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Supplement of

Source regions contributing to excess reactive nitrogen deposition in the Greater Yellowstone Area (GYA) of the United States

Rui Zhang et al.

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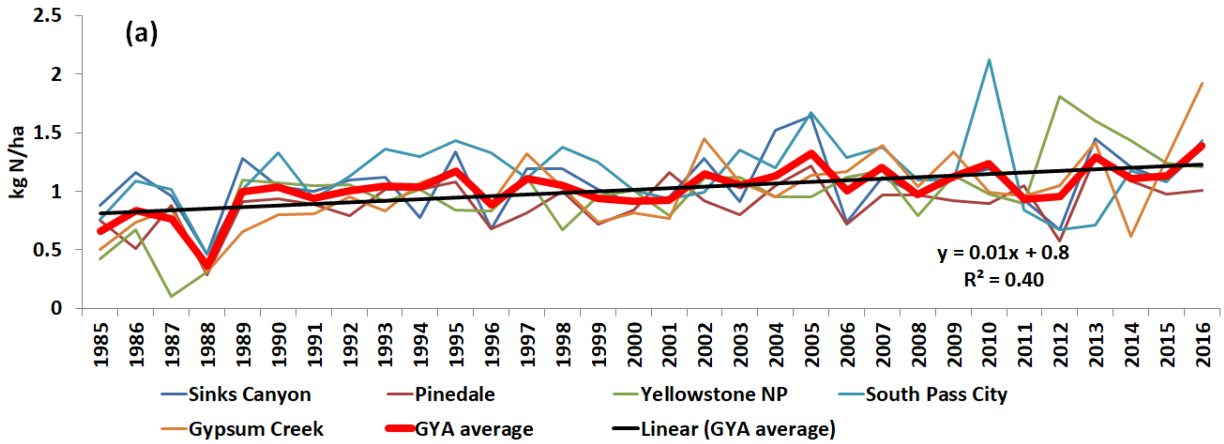


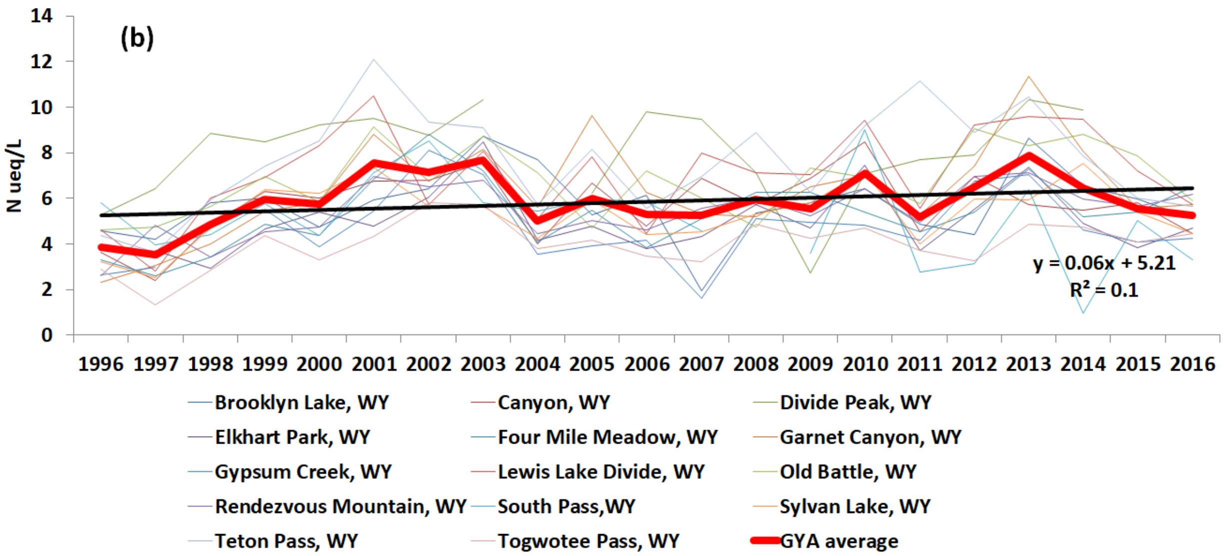


Figure S1. (a) Annual trend of measured wet N_r deposition measured at National Trend Network (NTN) sites (the Grand Teton site (WY94) was excluded from the trend analysis since began operation in 2011), (b) mean total N concentration (NH_4+NO_3) in snowpack measured at Rocky Mountain Regional snowpack Chemistry (RMRSC) network sites within the Greater Yellowstone Area (GYA).

