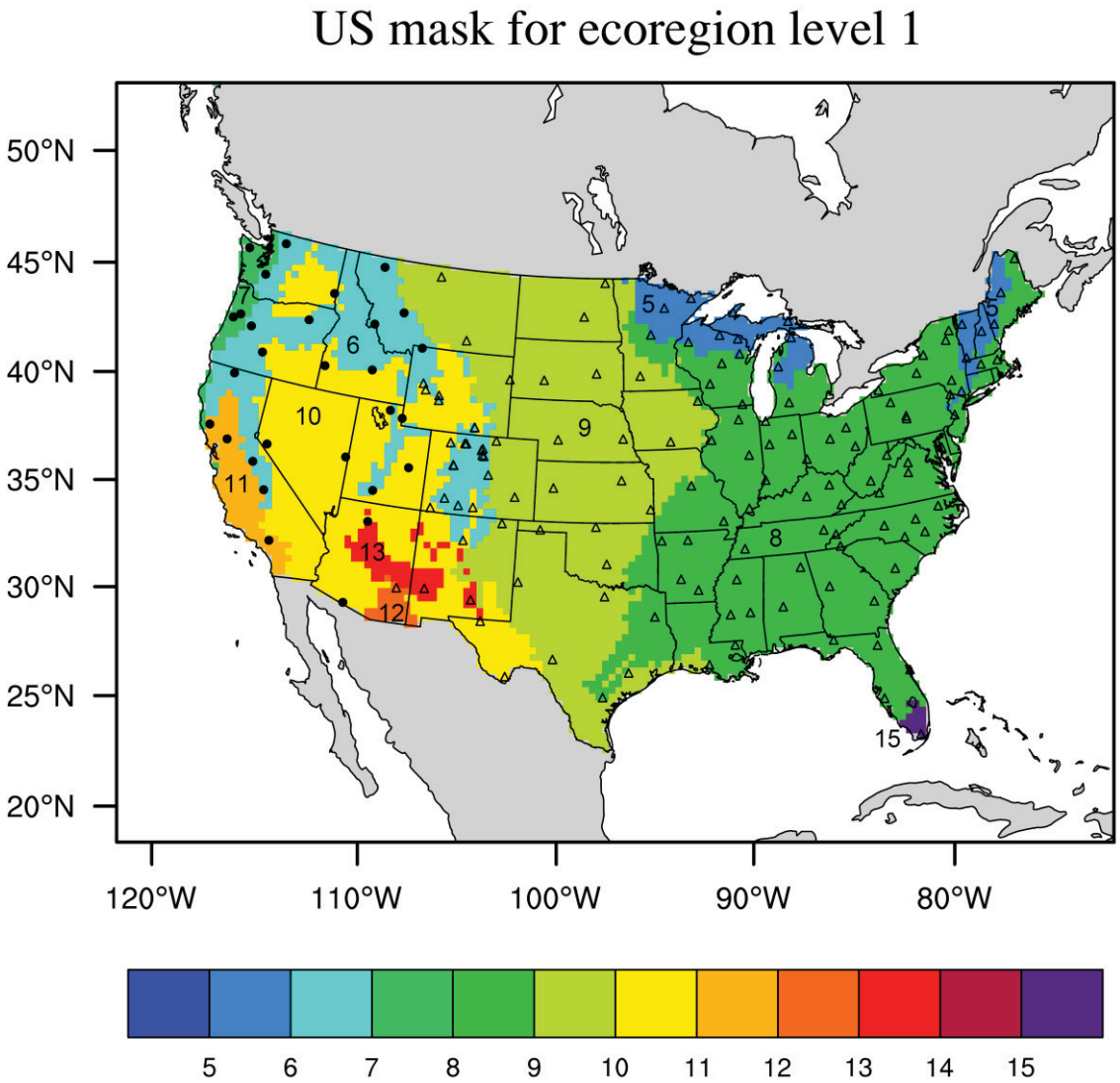

Figure S1. The 10 Level I ecoregions in the continental US: 5 = Northern Forests, 6 = Northwestern Forested Mountains, 7 = Marine West Coast Forest, 8 = Eastern Temperate Forests, 9 = Great Plains, 10 = North American Deserts, 11 = Mediterranean California, 12 =Southern Semi-arid Highlands, 13 = Temperate Sierras, and 15 = Tropical Wet Forests. The black circles are the NADP stations in the western US, and the up triangles are the NADP stations in the eastern US, as used in Figs. S2-S3, and Figs. 1-3 in the main manuscript. For detailed info of the NADP observations, please also see Table S2.

