5347	-106.781	16	Southwest
CO94	Sugarloaf	39.9939	-105.48	16	Southwest
CO96	Molas Pass	37.75	-107.689	15	Southwest
CO97	Buffalo Pass-Summit Lake	40.5383	-106.677	16	Southwest

CO98	Rocky Mountain National Park-Loch Vale	40.2878	-105.663	13	Southwest
CO99	Mesa Verde National Park-Chapin Mesa	37.1979	-108.491	16	Southwest
CT15	Abington	41.84	-72.0101	16	Northeast
FL03	Bradford Forest	29.9748	-82.1978	14	Southeast
FL05	Chassahowitzka National Wildlife Refuge	28.7486	-82.5551	16	Southeast
FL11	Everglades National Park-Research Center	25.39	-80.68	16	Southeast
FL14	Quincy	30.5486	-84.6004	16	Southeast
FL23	Sumatra	30.1106	-84.9902	16	Southeast
FL41	Verna Well Field	27.3801	-82.2831	16	Southeast
FL99	Kennedy Space Center	28.5428	-80.644	13	Southeast
GA09	Okefenokee National Wildlife Refuge	30.7404	-82.1283	16	Southeast
GA20	Bellville	32.0849	-81.9367	15	Southeast
GA33	Sapelo Island	31.3961	-81.2811	13	Southeast
GA41	Georgia Station	33.1805	-84.4103	15	Southeast
GA99	Chula	31.5217	-83.5482	16	Southeast
IA08	Big Springs Fish Hatchery	42.9097	-91.47	15	Upper Midwest
IA23	McNay Research Center	40.9631	-93.3925	16	Upper Midwest
ID02	Priest River Experimental Forest	48.3518	-116.84	15	Northwest
ID03	Craters of the Moon National Monument	43.4605	-113.555	16	Northwest
ID11	Reynolds Creek	43.2049	-116.75	16	Northwest
IL11	Bondville	40.0528	-88.3719	16	Ohio Valley
IL18	Shabbona	41.8414	-88.8511	14	Ohio Valley
IL46	Alhambra	38.8689	-89.6219	16	Ohio Valley
IL63	Dixon Springs Agricultural Center	37.4356	-88.6719	15	Ohio Valley
IL78	Monmouth	40.9333	-90.7231	16	Ohio Valley
IN20	Roush Lake	40.8401	-85.4639	16	Ohio Valley
IN22	Southwest Purdue Agriculture Center	38.7408	-87.4855	15	Ohio Valley
IN34	Indiana Dunes National Lakeshore	41.6318	-87.0881	16	Ohio Valley
IN41	Agronomy Center for Research and Extension	40.4749	-86.9924	16	Ohio Valley
KS07	Farlington Fish Hatchery	37.6511	-94.8036	16	South
KS31	Konza Prairie	39.1022	-96.6092	16	South
KS32	Lake Scott State Park	38.6717	-100.916	16	South
KY03	Mackville	37.7047	-85.0489	16	Ohio Valley
KY10	Mammoth Cave National Park-Houchin Meadow	37.1317	-86.148	15	Ohio Valley
KY19	Cannons Lane	38.2288	-85.6545	13	Ohio Valley

KY22	Lilley Cornett Woods	37.0778	-82.9936	16	Ohio Valley
KY35	Clark State Fish Hatchery	38.1183	-83.5469	14	Ohio Valley
KY99	Mulberry Flatt	36.9029	-88.0121	16	Ohio Valley
LA30	Southeast Research Station	30.7819	-90.2021	16	South
MA01	North Atlantic Coastal Lab	41.9759	-70.0241	15	Northeast
MA08	Quabbin Reservoir	42.3925	-72.3444	16	Northeast
MD13	Wye	38.9131	-76.1525	16	Northeast
MD15	Smith Island	37.9925	-76.0345	13	Northeast
MD18	Assateague Island National Seashore-Woodcock	38.251	-75.1593	16	Northeast
MD99	Beltsville	39.028	-76.8171	13	Northeast
ME00	Caribou	46.8675	-68.0134	15	Northeast
ME02	Bridgton	44.1075	-70.7289	16	Northeast
ME04	Carrabassett Valley	45.0803	-70.2118	13	Northeast
ME09	Greenville Station	45.4891	-69.6647	15	Northeast
ME96	Casco Bay-Wolfe's Neck Farm	43.8325	-70.0645	16	Northeast
ME98	Acadia National Park-McFarland Hill	44.3772	-68.2608	16	Northeast
MI09	Douglas Lake	45.5608	-84.6783	16	Upper Midwest
MI26	Kellogg Biological Station	42.4103	-85.3928	16	Upper Midwest
MI48	Seney National Wildlife Refuge-Headquarters	46.2889	-85.9504	16	Upper Midwest
MI51	Unionville	43.6135	-83.3599	16	Upper Midwest
MI52	Ann Arbor	42.4164	-83.9019	16	Upper Midwest
MI53	Wellston	44.2242	-85.8186	16	Upper Midwest
MI99	Chassell	47.1046	-88.5516	16	Upper Midwest
MN01	Cedar Creek	45.4017	-93.2031	16	Upper Midwest
MN08	Hovland	47.8471	-89.965	16	Upper Midwest
MN16	Marcell Experimental Forest	47.5311	-93.4686	16	Upper Midwest
MN18	Fernberg	47.9464	-91.4961	15	Upper Midwest
MN23	Camp Ripley	46.2494	-94.4972	15	Upper Midwest
MN27	Lamberton	44.237	-95.3011	15	Upper Midwest
MN28	Grindstone Lake	46.1217	-93.0001	16	Upper Midwest
MN32	Voyageurs National Park-Sullivan Bay	48.4132	-92.8305	15	Upper Midwest
MN99	Wolf Ridge	47.3841	-91.2067	16	Upper Midwest
MO03	Ashland Wildlife Area	38.754	-92.1994	15	Ohio Valley
MO05	University Forest	36.9108	-90.3187	16	Ohio Valley
MS10	Clinton	32.3069	-90.3186	14	South