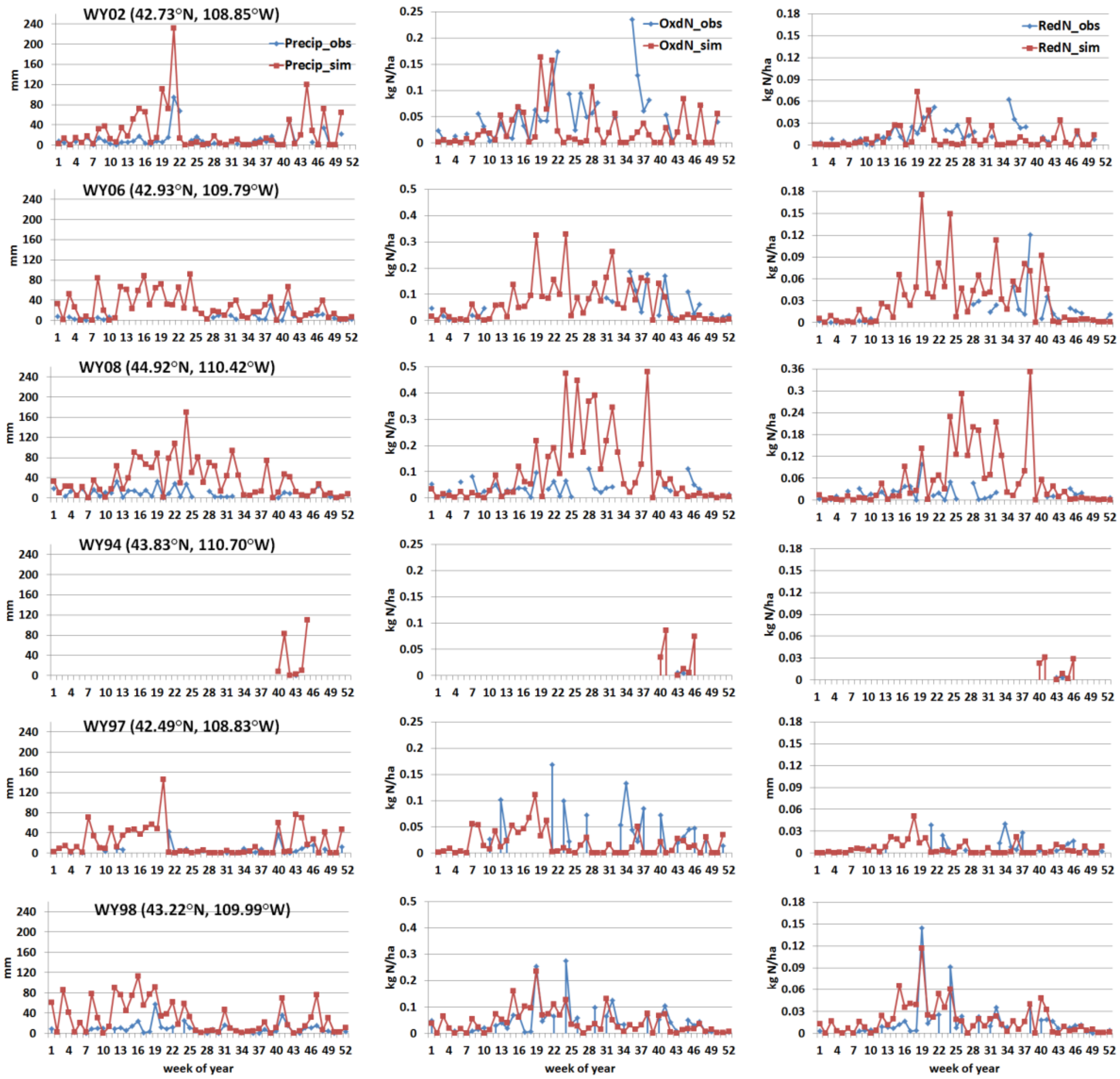


Figure S2. Time series comparison of measured (red lines) precipitation (left panels), wet deposition for oxidized nitrogen (middle panels) as well as wet deposition for reduced nitrogen with corresponding model values (blue lines) on the 6 NTN sites over Greater Yellowstone Area (GYA) in 2011. The Grand Teton site (WY94) began operation in September 2011. Due to the incomplete time series, these data were excluded from the model evaluation.

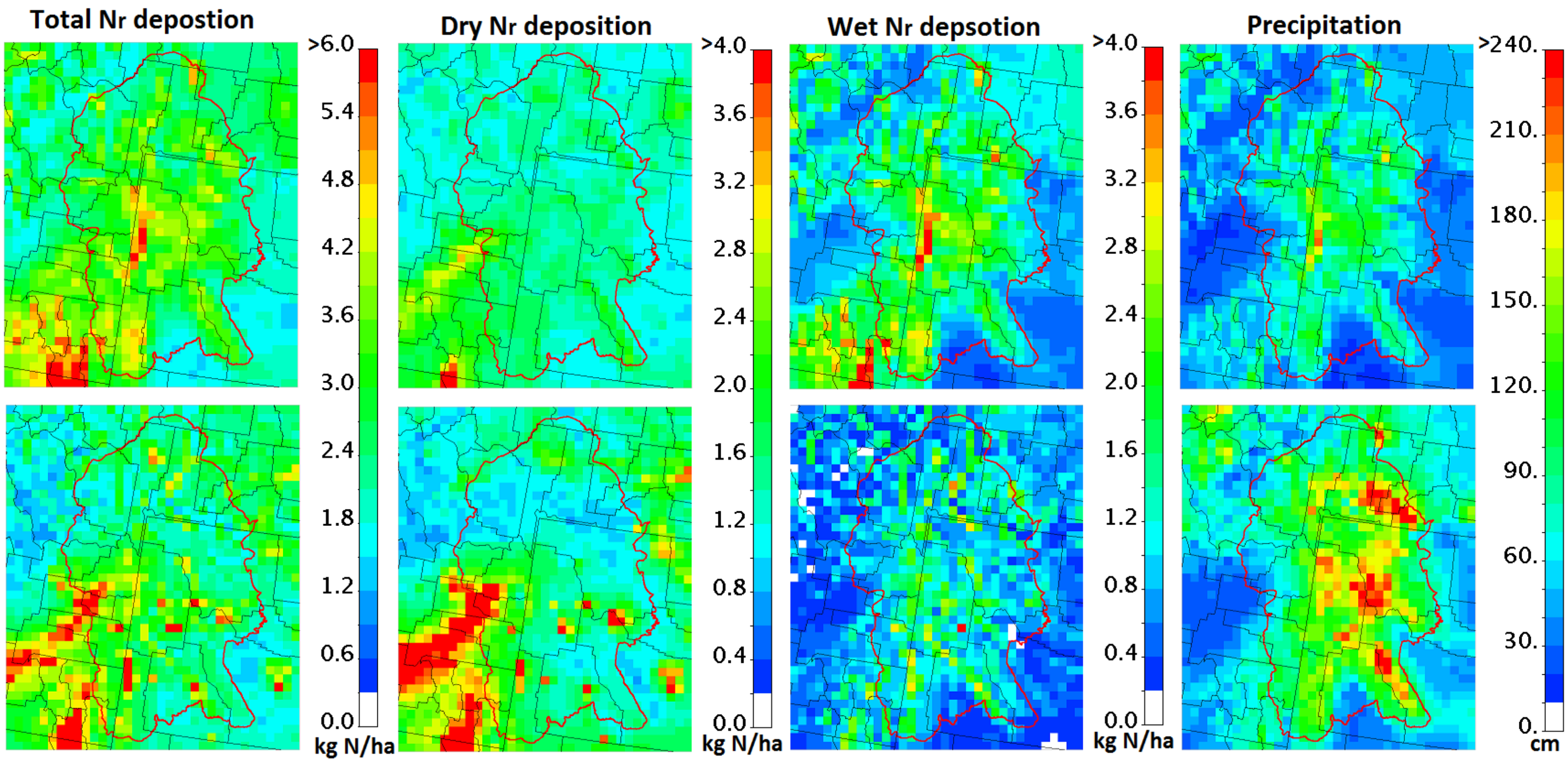


Figure S3. Spatial pattern comparison of the annual total, dry, and wet deposition, as well as precipitation from the NADP Total Deposition Map (TDEP, above panel) and corresponding CAMx/WRF simulation results (lower panel) over the Greater Yellowstone Area (GYA).

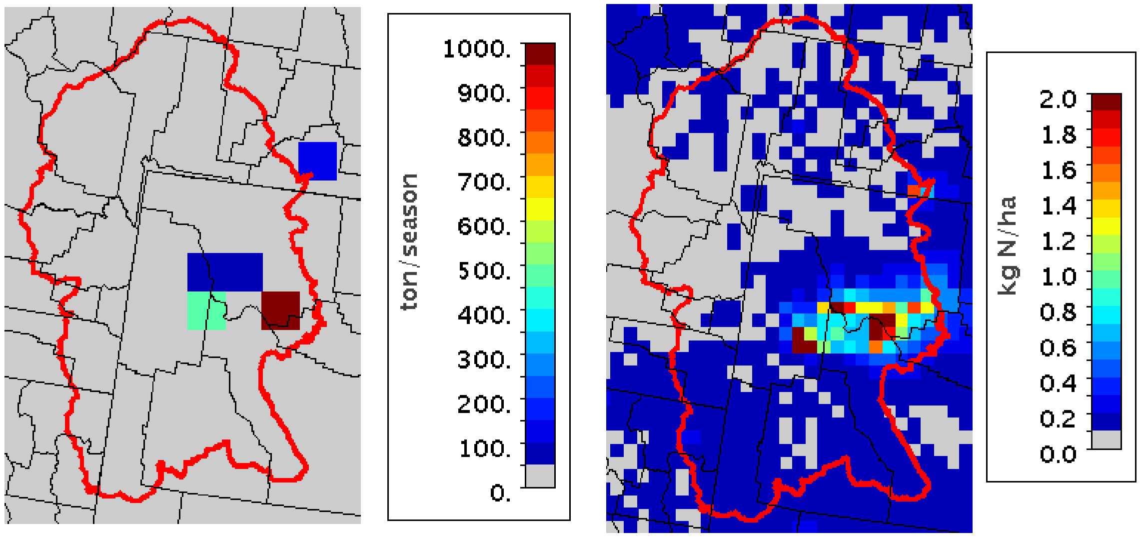


Figure S4. (left) Spatial pattern of total NOx emission from Fire emission sectors during summer (June, July, August) 2011 near the Greater Yellowstone Area (GYA). (right) Spatial pattern of total N deposition attributed to Fire emission during summer 2011.