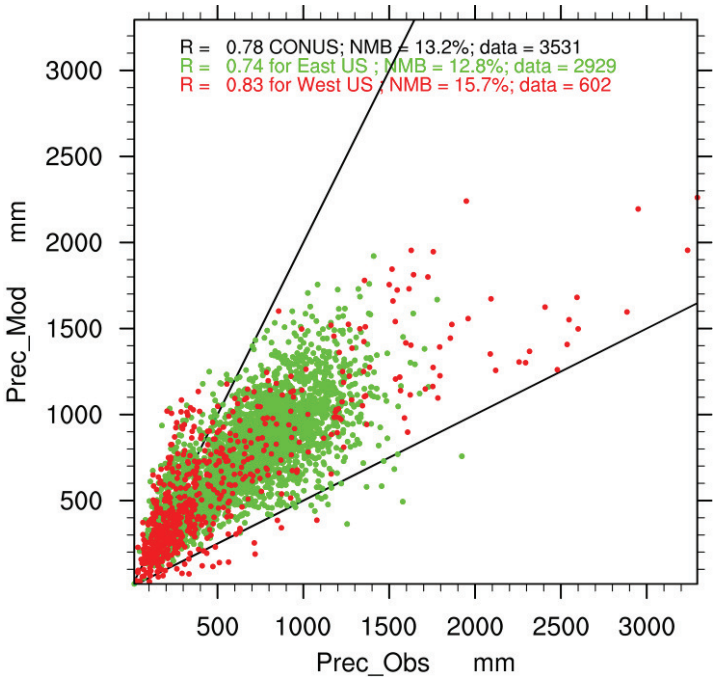

Figure S2. Scatter plots for the annual accumulated precipitation between observation (Prec_Obs) and model results (Prec_Mod) from 1990 to 2010 for 170 valid sites with 3531 valid data. The data at each NADP site is assumed valid only if at least 18 years of observation data are available with 75% annual coverage for that site. Each point in the plots represents the annual precipitation for a given site and year. Note that the annual accumulated precipitation may not be the actual annual totals because of missing data in the observations. The lines are for the 1:2 and 2:1 ratio.

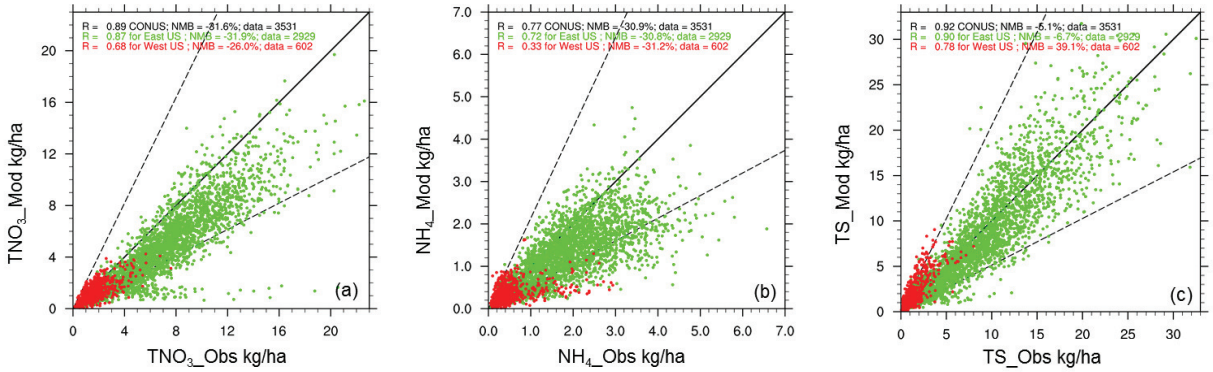

Figure S3. Scatter plots for the annual accumulated deposition (total oxidized nitrogen (TNO_3, a), reduced nitrogen (NH_X, b), and total sulfate (TS, c)) without considering the precipitation adjustment between observation (TNO_3_Obs, NH_4_Obs, TS_Obs) and model results (TNO_3_Mod, NH_4_Mod, TS_Mod) from 1990 to 2010 for 170 valid sites with 3531 valid data. The site in NADP is assumed valid if only at least 18 years of observation data is available with 75% annual coverage for the site. Note that the annual accumulated deposition may not be the actual annual totals because of the missing data in the observation. The green color is for the eastern US, and the red color is for the western US, with the dashed line for the 1:2 and 2:1 ratio, and the solid line for the 1:1 ratio.