MS19	Newton	32.3269	-89.2086	15	South
MS30	Coffeeville	34.0025	-89.7993	13	South
MT00	Little Bighorn Battlefield National Monument	45.5701	-107.438	14	Northern Rockies
MT05	Glacier National Park-Fire Weather Station	48.5102	-113.997	16	Northern Rockies
MT07	Clancy	46.485	-112.065	14	Northern Rockies
MT98	Havre-Northern Agricultural Research Center	48.5007	-109.798	15	Northern Rockies
NC03	Lewiston	36.1325	-77.1708	16	Southeast
NC06	Beaufort	34.8846	-76.6207	15	Southeast
NC25	Coweeta	35.0605	-83.4305	16	Southeast
NC29	Hofmann Forest	34.825	-77.3228	15	Southeast
NC34	Piedmont Research Station	35.697	-80.6225	16	Southeast
NC35	Clinton Crops Research Station	35.0258	-78.2783	16	Southeast
NC36	Jordan Creek	34.9705	-79.5281	15	Southeast
NC41	Finley Farm	35.7288	-78.6802	16	Southeast
ND00	Theodore Roosevelt National Park-Painted Canyon	46.8951	-103.378	16	Northern Rockies
ND08	Icelandic State Park	48.782	-97.7546	13	Northern Rockies
ND11	Woodworth	47.1247	-99.2381	15	Northern Rockies
NE15	Mead	41.1528	-96.4912	16	Northern Rockies
NE99	North Platte Agricultural Experiment Station	41.0592	-100.746	14	Northern Rockies
NH02	Hubbard Brook	43.9433	-71.7029	16	Northeast
NJ00	Edwin B. Forsythe National Wildlife Refuge	39.4728	-74.4369	15	Northeast
NJ99	Washington Crossing	40.3154	-74.8536	16	Northeast
NM07	Bandelier National Monument	35.7788	-106.266	16	Southwest
NM08	Mayhill	32.9096	-105.471	14	Southwest
NV03	Smith Valley	38.7992	-119.257	16	West
NV05	Great Basin National Park-Lehman Caves	39.0054	-114.217	15	West
NY01	Alfred	42.2276	-77.8016	13	Northeast
NY08	Aurora Research Farm	42.7339	-76.6597	16	Northeast
NY10	Chautauqua	42.2994	-79.3964	15	Northeast
NY20	Huntington Wildlife	43.9731	-74.2231	16	Northeast
NY22	Akwesasne Mohawk-Fort Covington	44.9226	-74.4806	15	Northeast
NY52	Bennett Bridge	43.5282	-75.9492	13	Northeast
NY68	Biscuit Brook	41.9936	-74.5031	16	Northeast
NY96	Cedar Beach-Southold	41.0347	-72.3891	13	Northeast
NY98	Whiteface Mountain	44.3933	-73.8594	16	Northeast