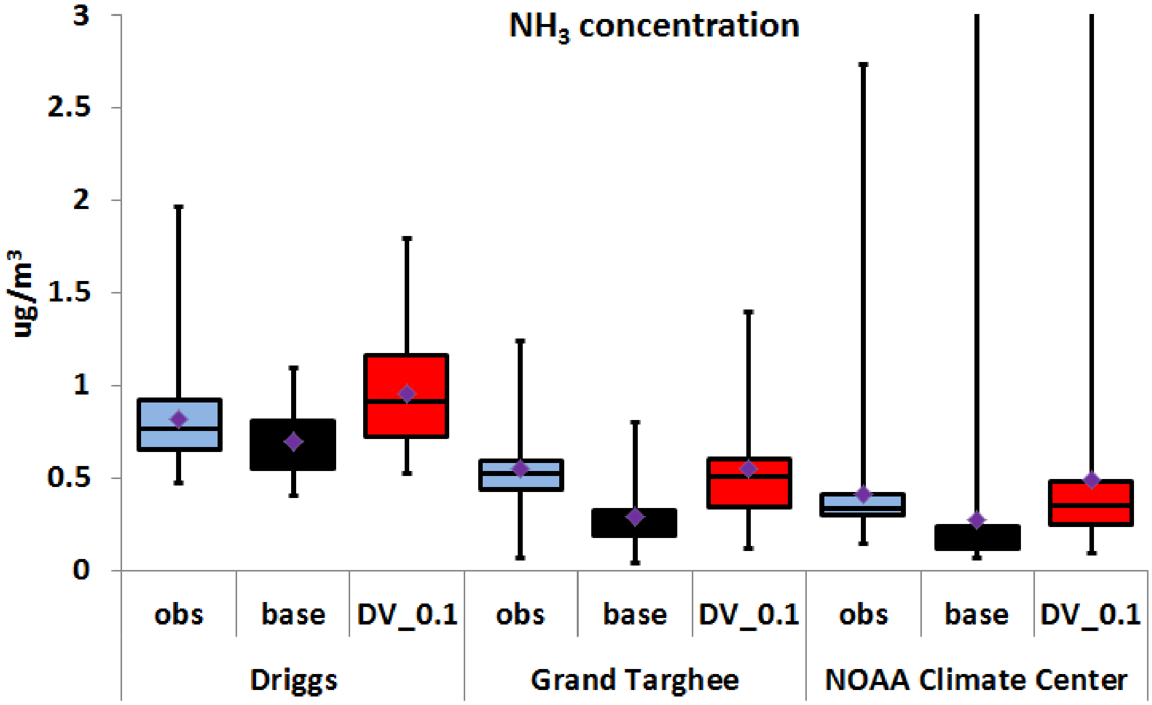


Figure S5. The sensitivity of the NH_3 dry deposition velocity to the simulated NH_3 concentrations at the three core sites during the GrandTReNDS study in July–August 2011.

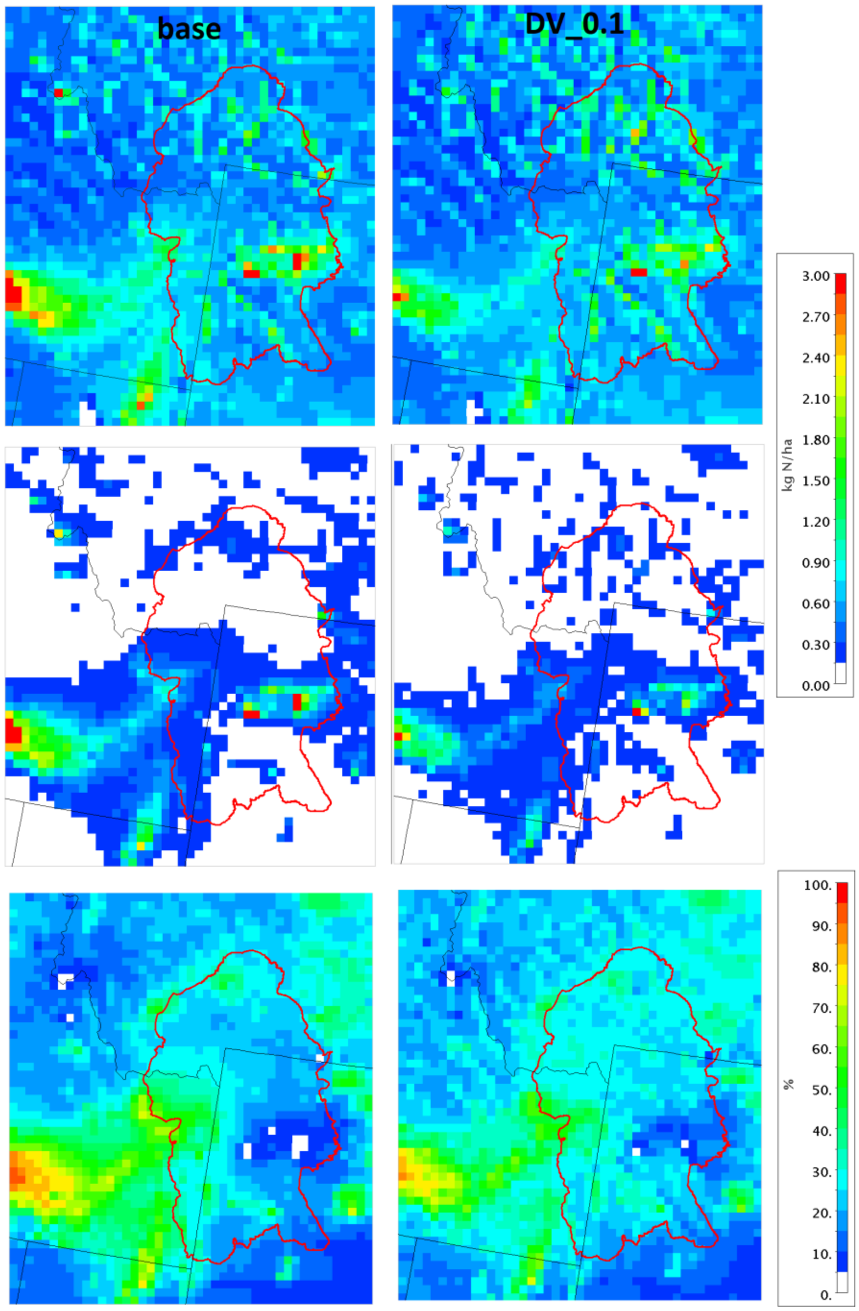


Figure S6. Change of spatial patterns of the simulated total N_r deposition (top panel), total NH3 deposition (middle panel) as well as contributions from agricultural emissions sector to total N_r deposition budget (bottom panel) over the Greater Yellowstone Area (GYA) during July–August 2011 due to the change of NH_3 deposition velocity in CAMx.