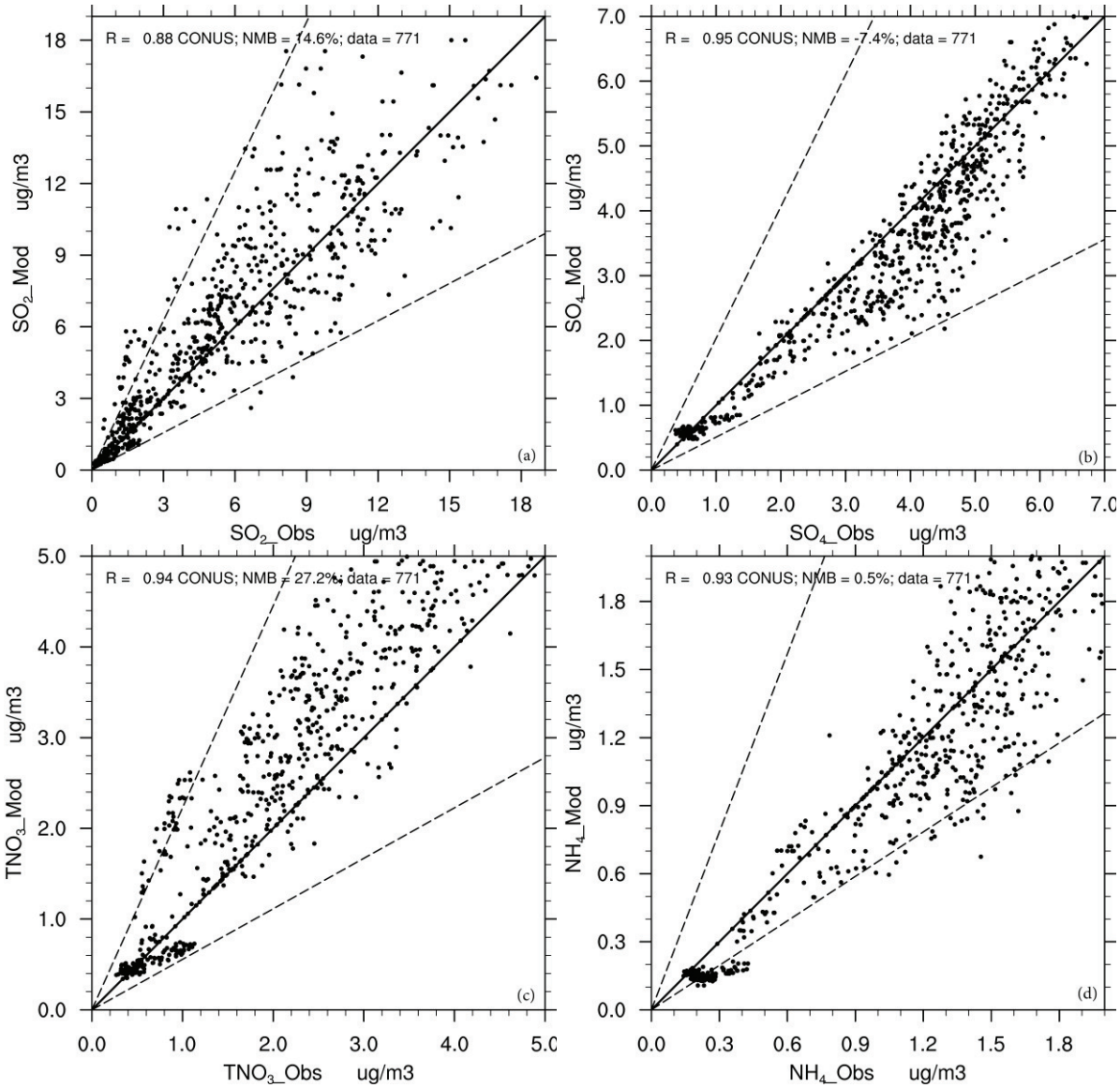

Figure S4. Scatter plots for the annual air concentration (sulfur dioxide (SO_2, a), particulate sulfate (SO_4, b), total particulate nitrate (TNO_3= NO_3 +HNO_3, c), and particulate ammonium (NH_4, d) between the observational and model results from 1990 to 2010 for 39 valid sites with 771 valid data. The site in CASTNET is assumed valid if only at least 18 years of observation data is available with 75% annual coverage for the site. The dashed lines are for the 1:2 and 2:1 ratio, and the solid line for the 1:1 ratio.