NY99	West Point	41.3511	-74.0484	16	Northeast
OH09	Oxford	39.5309	-84.7238	16	Ohio Valley
OH17	Delaware	40.3555	-83.0661	16	Ohio Valley
OH49	Caldwell	39.7928	-81.5311	16	Ohio Valley
OH54	Deer Creek State Park	39.6359	-83.2606	16	Ohio Valley
OH71	Wooster	40.7813	-81.9197	16	Ohio Valley
OK00	Salt Northern Rockies National Wildlife Refuge	36.7863	-98.18	16	South
OK17	Kessler Farm Field Laboratory	34.98	-97.5214	15	South
OK29	Goodwell Research Station	36.5908	-101.618	16	South
OR10	H. J. Andrews Experimental Forest	44.2118	-122.256	16	Northwest
OR18	Starkey Experimental Forest	45.2247	-118.513	16	Northwest
OR97	Hyslop Farm	44.6347	-123.19	16	Northwest
PA00	Arendtsville	39.9231	-77.3078	16	Northeast
PA15	Penn State	40.7883	-77.9458	16	Northeast
PA18	Young Woman's Creek	41.4142	-77.6799	16	Northeast
PA29	Kane Experimental Forest	41.5978	-78.7675	16	Northeast
PA42	Leading Ridge	40.6575	-77.9397	16	Northeast
PA47	Millersville	39.9909	-76.3856	14	Northeast
PA72	Milford	41.3273	-74.8199	13	Northeast
SC05	Cape Romain National Wildlife Refuge	32.943	-79.6592	16	Southeast
SC06	Santee National Wildlife Refuge	33.5394	-80.435	15	Southeast
SD04	Wind Cave National Park-Elk Mountain	43.5577	-103.484	14	Northern Rockies
SD08	Cottonwood	43.9461	-101.855	15	Northern Rockies
SD99	Huron Well Field	44.355	-98.2917	16	Northern Rockies
TN04	Speedwell	36.4698	-83.8272	16	Ohio Valley
TN11	Great Smoky Mountains National Park-Elkmount	35.6645	-83.5903	16	Ohio Valley
TN14	Hatchie National Wildlife Refuge	35.4688	-89.1713	14	Ohio Valley
TX03	Beeville	28.4667	-97.7069	16	South
TX04	Big Bend National Park-K-Bar	29.3025	-103.178	15	South
TX10	Attwater Prairie Chicken National Wildlife Refuge	29.6614	-96.2594	14	South
TX16	Sonora	30.2613	-100.555	16	South
TX21	Longview	32.3786	-94.7117	16	South
TX22	Guadalupe Mountains National Park Frijole Ranger Station	31.9069	-104.805	15	South
TX56	L.B.J. National Grasslands	33.3917	-97.6397	16	South
UT01	Logan	41.6661	-111.891	15	Southwest

UT09	Canyonlands National Park-Island in the Sky	38.4584	-109.821	15	Southwest
UT98	Green River	39.0001	-110.174	16	Southwest
UT99	Bryce Canyon National Park-Repeater Hill	37.6186	-112.173	16	Southwest
VA00	Charlottesville	38.0402	-78.5427	16	Southeast
VA13	Hortons Station	37.3232	-80.4572	16	Southeast
VA24	Prince Edward	37.1652	-78.3073	16	Southeast
VA28	Shenandoah National Park-Big Meadows	38.5231	-78.4348	15	Southeast
VA99	Natural Bridge Station	37.6265	-79.5126	15	Southeast
VT01	Bennington	42.8761	-73.1633	16	Northeast
VT99	Underhill	44.5283	-72.8684	16	Northeast
WA14	Olympic National Park-Hoh Ranger Station	47.8597	-123.933	14	Northwest
WA19	North Cascades National Park-Marblemount Ranger Station	48.5403	-121.446	16	Northwest
WA21	La Grande	46.8353	-122.287	14	Northwest
WA24	Palouse Conservation Farm	46.7606	-117.185	16	Northwest
WA98	Columbia River Gorge	45.5694	-122.21	14	Northwest
WA99	Mount Rainier National Park-Tahoma Woods	46.7582	-122.124	15	Northwest
WI35	Perkinstown	45.2064	-90.5978	16	Upper Midwest
WI36	Trout Lake	46.0512	-89.6541	16	Upper Midwest
WI37	Spooner	45.8228	-91.8744	15	Upper Midwest
WV05	Cedar Creek State Park	38.8794	-80.8476	15	Ohio Valley
WV18	Parsons	39.0897	-79.6622	16	Ohio Valley
WY00	Snowy Range	41.3762	-106.26	16	Northern Rockies
WY02	Sinks Canyon	42.7336	-108.85	16	Northern Rockies
WY06	Pinedale	42.929	-109.788	13	Northern Rockies
WY08	Yellowstone National Park-Tower Falls	44.9166	-110.42	16	Northern Rockies
WY95	Brooklyn Lake	41.3647	-106.241	16	Northern Rockies
WY98	Gypsum Creek	43.2227	-109.992	13	Northern Rockies
WY99	Newcastle	43.873	-104.192	16	Northern Rockies

Table S2. Location of CASTNET sites assessed in this study with years of valid data and climate region designations.