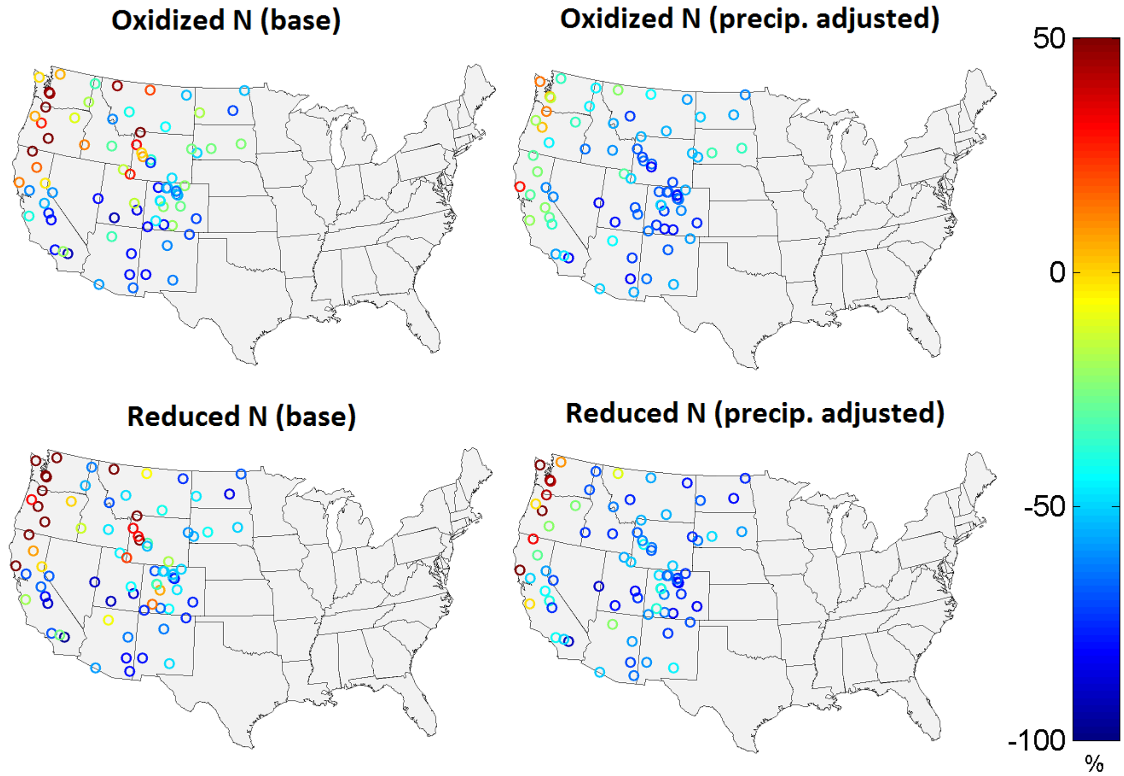


Figure S7. Change of model performance of CAMx wet oxidized and reduced nitrogen deposition simulation in terms of normalized mean bias at NADP NTN sites by implementing the precipitation adjustment technique followed by Appel et al. (2011).

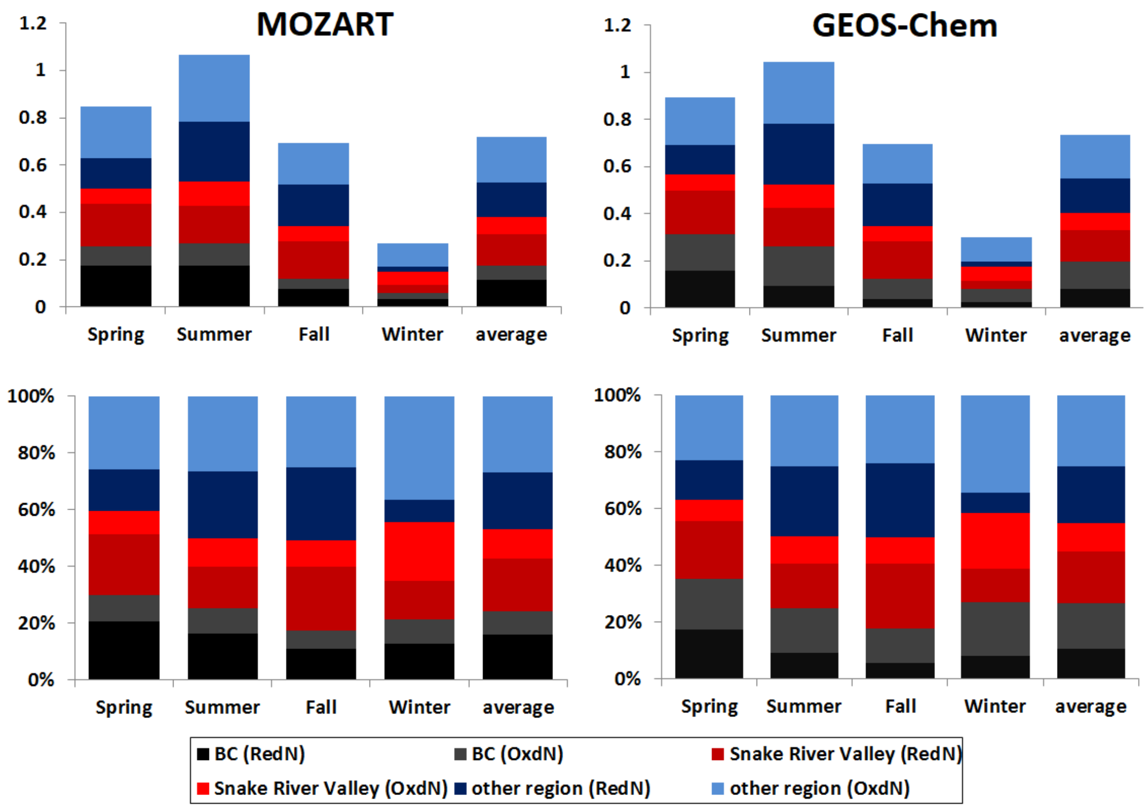
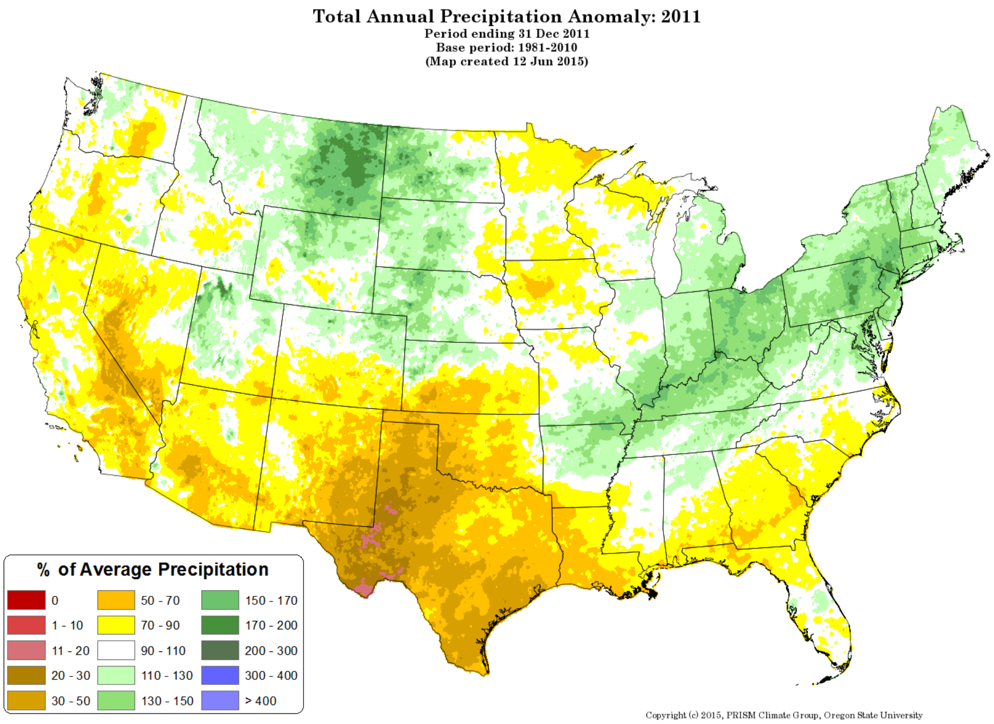


Figure S8. Sensitivity of MOZART (left) and GEOS-Chem (right) boundary conditions to average seasonal source apportionment results in 2011.