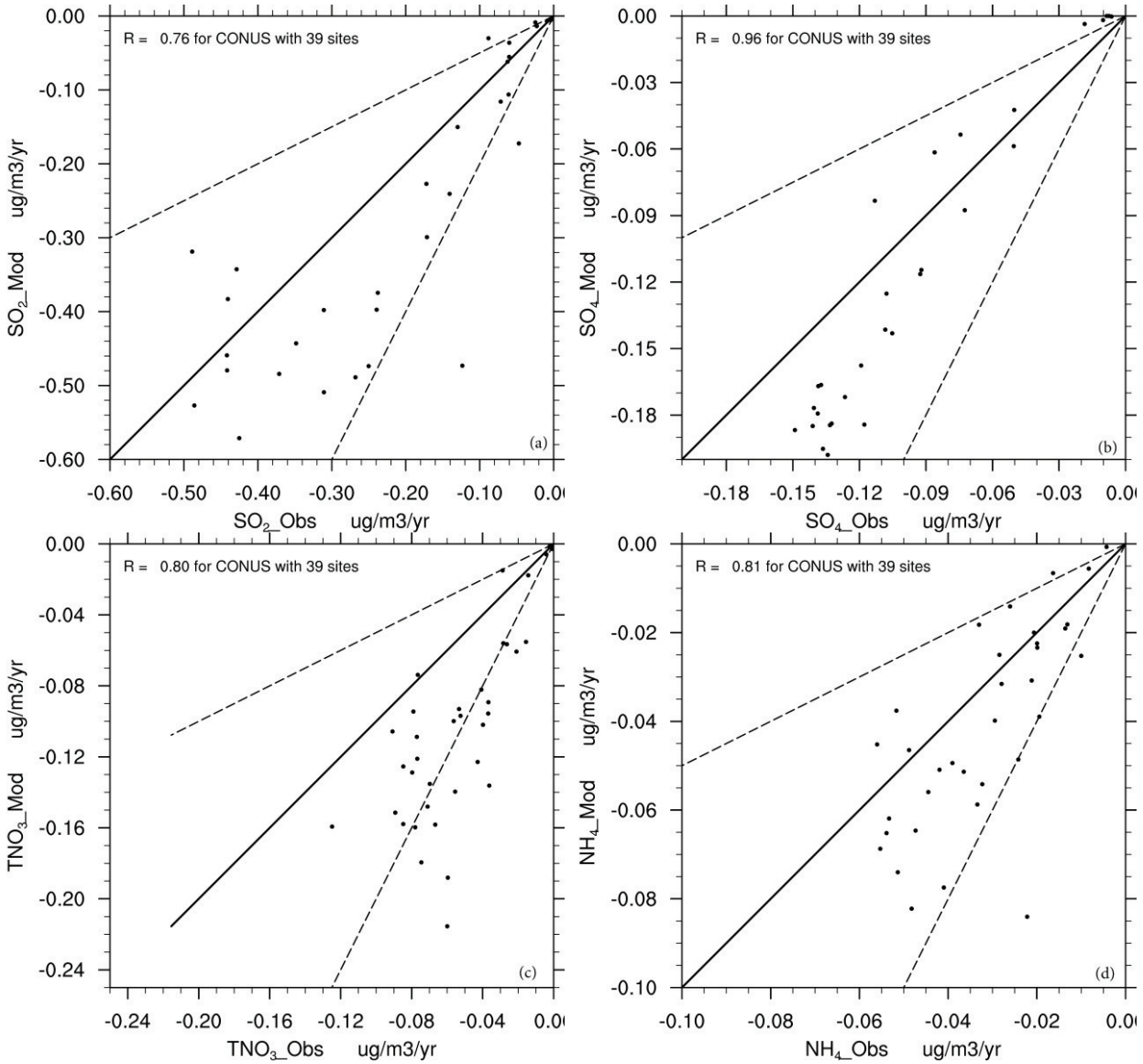

Figure S5. Comparison of the CASTNET trend for each valid site between the model values and observational for annual air concentration (sulfur dioxide (SO_2, a), particulate sulfate (SO_4, b), total particulate nitrate (TNO_3= NO_3 +HNO_3, c), and particulate ammonium (NH_4, d). Each CASTNET site is assumed to be valid for our analysis only if at least 18 years of observation data are available at that site and the data coverage is at least 75% for each year. The dashed line is for the 1:2 and 2:1 ratio, and the solid line for the 1:1 ratio.

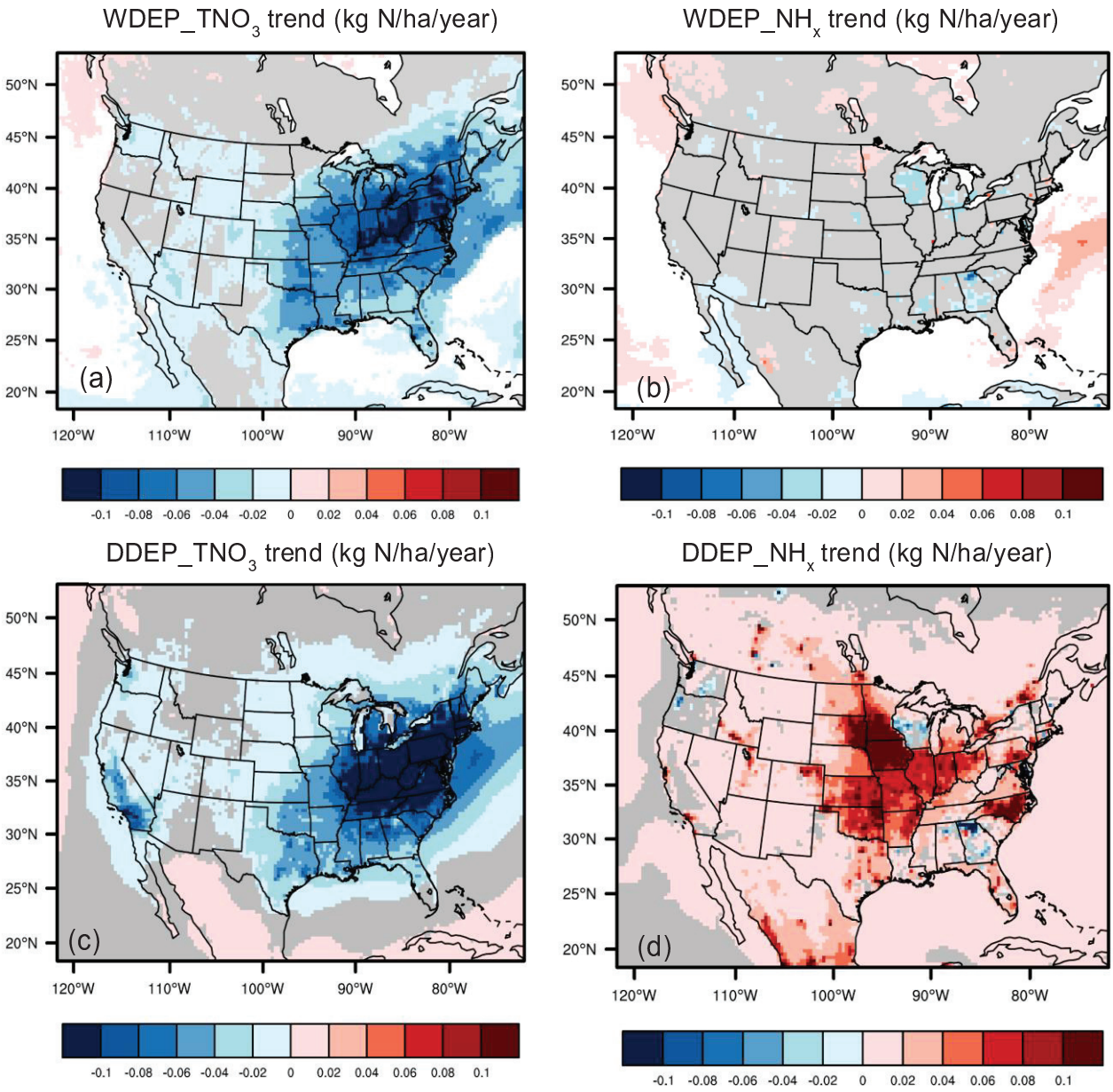

Figure S6. The spatial distribution of the wet deposition (a,b, WDEP) and dry deposition (c,d, DDEP) trends for oxidized nitrogen (TNO_3, a,c) and reduced nitrogen (NH_X, b,d).