Site ID	Latitude	Longitude	NOAA Region	Valid Years
ABT147	41.8405	-72.0104	Northeast	16
ACA416	44.3771	-68.2608	Northeast	16
ALC188	30.7016	-94.674	South	14
ALH157	38.869	-89.6228	Ohio Valley	16
ANA115	42.4166	-83.9022	Upper Midwest	16
ARE128	39.9232	-77.3079	Northeast	16
ASH135	46.6038	-68.4132	Northeast	16
BBE401	29.3027	-103.178	South	16
BEL116	39.0282	-76.8171	Northeast	15
BFT142	34.8847	-76.6207	Southeast	16
BVL130	40.052	-88.3725	Ohio Valley	16
BWR139	38.445	-76.1113	Northeast	16
CAD150	34.1793	-93.0988	South	16
CAN407	38.4583	-109.821	Southwest	16
CAT175	41.9423	-74.552	Northeast	16
CDR119	38.8795	-80.8477	Ohio Valley	16
CDZ171	36.7841	-87.8502	Ohio Valley	16
CHA467	32.0094	-109.389	Southwest	16
CHE185	35.7508	-94.6698	South	16
CKT136	37.9215	-83.0663	Ohio Valley	16
CND125	35.2633	-79.8375	Southeast	16
CNT169	41.3645	-106.24	Northern Rockies	16
COW137	35.0605	-83.4303	Southeast	16
CTH110	42.4009	-76.6535	Northeast	16
CVL151	34.0027	-89.7992	South	16
DCP114	39.6359	-83.2606	Ohio Valley	16
ESP127	36.0389	-85.733	Ohio Valley	16
EVE419	25.3912	-80.6808	Southeast	15
GAS153	33.1812	-84.4101	Southeast	16
GLR468	48.5103	-113.997	Northern Rockies	16
GRB411	39.0051	-114.216	West	15
GRC474	36.0586	-112.184	Southwest	16
GRS420	35.6335	-83.9416	Ohio Valley	15
GTH161	38.9563	-106.986	Southwest	16
HOX148	44.1809	-85.739	Upper Midwest	15
HWF187	43.973	-74.2233	Northeast	16
IRL141	27.8492	-80.4556	Southeast	15
JOT403	34.0696	-116.389	West	16
KEF112	41.5981	-78.7679	Northeast	15
KNZ184	39.1022	-96.6096	South	16

LAV410	40.54	-121.577	West	16
LRL117	39.9883	-79.2516	Northeast	16
MAC426	37.1318	-86.143	Ohio Valley	16
MCK131	37.7047	-85.0487	Ohio Valley	15
MCK231	37.7047	-85.0487	Ohio Valley	16
MEV405	37.1984	-108.491	Southwest	14
MKG113	41.4268	-80.1452	Northeast	16
OXF122	39.5311	-84.7235	Ohio Valley	16
PAR107	39.0904	-79.6617	Ohio Valley	16
PED108	37.1652	-78.3071	Southeast	13
PET427	34.8225	-109.893	Southwest	16
PIN414	36.4832	-121.157	West	16
PND165	42.929	-109.788	Northern Rockies	16
PNF126	36.1054	-82.045	Southeast	16
PRK134	45.2065	-90.5972	Upper Midwest	15
PSU106	40.7209	-77.9318	Northeast	16
QAK172	39.9427	-81.3379	Ohio Valley	16
ROM206	40.2781	-105.546	Southwest	16
ROM406	40.2781	-105.546	Southwest	16
SAL133	40.816	-85.6614	Ohio Valley	15
SEK430	36.4895	-118.829	West	13
SHN418	38.5231	-78.4347	Southeast	15
SND152	34.289	-85.9701	Southeast	15
SPD111	36.4698	-83.8265	Ohio Valley	15
STK138	42.2872	-89.9999	Ohio Valley	16
SUM156	30.1102	-84.9904	Southeast	16
THR422	46.8948	-103.378	Northern Rockies	15
UVL124	43.6136	-83.3599	Upper Midwest	16
VIN140	38.7408	-87.4849	Ohio Valley	16
VOY413	48.4125	-92.8292	Upper Midwest	16
VPI120	37.3232	-80.4572	Southeast	16
WSP144	40.3123	-74.8727	Northeast	16
WST109	43.9445	-71.7008	Northeast	15
YEL408	44.5654	-110.4	Northern Rockies	16
YOS404	37.7132	-119.706	West	16