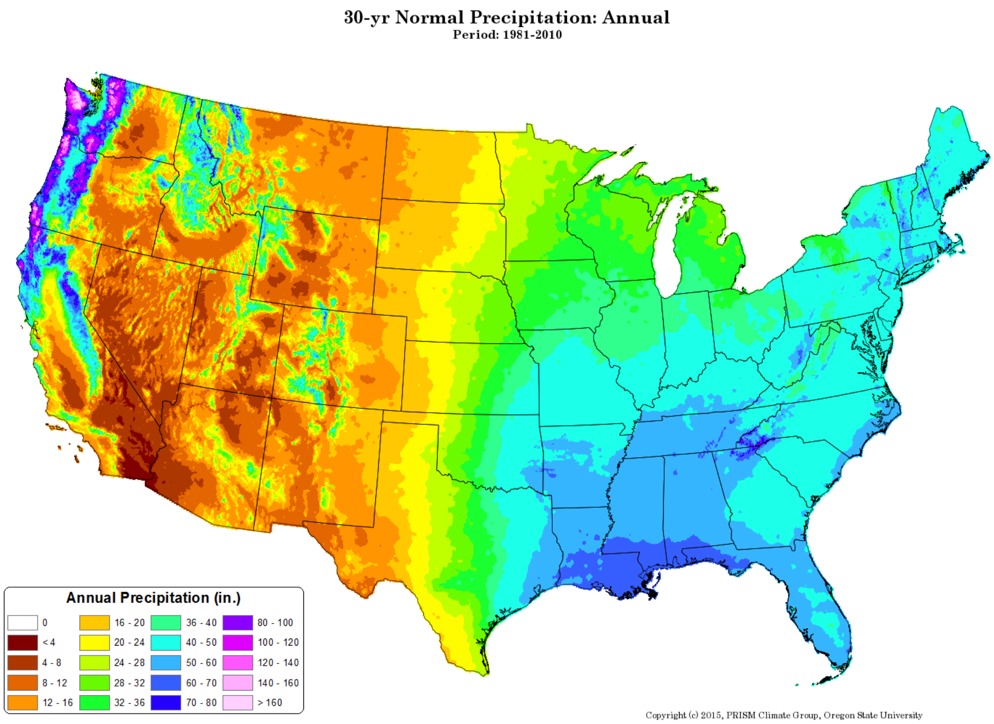


Figure S9. Total annual precipitation anomaly (in percentage) in the year 2011 compared with 30 years (1981-2010) normal annual precipitation from PRISM model (Copyright@2015, PRISM Climate Group, Oregon State University, http://prism.oregonstate.edu Map created July 2017).

Table S1. The model configuration of WRF-SMOKE-CAMx simulation platform for reactive nitrogen source apportionment study

Meteorological Modeling: WRF-ARW version 3.5.1	
Domain definition	Outer 36km domain (165x129 grid cells); Inner 12km domain (256x253 grid cell); Vertical layer: 37 layers from the ground to 50mb with 16 layers within first 1km height
Physics options	Microphysics: Thompson ice, snow and graupel scheme Longwave radiation: RRTMG Shortwave radiation: RRTMG PBL scheme: YSU planetary boundary layer Surface layer scheme: Monin-Obukhov Cumulus parameterization: Kain-Fritsch scheme Land-surface model: Unified NOAH
Data assimilation	Analysis nudging for winds, temperature and mixing ratio above PBL with nudging coefficients $5x10^{-4}$, $3x10^{-4}$ and $1x10^{-5}$ respectively
Initial condition	12km (Grid #218) North American Model (NAM)
Emission Modeling: SMOKE version 3.0	
Anthropogenic emission:	SMOKE version 3.0 with NEI2011v6 MOVES version 2010b for on-road mobile sources
Biogenic emission:	MEGAN version 2.1
Dust emission:	WRAP windblown dust model (WRAP-WBD)
Oil and gas emission:	SMOKE with Independent Petroleum Association of the Mountain States (IPAMS)
Lightning NOx:	ENVIRON generated based on NLCD lightning flash counts
Sea salt:	ENVIRON generated surf zone and open ocean PM emissions
Photochemical Modeling: CAMx version 6.10	
Domain definition	Outer 36km domain (148x112 grid cells); Inner 12km domain (227x230 grid cell); Vertical layer: 25 layer with layer 1 ~24m and model top ~ 19km MSL
Gas phase chemistry:	CB6r2
Deposition scheme:	Zhang et al. (2003)[1] dry deposition scheme CAMx-specific formulation for wet deposition
Aerosol module:	CF scheme for aerosol size distribution
Numeric options:	Gas phase chemistry solver: Euler Backward Iterative (EBI) Vertical advection scheme: Implicit scheme w/ vertical velocity update Horizontal advection scheme: Piecewise Parabolic Method (PPM)
Photolysis rate:	Day-specific ozone column data based on TOMS data measured by OMI
Boundary condition	MOZART global chemistry model (GCM) version 4.6
Initial condition	A fresh start with 15 days spin-up time

[1]Zhang, L., J.R. Brook, and R.Vet, (2003). A revised parameterization for gaseous dry deposition in air-quality models, Atmospheric Chemistry and Physics, 3, 2067-2082, doi:10.5194/acp-3-2067-2003.

Table S2. Summary of 27 tagged regions in CAMx PSAT of this study their corresponding annual emissions for NH_3 and NOx with agriculture (AG), oil and gas (OG), wildfires and prescribed fires (fire) and remaining emission source sectors (Other)

Tagged region	**Total emission for nitrogen species (tons/yr)**									
	NH3					**NOx**				
	AG	**OG**	**Fire**	**Other**	**total**	**AG**	**OG**	**Fire**	**Other**	**total**
NW Colorado (Southwest)[1]	4,900	0	55	418	5,373	0	12,046	564	54,827	67,437
NE Colorado (Southwest)	37,041	0	415	3,157	40,613	0	16,002	749	72,830	89,581
SE Colorado (Southwest)	20,281	0	227	1,728	22,237	0	20,869	976	94,980	116,825
SW Colorado (Southwest)	6,672	0	75	569	7,315	0	5,504	258	25,051	30,812
Upper Green River, Wyoming	2,358	0	525	110	2,993	0	11,412	3,016	43,523	57,952
Jackson, Wyoming	2,375	0	529	111	3,015	0	477	126	1,817	2,420
Eastern Wyoming (Other WY)	7,298	0	1,625	342	9,265	0	3,013	796	11,490	15,299
Western Wyoming (Other WY)	18,046	0	4,018	845	22,910	0	10,925	2,887	41,662	55,474
Yellowstone (Other WY)	1,511	0	336	71	1,918	0	761	201	2,902	3,864
Northern Idaho (Northwest)	16,887	0	2,193	910	19,991	0	669	6,906	47,036	54,612
Snake River Valley, Idaho	43,696	0	5,674	2,356	51,726	0	682	7,030	47,882	55,594
Northern Utah	12,946	0	69	2,163	15,178	0	10,235	200	92,312	102,747
Southern Utah (Southwest)	10,083	0	54	1,685	11,822	0	8,907	174	80,338	89,419
Nevada	5,569	0	825	2,533	8,926	0	189	2,725	107,900	110,814
Montana	54,343	0	7,531	1,313	63,187	0	13,806	11,510	153,220	178,537
Washington (Northwest)	44,118	3	825	7,400	52,345	0	467	2,458	268,831	271,757
Oregon (Northwest)	43,626	0	8,858	5,164	57,649	0	925	28,231	146,062	175,218
California	203,204	155	3,056	111,240	317,655	0	8,806	9,457	669,421	687,684
Mexico (Non U.S.)					246,344					782,600
New Mexico (Southwest)	35,327	0	4,374	2,673	42,374	0	71,863	15,197	170,550	257,609
Arizona (Southwest)	33,247	0	9,041	8,520	50,808	0	1,489	26,817	250,201	278,506
Texas & Oklahoma (Southwest)	364,835	44	24,481	39,179	428,539	0	410,736	35,635	1,450,095	1,896,465
Canada (Non U.S.)					421,830					934,900