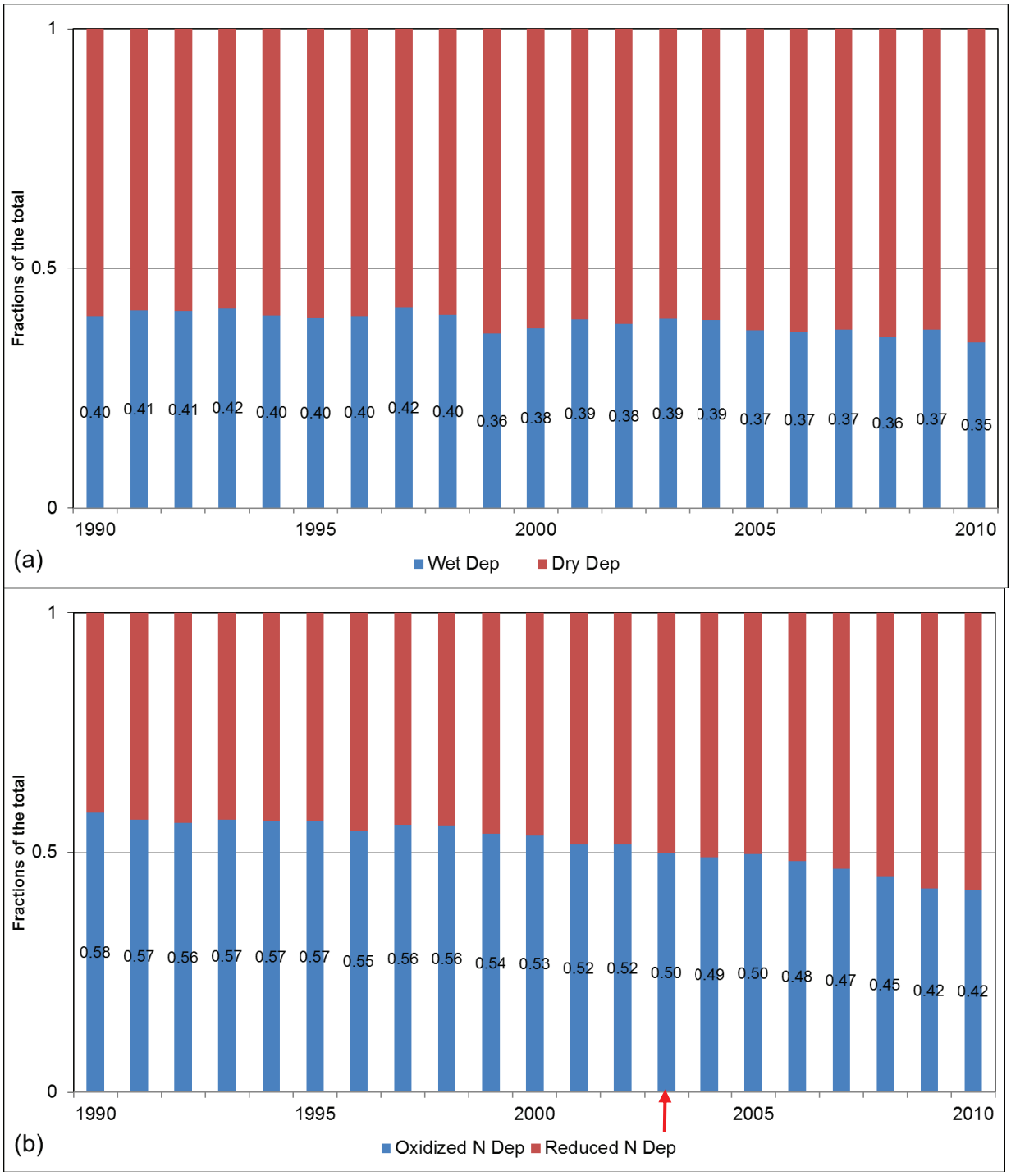

Figure S7. Interannual variability of the fractions of the total inorganic nitrogen deposition in the US from 1990 to 2010, including (a) wet versus dry, and (b) oxidized versus reduced. The red arrow points to the year 2003.

Table S1. List of configurations for the coupled WRF-CMAQ model from 1990 to 2010.

Parameter	Configuration
Emission	Xing et al. (2013)
Aerosol Chemistry	AERO6
Gas-phase Chemistry	Carbon Bond 05
Chemical Boundary Condition	Xing et al. (2015a)
Cumulus	Kain-Fritsch 2
Land Surface Model	Pleim-Xiu
Land Use Category	NLCD 50
Microphysics	Morrison 2-moment
Radiation	RRTMG SW & LW
Planetary Boundary Layer	ACM2 (Pleim, 2007)

Table S2. Location of the NADP observation networks with years of valid data, and the ecoregion definitions.