North Dakota (Eastern U.S. + Great Plains)	93,163	0	952	6,995	101,110	0	8,408	1,407	171,869	181,683
Pacific (Non U.S.)					292					251,698
Far East U.S. (Eastern U.S. + Great Plains)					2,627,200					9,296,000
SD_KS_NE (Eastern U.S. + Great Plains)	480,670	4	6,245	9,439	496,359	0	96,945	25,572	666,950	789,467
Total:					**5,128,972**					**16,834,975**

Note:[1]the items in the parentheses are aggregate regions based on prevailing wind patterns over the GYA for the source apportionment results report in Figure 9-11

Table S3. Summary of regional air quality model nitrogen related species performance in terms of normalized mean bias (NMB) evaluated by the observations over the continental United States

Species	Photochemical model	Duration	Model resolution	Region evaluated	NMB value	Reference
NH3	CAMx	2011 full year	12km	GYA	-65%	this study
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest U.S.	[-23% -79%]	Zhang et al. (2013)
	CAMx	2009 full year	36km/12km	Colorado	-55%	Thompson et al. (2015)
	CAMx	Summer 2011	4km	Colorado	[-83% 46%]	Li et al. (2017)
HNO3	CAMx	2011 full year	12km	GYA	108%	this study
	CAMx	2009 full year	36km/12km	Colorado	23%	Thompson et al. (2015)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest U.S.	[-17% 45%]	Zhang et al. (2013)
PM25 nitrate	CAMx	2011 full year	12km	GYA	[37% 58%]	this study
	CAMx,CMAQ, WRF-Chem (n=34)	varies, case study to full year simulation	9-45km	varies, states to CONUS	[-49% 11%]	Simon et al. (2012)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest U.S.	[-92% -103%]	Zhang et al. (2013)
	CAMx	2009 full year	36km/12km	Colorado	57%	Thompson et al. (2015)
	CMAQ	2002 full year	12km	CONUS	[-24% 45%]	Bash et al. (2013)
	CMAQ	1990-2010	108km	CONUS	[-41% 106%]	Xing et al. (2015)
	WRF-CMAQ	Aug, Sep 2006	12/4km	CONUS/Texas	[-82% 83%]	Yu et al. (2014)
PM25 ammonia	CAMx	2011 full year	12km	GYA	3%	this study
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest US	[-57% 61%]	Zhang et al. (2013)
	CAMx,CMAQ, WRF-Chem (n=31)	varies, case study to full year simulation	9-45km	varies, states to CONUS	[-17% 7%]	Simon et al. (2012)
	CAMx	2009 full year	36km/12km	Colorado	-31%	Thompson et al. (2015)
	CMAQ	1990-2010	108km	CONUS	[-54% 23%]	Xing et al. (2015)
	WRF-CMAQ	Aug, Sep 2006	12/4km	CONUS/Texas	[-75% 48]	Yu et al. (2014)
NOx wet deposition	CAMx	2011 full year	12km	GYA	31%	this study
	CAMx,CMAQ, WRF-Chem (n=16)	varies, case study to full year simulation	9-45km	varies, states to CONUS	[-45% 19%]	Simon et al. (2012)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest U.S.	[-94% 52%]	Zhang et al. (2013)

NH4 wet deposition	CAMx	2011 full year	12km	GYA	49%	this study
	CAMx,CMAQ, WRF-Chem (n=16)	varies, case study to full year simulation	36km/12km	varies, states to CONUS	[-33% 28%]	Simon et al. (2012)
	CMAQ,CAMx	Jan, Jul 2002	4km	Southwest U.S.	[-51% 19%]	Zhang et al. (2013)
	CMAQ	2002 full year	12km	CONUS	[-16% 18%]	Bash et al. (2013)

Table S4. Detail source attribution results of 27 tagged source regions as well as agriculture (AG), oil and gas (OG), wildfires and prescribed fires (Fire) and remaining emission source sectors (Other) as well as boundary conditions (BC) to average N_r deposition at each season in 2011

Tagged region	**Season**	**Total N ($g\ N\ ha^{-1}\ season^{-1}$)**					**Dry N (percentage)**					**Reduced N (percentage)**				
		AG	**OG**	**Fire**	**Other**	**BC**	**AG**	**OG**	**Fire**	**Other**	**BC**	**AG**	**OG**	**Fire**	**Other**	**BC**
NW Colorado	Winter	0.0	0.0	0.0	0.0		15%	53%	85%	71%		100%	0%	1%	5%	
	Spring	0.1	0.3	0.0	0.4		26%	63%	51%	66%		100%	0%	2%	1%	
	Summer	0.0	0.1	0.0	0.1		19%	60%	61%	51%		100%	0%	3%	2%	
	Fall	0.1	0.3	0.0	0.5		29%	63%	57%	63%		100%	0%	2%	2%	
NE Colorado	Winter	0.0	0.0	0.0	0.0		93%	98%	98%	88%		100%	0%	1%	7%	
	Spring	0.2	0.1	0.0	0.4		20%	67%	51%	60%		100%	0%	2%	2%	
	Summer	0.0	0.0	0.0	0.1		33%	60%	61%	59%		100%	0%	3%	2%	
	Fall	0.6	0.2	0.0	1.1		42%	73%	57%	67%		100%	0%	2%	4%	
SE Colorado	Winter	0.0	0.0	0.0	0.0		80%	98%	98%	85%		100%	0%	2%	8%	
	Spring	0.0	0.0	0.0	0.1		9%	45%	51%	48%		100%	0%	2%	2%	
	Summer	0.0	0.0	0.0	0.1		31%	55%	61%	49%		100%	0%	3%	2%	
	Fall	0.2	0.0	0.0	0.4		32%	64%	57%	62%		100%	0%	2%	4%	
SW Colorado	Winter	0.0	0.1	0.0	0.0		27%	78%	85%	61%		100%	0%	1%	4%	
	Spring	0.1	0.2	0.0	0.2		20%	63%	51%	62%		100%	0%	2%	3%	
	Summer	0.1	0.2	0.0	0.3		10%	43%	61%	39%		100%	0%	3%	3%	
	Fall	0.2	0.3	0.0	0.3		25%	62%	57%	54%		100%	0%	2%	1%	
Upper Green River	Winter	1.2	1.0	2.3	0.7		48%	79%	82%	61%		100%	0%	0%	4%	
	Spring	12.0	3.5	8.8	1.9		32%	54%	51%	41%		100%	0%	1%	5%	
	Summer	8.3	3.2	8.2	4.8		54%	77%	69%	78%		100%	0%	1%	2%	
	Fall	6.9	2.3	6.4	1.9		62%	84%	76%	100%		100%	0%	6%	5%	
Jackson	Winter	0.6	0.0	0.1	4.0		64%	78%	73%	94%		100%	1%	10%	9%	
	Spring	4.5	0.0	1.2	4.7		60%	96%	44%	78%		100%	0%	0%	3%	

	Summer	3.1	0.0	0.4	5.8	81%	60%	33%	89%	100%	43%	1%	8%
	Fall	3.6	0.0	3.5	4.5	85%	72%	89%	94%	100%	1%	72%	5%
Eastern Wyoming	Winter	0.0	0.1	0.2	0.0	79%	97%	97%	94%	100%	0%	67%	0%
	Spring	0.5	0.4	1.1	1.1	33%	68%	53%	83%	100%	0%	1%	1%
	Summer	0.1	0.1	0.3	0.0	41%	85%	79%	46%	100%	0%	84%	1%
	Fall	1.3	0.7	3.4	0.0	70%	92%	91%	10%	100%	0%	63%	0%
Western Wyoming	Winter	1.2	0.7	0.6	0.1	70%	84%	82%	99%	100%	0%	69%	0%
	Spring	10.8	1.9	2.6	0.1	48%	61%	51%	68%	100%	0%	1%	2%
	Summer	5.3	1.6	46.9	0.5	61%	83%	89%	81%	100%	0%	90%	2%
	Fall	6.6	1.5	9.9	0.0	73%	87%	85%	90%	100%	0%	71%	4%
Yellowstone	Winter	0.1	0.0	0.2	2.3	82%	92%	70%	81%	100%	0%	55%	5%
	Spring	0.6	0.0	0.9	2.2	68%	75%	53%	65%	100%	0%	1%	1%
	Summer	0.4	0.0	25.7	62.9	85%	90%	79%	75%	100%	0%	98%	1%
	Fall	0.5	0.0	13.9	43.6	86%	93%	94%	94%	100%	0%	97%	3%
Northern Idaho	Winter	0.3	0.0	1.6	0.8	40%	86%	55%	90%	100%	0%	5%	2%
	Spring	1.7	0.0	3.6	0.0	45%	77%	57%	88%	100%	0%	5%	3%
	Summer	0.8	0.0	1.9	1.8	57%	60%	56%	82%	100%	0%	12%	2%
	Fall	2.2	0.0	9.4	15.0	51%	78%	55%	46%	100%	0%	68%	4%
Snake River Valley	Winter	45.7	1.5	11.6	61.1	59%	70%	69%	73%	100%	0%	23%	7%
	Spring	259.5	1.7	18.0	62.0	65%	52%	54%	57%	100%	0%	18%	2%
	Summer	230.6	2.3	84.7	43.1	76%	77%	68%	99%	100%	0%	14%	9%
	Fall	259.3	1.8	66.3	22.0	82%	79%	86%	63%	100%	0%	28%	1%
Northern Utah	Winter	2.8	0.3	3.3	17.2	40%	73%	74%	73%	100%	0%	17%	4%
	Spring	14.7	1.1	6.0	31.8	25%	48%	41%	45%	100%	0%	5%	4%
	Summer	16.8	1.0	5.0	42.9	51%	70%	66%	70%	100%	0%	8%	3%
	Fall	12.4	0.8	3.6	23.9	49%	73%	63%	71%	100%	0%	6%	3%
Southern Utah	Winter	0.6	0.1	3.7	2.4	38%	74%	85%	77%	100%	0%	1%	1%
	Spring	3.6	0.2	4.9	3.5	14%	51%	51%	44%	100%	0%	2%	1%
	Summer	4.6	0.2	9.2	9.5	47%	63%	61%	78%	100%	0%	2%	0%

	Fall	4.6	0.2	7.5	6.0	36%	69%	57%	76%	100%	0%	2%	0%
Nevada	Winter	0.5	0.0	1.4	5.1	26%	54%	70%	65%	100%	0%	2%	3%
	Spring	3.5	0.1	2.5	12.9	18%	29%	37%	31%	100%	0%	2%	3%
	Summer	4.4	0.1	4.6	32.3	38%	61%	56%	67%	100%	0%	7%	3%
	Fall	1.8	0.0	2.3	8.5	43%	64%	53%	64%	100%	0%	10%	4%
Montana	Winter	0.6	0.2	0.8	2.2	47%	76%	82%	81%	100%	0%	4%	8%
	Spring	7.0	0.7	4.0	3.9	34%	61%	54%	69%	100%	0%	1%	5%
	Summer	0.8	0.1	0.9	1.6	38%	61%	55%	75%	100%	0%	6%	4%
	Fall	4.3	0.4	13.3	3.7	57%	82%	56%	61%	100%	0%	68%	3%
Washington	Winter	0.7	0.0	1.6	7.3	49%	80%	55%	78%	100%	1%	5%	6%
	Spring	3.7	0.0	4.4	7.3	50%	67%	61%	75%	100%	1%	4%	4%
	Summer	1.9	0.0	2.0	5.5	41%	65%	56%	65%	100%	7%	12%	4%
	Fall	2.7	0.0	7.7	0.5	48%	71%	70%	98%	100%	1%	11%	0%
Oregon	Winter	1.5	0.1	3.1	5.2	34%	66%	55%	54%	100%	0%	5%	2%
	Spring	10.4	0.1	8.2	11.0	40%	44%	61%	43%	100%	0%	4%	3%
	Summer	10.8	0.1	3.8	17.0	54%	73%	56%	71%	100%	0%	12%	0%
	Fall	6.2	0.1	17.4	2.7	54%	70%	58%	99%	100%	0%	28%	1%
California	Winter	1.2	0.2	4.9	13.2	23%	57%	67%	56%	100%	2%	6%	2%
	Spring	21.5	0.5	12.0	45.2	12%	31%	47%	26%	100%	3%	7%	2%
	Summer	29.3	0.6	10.6	56.0	35%	64%	57%	60%	100%	4%	18%	3%
	Fall	7.0	0.2	5.1	18.5	40%	61%	51%	56%	100%	2%	18%	2%
Mexico	Winter	0.0	0.0	0.3	0.0	78%	83%	76%	100%	1%	1%	15%	3%
	Spring	0.0	0.0	0.6	1.1	96%	57%	61%	33%	0%	0%	4%	1%
	Summer	0.0	0.0	0.6	20.2	60%	69%	54%	52%	43%	0%	1%	5%
	Fall	0.0	0.0	0.7	3.3	72%	58%	75%	41%	1%	0%	36%	6%
New Mexico	Winter	0.0	0.1	0.1	0.3	60%	91%	85%	89%	100%	0%	1%	1%
	Spring	0.1	0.1	0.1	0.5	17%	61%	51%	70%	100%	0%	2%	5%
	Summer	0.8	0.5	0.2	3.4	21%	41%	61%	40%	100%	0%	3%	8%
	Fall	0.6	0.4	0.1	1.5	24%	60%	57%	56%	100%	0%	2%	4%