Site ID	Site Name	Valid Years	Ecoregion Level I
AL10	Black Belt Research & Extension Center	21	Eastern Temperate Forests
AL99	Sand Mountain Research & Extension Center	21	Eastern Temperate Forests
AR02	Warren 2WSW	21	Eastern Temperate Forests
AR03	Caddo Valley	21	Eastern Temperate Forests
AR16	Buffalo National River-Buffalo Point	21	Eastern Temperate Forests
AR27	Fayetteville	20	Eastern Temperate Forests
AZ03	Grand Canyon National Park-Hopi Point	21	North American Deserts
AZ06	Organ Pipe Cactus National Monument	20	North American Deserts
AZ99	Oliver Knoll	20	Southern Semi-arid Highlands
CA42	Tanbark Flat	21	Mediterranean California
CA45	Hopland	21	Mediterranean California
CA75	Sequoia National Park-Giant Forest	21	Mediterranean California
CA76	Montague	21	Northwestern Forested Mountains
CA88	Davis	21	Mediterranean California
CA99	Yosemite National Park-Hodgdon Meadow	21	Northwestern Forested Mountains
CO00	Alamosa	20	North American Deserts
CO01	Las Animas Fish Hatchery	21	Great Plains
CO02	Niwot Saddle	21	Northwestern Forested Mountains
CO08	Four Mile Park	21	Northwestern Forested Mountains
CO15	Sand Spring	21	North American Deserts
CO19	Rocky Mountain National Park-Beaver Meadows	21	Northwestern Forested Mountains
CO21	Manitou	21	Northwestern Forested Mountains
CO22	Pawnee	21	Great Plains
CO91	Wolf Creek Pass	18	Northwestern Forested Mountains
CO92	Sunlight Peak	18	Northwestern Forested Mountains
CO93	Buffalo Pass - Dry Lake	21	Northwestern Forested Mountains
CO94	Sugarloaf	21	Northwestern Forested Mountains
CO96	Molas Pass	21	Northwestern Forested Mountains
CO97	Buffalo Pass - Summit Lake	20	Northwestern Forested Mountains
CO98	Rocky Mountain National Park-Loch Vale	21	Northwestern Forested Mountains
CO99	Mesa Verde National Park-Chapin Mesa	21	North American Deserts
FL03	Bradford Forest	21	Eastern Temperate Forests
FL11	Everglades National Park-Research Center	21	Tropical Wet Forests
FL14	Quincy	21	Eastern Temperate Forests
FL41	Verna Well Field	21	Eastern Temperate Forests
FL99	Kennedy Space Center	21	Eastern Temperate Forests
GA20	Bellville	19	Eastern Temperate Forests
GA41	Georgia Station	21	Eastern Temperate Forests
IA08	Big Springs Fish Hatchery	20	Eastern Temperate Forests
IA23	McNay Research Center	21	Great Plains
ID03	Craters of the Moon National Monument	21	Northwestern Forested Mountains
ID11	Reynolds Creek	21	North American Deserts
IL11	Bondville	21	Eastern Temperate Forests

IL18	Shabbona	21	Eastern Temperate Forests
IL63	Dixon Springs Agricultural Center	21	Eastern Temperate Forests
IL78	Monmouth	21	Eastern Temperate Forests
IN20	Roush Lake	21	Eastern Temperate Forests
IN22	Southwest Purdue Agriculture Center	21	Eastern Temperate Forests
IN34	Indiana Dunes National Lakeshore	21	Eastern Temperate Forests
IN41	Agronomy Center for Research and Extension	21	Eastern Temperate Forests
KS07	Farlington Fish Hatchery	21	Great Plains
KS31	Konza Prairie	21	Great Plains
KS32	Lake Scott State Park	21	Great Plains
KY03	Mackville	21	Eastern Temperate Forests
KY22	Lilley Cornett Woods	21	Eastern Temperate Forests
KY35	Clark State Fish Hatchery	21	Eastern Temperate Forests
LA12	Iberia Research Station	20	Eastern Temperate Forests
LA30	Southeast Research Station	21	Eastern Temperate Forests
MA01	North Atlantic Coastal Lab	21	Eastern Temperate Forests
MA08	Quabbin Reservoir	21	Eastern Temperate Forests
MA13	East	20	Eastern Temperate Forests
MD13	Wye	21	Eastern Temperate Forests
ME00	Caribou	20	Eastern Temperate Forests
ME02	Bridgton	21	Northern Forests
ME09	Greenville Station	21	Northern Forests
ME98	Acadia National Park-McFarland Hill	21	Northern Forests
MI09	Douglas Lake	21	Northern Forests
MI26	Kellogg Biological Station	20	Eastern Temperate Forests
MI53	Wellston	21	Northern Forests
MI98	Raco	20	Northern Forests
MI99	Chassell	21	Northern Forests
MN16	Marcell Experimental Forest	21	Northern Forests
MN18	Fernberg	21	Northern Forests
MN23	Camp Ripley	21	Northern Forests
MN27	Lamberton	21	Great Plains
MO03	Ashland Wildlife Area	20	Eastern Temperate Forests
MO05	University Forest	21	Eastern Temperate Forests
MS10	Clinton	21	Eastern Temperate Forests
MS19	Newton	21	Eastern Temperate Forests
MS30	Coffeeville	21	Eastern Temperate Forests
MT00	Little Bighorn Battlefield National Monument	21	Great Plains
MT05	Glacier National Park-Fire Weather Station	21	Northwestern Forested Mountains
MT07	Clancy	21	Northwestern Forested Mountains
MT97	Lost Trail Pass	20	Northwestern Forested Mountains
MT98	Havre - Northern Agricultural Research Center	21	Great Plains
NC03	Lewiston	21	Eastern Temperate Forests
NC25	Coweeta	21	Eastern Temperate Forests
NC34	Piedmont Research Station	21	Eastern Temperate Forests
NC35	Clinton Crops Research Station	21	Eastern Temperate Forests
NC36	Jordan Creek	21	Eastern Temperate Forests
NC41	Finley Farm	21	Eastern Temperate Forests
ND08	Icelandic State Park	21	Great Plains

ND11	Woodworth	21	Great Plains
NE15	Mead	21	Great Plains
NE99	North Platte Agricultural Experiment Station	21	Great Plains
NH02	Hubbard Brook	21	Northern Forests
NJ99	Washington Crossing	21	Eastern Temperate Forests
NM01	Gila Cliff Dwellings National Monument	20	Temperate Sierras
NM07	Bandelier National Monument	21	Northwestern Forested Mountains
NM08	Mayhill	20	Temperate Sierras
NM12	Capulin Volcano National Monument	21	Great Plains
NV03	Smith Valley	21	North American Deserts
NV05	Great Basin National Park-Lehman Caves	21	North American Deserts
NY08	Aurora Research Farm	21	Eastern Temperate Forests
NY10	Chautauqua	20	Eastern Temperate Forests
NY20	Huntington Wildlife	21	Eastern Temperate Forests
NY52	Bennett Bridge	21	Eastern Temperate Forests
NY68	Biscuit Brook	21	Eastern Temperate Forests
NY98	Whiteface Mountain	21	Eastern Temperate Forests
NY99	West Point	21	Northern Forests
OH09	Oxford	21	Eastern Temperate Forests
OH17	Delaware	21	Eastern Temperate Forests
OH49	Caldwell	21	Eastern Temperate Forests
OH71	Wooster	21	Eastern Temperate Forests
OK00	Salt Plains National Wildlife Refuge	21	Great Plains
OK17	Kessler Farm Field Laboratory	19	Great Plains
OK29	Goodwell Research Station	20	Great Plains
OR02	Alsea Guard Ranger Station	18	Marine West Coast Forest
OR09	Silver Lake Ranger Station	20	Northwestern Forested Mountains
OR10	H. J. Andrews Experimental Forest	21	Northwestern Forested Mountains
OR18	Starkey Experimental Forest	21	Northwestern Forested Mountains
OR97	Hyslop Farm	21	Marine West Coast Forest
PA15	Penn State	21	Eastern Temperate Forests
PA29	Kane Experimental Forest	21	Eastern Temperate Forests
PA42	Leading Ridge	21	Eastern Temperate Forests
PA72	Milford	21	Eastern Temperate Forests
SC06	Santee National Wildlife Refuge	21	Eastern Temperate Forests
SD08	Cottonwood	21	Great Plains
SD99	Huron Well Field	21	Great Plains
TN00	Walker Branch Watershed	21	Eastern Temperate Forests
TN11	Great Smoky Mountains National Park-Elkmont	21	Eastern Temperate Forests
TN14	Hatchie National Wildlife Refuge	21	Eastern Temperate Forests
TX02	Muleshoe National Wildlife Refuge	19	Great Plains
TX03	Beeville	21	Eastern Temperate Forests
TX04	Big Bend National Park - K-Bar	21	North American Deserts
TX10	Attwater Prairie Chicken National Wildlife Refuge	21	Great Plains
TX16	Sonora	21	Great Plains
TX21	Longview	21	Eastern Temperate Forests
TX22	Guadalupe Mountains National Park Frijole Ranger Station	21	North American Deserts

TX56	L.B.J. National Grasslands	21	Great Plains
UT01	Logan	21	North American Deserts
UT08	Murphy Ridge	21	North American Deserts
UT98	Green River	21	North American Deserts
UT99	Bryce Canyon National Park-Repeater Hill	21	North American Deserts
VA00	Charlottesville	21	Eastern Temperate Forests
VA13	Hortons Station	21	Eastern Temperate Forests
VA28	Shenandoah National Park-Big Meadows	21	Eastern Temperate Forests
VT01	Bennington	21	Northern Forests
VT99	Underhill	21	Northern Forests
WA14	Olympic National Park-Hoh Ranger Station	20	Marine West Coast Forest
WA19	North Cascades National Park-Marblemount Ranger Station	21	Northwestern Forested Mountains
WA21	La Grande	21	Northwestern Forested Mountains
WA24	Palouse Conservation Farm	21	North American Deserts
WI09	Popple River	20	Northern Forests
WI25	Suring	21	Northern Forests
WI28	Lake Dubay	21	Eastern Temperate Forests
WI36	Trout Lake	21	Northern Forests
WI37	Spooner	21	Northern Forests
WI98	Wildcat Mountain	21	Eastern Temperate Forests
WI99	Lake Geneva	21	Eastern Temperate Forests
WV04	Babcock State Park	21	Eastern Temperate Forests
WV18	Parsons	21	Eastern Temperate Forests
WY00	Snowy Range	21	Northwestern Forested Mountains
WY02	Sinks Canyon	21	North American Deserts
WY06	Pinedale	21	Northwestern Forested Mountains
WY08	Yellowstone National Park-Tower Falls	21	Northwestern Forested Mountains
WY95	Brooklyn Lake	18	Northwestern Forested Mountains
WY97	South Pass City	20	Northwestern Forested Mountains
WY98	Gypsum Creek	21	North American Deserts
WY99	Newcastle	21	Great Plains

Table S3. Location of the CASTNET observation networks with years of valid data, the longitudes and latitudes of these stations.