Arizona	Winter	0.1	0.0	1.9	0.2		41%	82%	78%	89%		100%	0%	1%	3%	
	Spring	1.1	0.1	3.7	0.3		13%	50%	48%	57%		100%	0%	2%	1%	
	Summer	3.0	0.1	7.3	5.8		36%	59%	61%	60%		100%	0%	3%	2%	
	Fall	2.4	0.1	6.0	3.2		29%	66%	57%	70%		100%	0%	2%	2%	
Texas & Oklahoma	Winter	0.0	0.0	0.0	0.0		71%	97%	95%	51%		100%	0%	1%	6%	
	Spring	0.4	0.2	0.1	0.7		11%	55%	51%	51%		100%	0%	2%	7%	
	Summer	2.2	1.0	0.2	5.2		28%	52%	61%	52%		100%	0%	3%	8%	
	Fall	2.1	0.5	0.1	1.7		25%	56%	57%	50%		100%	0%	2%	2%	
Canada	Winter	0.0	0.0	7.5	2.8		78%	78%	73%	81%		1%	1%	9%	2%	
	Spring	0.0	0.0	6.9	1.9		96%	96%	67%	100%		0%	0%	4%	2%	
	Summer	0.0	0.0	4.6	0.0		60%	60%	63%	40%		43%	43%	2%	4%	
	Fall	0.0	0.0	9.3	1.8		72%	72%	75%	64%		1%	1%	40%	2%	
North Dakota	Winter	0.0	0.0	0.0	0.0		85%	94%	90%	93%		100%	0%	5%	6%	
	Spring	0.9	0.1	0.9	1.2		10%	52%	35%	54%		100%	0%	6%	2%	
	Summer	0.0	0.0	0.0	0.0		40%	81%	75%	93%		100%	0%	1%	4%	
	Fall	0.4	0.0	0.1	0.1		68%	86%	60%	100%		100%	0%	15%	0%	
Pacific	Winter	0.0	0.0	1.7	1.8		20%	78%	73%	40%		99%	1%	0%	0%	
	Spring	0.0	0.0	1.8	3.2		14%	96%	61%	47%		100%	0%	0%	0%	
	Summer	0.0	0.0	1.5	0.1		40%	60%	68%	99%		100%	43%	0%	0%	
	Fall	0.0	0.0	2.2	1.1		34%	72%	75%	49%		100%	1%	2%	0%	
Far East US	Winter	0.0	0.0	0.0	0.0		99%	100%	100%	100%		100%	0%	3%	0%	
	Spring	3.4	0.1	3.1	1.2		8%	55%	35%	87%		100%	1%	6%	2%	
	Summer	0.5	0.2	0.9	2.3		30%	60%	46%	57%		100%	0%	3%	6%	
	Fall	0.3	0.0	0.3	0.3		24%	54%	55%	45%		100%	0%	17%	3%	
SD_KS_NE	Winter	0.0	0.0	0.0	0.0		92%	99%	95%	100%		100%	0%	5%	2%	
	Spring	2.0	0.1	1.4	1.2		12%	55%	35%	61%		100%	0%	3%	0%	
	Summer	0.2	0.0	0.2	0.0		34%	59%	59%	64%		100%	0%	2%	0%	
	Fall	1.2	0.1	0.1	0.7		48%	67%	55%	86%		100%	0%	12%	0%	
	Winter					55.0					50%					58.4%

BC	Spring	235.4	21%	67.8%
	Summer	246.3	38%	64.1%
	Fall	115.1	52%	63.2%

Supplement File S1

Regional evaluation of CAMx nitrogen deposition in 2011

The Three-State Air Quality Study (3SAQS) performed photochemical grid modeling using the same modeling platform and input files as this study, but with the addition of a nested, 4-km domain centered over Colorado, Utah, and Wyoming (UNC and ENVIRON, 2014). The 3SAQS comprehensive model evaluation report (UNC and ENVIRON, 2015) compared the simulated Nr compound with regard to concentration and deposition against the routine measured data for all sites within the 12-km and 4-km domains. A subset of these results is reproduced in the attached Table. With the exception of NH3, the 3SAQS simulation generally reproduced the spatial and temporal variations of the ambient nitrogen concentrations over the western United States with a fractional bias (FB) smaller than ±60% (Boylan and Russell, 2006) (Table S3). However, there were important systematic biases. The oxidized nitrogen gases of NO2 and HNO3 were overestimated with FBs of 29% and 81%, respectively, while NH3 was underestimated by -101%. The CAMx model had different systematic biases for the simulated PNO3 and PNH4 concentration in the different networks. The PNO3 were underestimated at CASTNet and CSN sites while overestimated those at IMPROVE sites. For PNH4, the simulation underestimated data at CASTNet sites while overestimated those at CSN sites. The measured PNH4 at CASTNet and CSN suffered from a negative artifact due to volatilization (Yu et al., 2005) and an accurate model simulation should overestimate these measured concentrations. Note that the dry deposition value provided by CASTNet is not a direct flux measurement but rather the product of a measured concentration and an estimated dry deposition velocity derived from the Multilayer Model (MLM, Cooter and Schwede, 2000). Different deposition velocity algorithm used between MLM and regional CTM model such as CMAQ can impose uncertainties for dry deposition estimates (Schwede and Lear, 2014). Both oxidized and reduced N were underestimated by more than -50% in the wet deposition, with reduced N bias greater than oxidized N (-70% versus -58%, respectively). The biases in particulate compounds did not have any systematic patterns and varied by network and species.

Reference:

Boylan, J.W. and Russell, A.G. (2006). PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. *Atmospheric Environment*, 40(26), pp.4946-4959.

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Schwede, D. B., and Lear, G. G. (2014). A novel hybrid approach for estimating total deposition in the United States. Atmospheric Environment, 92, 207-220.

UNC-Chapel Hill and ENVIRON International Corporation (2014), *Three-State Air Quality Modeling Study (3SAQS) – Final modeling protocol: 2011 emissions & air quality modeling platform*, Available at : http://vibe.cira.colostate.edu/wiki/Attachments/Modeling/3SAQS_2011_WRF_MPE_v8_draft_Aug04_2014.pdf.

UNC-Chapel Hill and ENVIRON International Corporation (2015), *Three-State Air Quality Modeling Study (3SAQS) – Model performance evaluation (simulation year 2011)*, Available at: http://vibe.cira.colostate.edu/wiki/Attachments/Modeling/3SAQS_Base11a_MPE_Final_18Jun2015.pdf

Table. Summary of CAMx model performance for nitrogen compound concentrations and deposition simulations at different network sites over the WRAP region, evaluated by 3SAQS study*

Species	Network	Domain resolution	Mean Obs	Mean Sim	R	NMB (%)	NME (%)	FB (%)	FE (%)
Gaseous N_r concentration evaluation									
NO_2 (ppb)	AQS	4km	9.6	13.7	N.A.	58	93	29	63
NH_3 (ppb)	AMoN	4km	1.2	0.42	0.71	-66	69	-101	115
Particulate matter N_r concentration evaluation									
PNO_3(ug m^{-3})	CASTNet[1]	12km	0.51	0.46	N.A.	-9	64	-12	74
	CSN	12km	1.63	1.27	N.A.	-22	66	-30	75
	IMPROVE	12km	0.34	0.44	N.A.	30	94	31	83
PNH_4(ug m^{-3})	CASTNet[2]	12km	0.34	0.33	N.A.	-4	43	-7	41
	CSN	12km	0.71	0.77	N.A.	8	70	26	64
Average N_r dry deposition evaluation									
HNO_3(kg N ha^{-1})	CASTNet	4km	0.0084	0.0195	0.45	130	131	81.	83
PNO_3(kg N ha^{-1})	CASTNet	4km	0.0005	0.0007	0.10	28	84	15	76
PNH_4(kg N ha^{-1})	CASTNet	4km	0.0016	0.0023	0.21	26	53	27	49
Accumulated annual N_r wet deposition evaluation									
PNO_3(kg N ha^{-1})	NTN	12km	0.58	0.36	0.77	-38	40	-58	60
PNH_4(kg N ha^{-1})	NTN	12km	0.91	0.47	0.71	-48	52	-70	79

*For more detailed model performance statistics, refer to UNC and Environ (2015), 1-the measured PNO_3 at CASTNet sites including fine and coarse mode, which should be greater than CAMx counterparts with only fine particulate nitrate, 2-the measured PNH_4 at CASTNet sites has negative bias so is a lower bound of "true" particulate ammonium.