Site ID	Valid Years	Longitude	Latitude
ALH157	20	-89.6228	38.869
ANA115	19	-83.9022	42.4166
ARE128	21	-77.3079	39.9232
ASH135	19	-68.4132	46.6038
BEL116	18	-76.8171	39.0282
BVL130	19	-88.3725	40.052
CAD150	18	-93.0988	34.1793
CDR119	20	-80.8477	38.8795
CHA467	21	-109.389	32.0094
CND125	19	-79.8375	35.2633
CNT169	21	-106.24	41.3645
COW137	21	-83.4303	35.0605
CTH110	20	-76.6535	42.4009
CVL151	18	-89.7992	34.0027
DCP114	19	-83.2606	39.6359
ESP127	19	-85.733	36.0389
GAS153	20	-84.4101	33.1812
GLR468	21	-113.997	48.5103
GRC474	21	-112.184	36.0586
GTH161	21	-106.986	38.9563
KEF112	18	-78.7679	41.5981
LRL117	19	-79.2516	39.9883
LYK123	19	-82.9982	40.917
MKG113	19	-80.1452	41.4268
OXF122	21	-84.7235	39.5311
PAR107	21	-79.6617	39.0904
PND165	20	-109.788	42.929
PNF126	19	-82.045	36.1054
PRK134	21	-90.5972	45.2065
PSU106	21	-77.9318	40.7209
SAL133	19	-85.6614	40.816
SHN418	20	-78.4347	38.5231
SPD111	18	-83.8265	36.4698
SUM156	21	-84.9904	30.1102
UVL124	19	-83.3599	43.6136
VIN140	21	-87.4849	38.7408
VPI120	20	-80.5575	37.3298
WSP144	20	-74.8727	40.3123
WST109	20	-71.7008	43.9445

Table S4. The normalized mean bias (NMB, %) between the model and the NADP sites from 2002 to 2006 from Appel et al. (2011) and this study including both precipitation adjustment and without precipitation adjustment.

		TNO_3	NH_X	TS
East	Appel et al., 2011	-21.9	-15.2	0.8
	No adjustment	-37.0	-33.4	-8.7
	Annual Precip-adjust	-39.6	-36.5	-12.0
West	Appel et al., 2011	-7.6	-15.7	18.9
	No adjustment	-23.0	-30.3	48.1
	Annual Precip-adjust	-22.3	-31.7	58.5

Table S5. The trends for the wet deposition (WDEP, units of kg N ha^{-1} yr^{-1} for nitrogen deposition including TNO_3, NH_X and TIN, and kg S ha^{-1} yr^{-1} for TS) over the ten ecoregions. The bolded values mean the trends are statistically significant with the P value less than 0.05 for the Student's t-test.

ID	Region Name	TNO_3	NH_X	TIN	TS
5	Northern Forests	**-0.036**	-0.002	**-0.037**	**-0.088**
6	Northwestern Forested Mountains	**-0.006**	0.002	-0.004	**-0.008**
7	Marine West Coast Forest	-0.002	0.007	0.005	0.005
8	Eastern Temperate Forests	**-0.065**	-0.005	**-0.070**	**-0.18**
9	Great Plains	**-0.022**	-0.001	**-0.023**	**-0.037**
10	North American Deserts	**-0.007**	0.00	**-0.007**	**-0.013**
11	Mediterranean California	-0.006	0.00	-0.006	0.002
12	Southern Semi-arid Highlands	**-0.014**	-0.007	**-0.021**	**-0.049**
13	Temperate Sierras	**-0.011**	-0.001	**-0.012**	**-0.034**
15	Tropical Wet Forests	**-0.023**	**0.011**	-0.012	**-0.036**

Table S6. The same as Table S5 but for dry deposition (DDEP, units of kg N ha^{-1} yr^{-1} for nitrogen deposition including TNO_3, NH_X and TIN, and kg S ha^{-1} yr^{-1} for TS) over the ten ecoregions.

ID	Region Name	TNO_3	NH_X	TIN	TS
5	Northern Forests	**-0.051**	**0.018**	**-0.033**	**-0.15**
6	Northwestern Forested Mountains	**-0.007**	**0.009**	0.002	**-0.013**
7	Marine West Coast Forest	**-0.016**	-0.005	**-0.022**	**-0.058**
8	Eastern Temperate Forests	**-0.088**	**0.039**	**-0.049**	**-0.33**
9	Great Plains	**-0.019**	**0.045**	**0.026**	**-0.045**
10	North American Deserts	**-0.009**	**0.009**	-0.001	**-0.011**
11	Mediterranean California	**-0.045**	0.012	**-0.032**	**-0.016**
12	Southern Semi-arid Highlands	0.001	**0.009**	**0.009**	**-0.025**
13	Temperate Sierras	-0.005	**0.010**	0.005	**-0.02**
15	Tropical Wet Forests	-0.004	**0.030**	**0.027**	**-0.019**