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Addendum to: "A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus"

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Supplementary material

Supplement 1. Description of the data screening and evaluation process (see Table 1)

The screening of measurement-based precipitation chemistry and wet and dry deposition data involved detailed assessments of site representativeness, sampling protocols, laboratory analyses, data completeness and overall measurement quality. A brief description of each follows.

Site representativeness: Site characteristics were screened at the network level to ensure that all data were regionally-representative (i.e., from rural/remote sites). Urban and industrial sites, i.e., located within 50 km of major industries or major urban areas, were explicitly excluded. At sites <100 km from saltwater coastlines, non-sea-salt sulfur wet deposition fluxes were calculated using the method described in WMO/GAW (2004). Sea salt corrections were also calculated at sites in Africa >100 km from the coastline where sea salt penetration was known to exceed 100 km. For all other sites >100 km from coastlines, measured sulfur wet deposition fluxes were assumed to be non-sea-salt in origin so no sea salt correction was done. Dry deposition S fluxes were not adjusted for sea salt.

Sampling protocols: Wet deposition fluxes were calculated from precipitation samples collected and chemically analyzed for major anions and cations, viz., SO_4^{2-}, NO_3^-, Cl^-, H^+, NH_4^+, Ca^{2+}, Mg^{2+}, Na^+ and K^+, and in some cases, organic acids and phosphorus. Data accepted for this assessment were required to meet the sampling and analysis protocols of the Global Atmospheric Watch (GAW) Precipitation Chemistry Progamme as documented in the Manual for the GAW Precipitation Chemistry Programme (WMO/GAW, 2004), namely: (1) sampling period ≤ 7 days (i.e., daily, event, weekly sampling), (2) samples collected in a wet-only collector accompanied by a national weather service standard precipitation gauge, (3) field quality control activities

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imbedded in the network operations, (4) suitable laboratory analytical and quality assurance/quality control procedures, (5) participation in external laboratory intercomparison studies and (6) acceptable data management and data QA/QC procedures. Special attention was given to "bulk" deposition data which involved the collection of samples in samplers left open to the atmosphere during wet and dry periods. Given the myriad problems and issues associated with bulk sampling (Vet, 1991; Wesley and Hicks, 2000; WMO/GAW, 2004), bulk deposition data were generally excluded. Exceptions were allowed where bulk measurements had been shown to be comparable to wet-only measurements and where no wet-only deposition data were available.

Calculations and Evaluation of Data Completeness: The 3-year average concentration and deposition values used in the assessment were calculated as the arithmetic means of the annual concentrations and deposition values (see the discussion below for details). The annual concentrations were calculated as precipitation-weighted (PW) mean concentrations and the annual wet deposition fluxes were calculated as the product of the annual PW mean concentration and the annual precipitation depth (mostly, but not always, measured by a standard gauge). All calculation procedures followed the guidelines documented in WMO/GAW (2004).

All of the wet deposition data were rigorously screened for data completeness and were required to meet the data completeness criteria discussed below (data that did not meet the data completeness criteria were excluded). All annual wet deposition data for the years 2000, 2001, 2002, 2005, 2006 and 2007 were required to meet the following criteria: %PCL $\geq$ 70% and % TP $\geq$ 50% where %PCL represents the Percent Precipitation Coverage Length (i.e., the percentage of days in a year during which precipitation was measured) and %TP represents the Percent Total Precipitation (i.e., the percentage of annual precipitation depth that was associated with valid concentration data). A complete description of these data completeness criteria can be found in WMO/GAW (2004).

Data Quality Rating: A data quality rating, either "*Satisfactory*" or "*Conditional*", was assigned to all 3-year average wet deposition and precipitation-weighted mean concentration values. *Satisfactory* data are indicated on global maps as circles and *Conditional* data as squares. The data quality ratings were assigned based on a multi-stage assessment of sampling methods and data completeness levels using the conditions shown in Table 1. A 3-year average value was deemed as *Satisfactory* if it met the sampling and data completeness conditions indicated in Table 1 and was derived from 2 or 3 *Satisfactory* annual values; otherwise, a value was deemed as *Conditional,* including the situation where it was derived from only a single year of *Satisfactory* or *Conditional* data. *Conditional* data, although considered of lower quality, were allowed in the final data set to fill gaps in the deposition patterns. While only 2–3% of the 3-year average wet deposition values were deemed to be *Conditional*, they tended to be located in data sparse areas and were therefore important to filling the spatial patterns. When calculating the % changes in wet deposition between 2000–2002 and 2005–2007, only Satisfactory data were used. This was done to minimize uncertainty in the calculated values.

In several regions of the world, measurement data were available only for time periods outside of the standard 2000–2002 and 2005–2007 periods, e.g., South Africa and Australia. These data were included on the global maps as "non-conforming" if they met two conditions, namely, if they were located in areas where no other data were available, and if the data were collected within a few years of the standard 3-year periods.

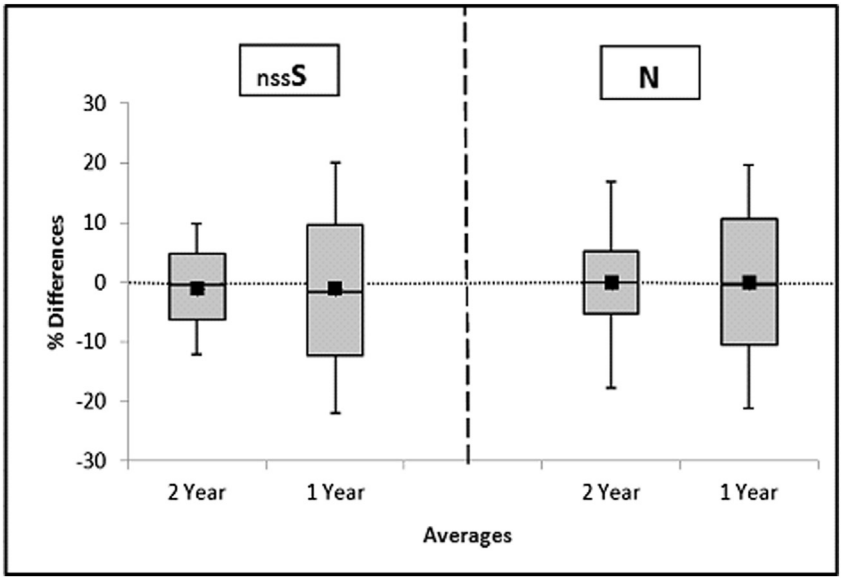

Fig. S1. Percent differences of the 2-year and 1-year averages from the 3-year averages for 2005–2007 wet deposition of nssS (left) and N (right) based on a sensitivity analysis of 411 sites with nssS data and 408 sites with N data. The bottom and top of the whiskers correspond to the 10th and 90th percentiles of the differences, the bottom and top of the boxes correspond to the 25th and 75th percentiles and the middle bars and squares correspond to the medians and means, respectively.

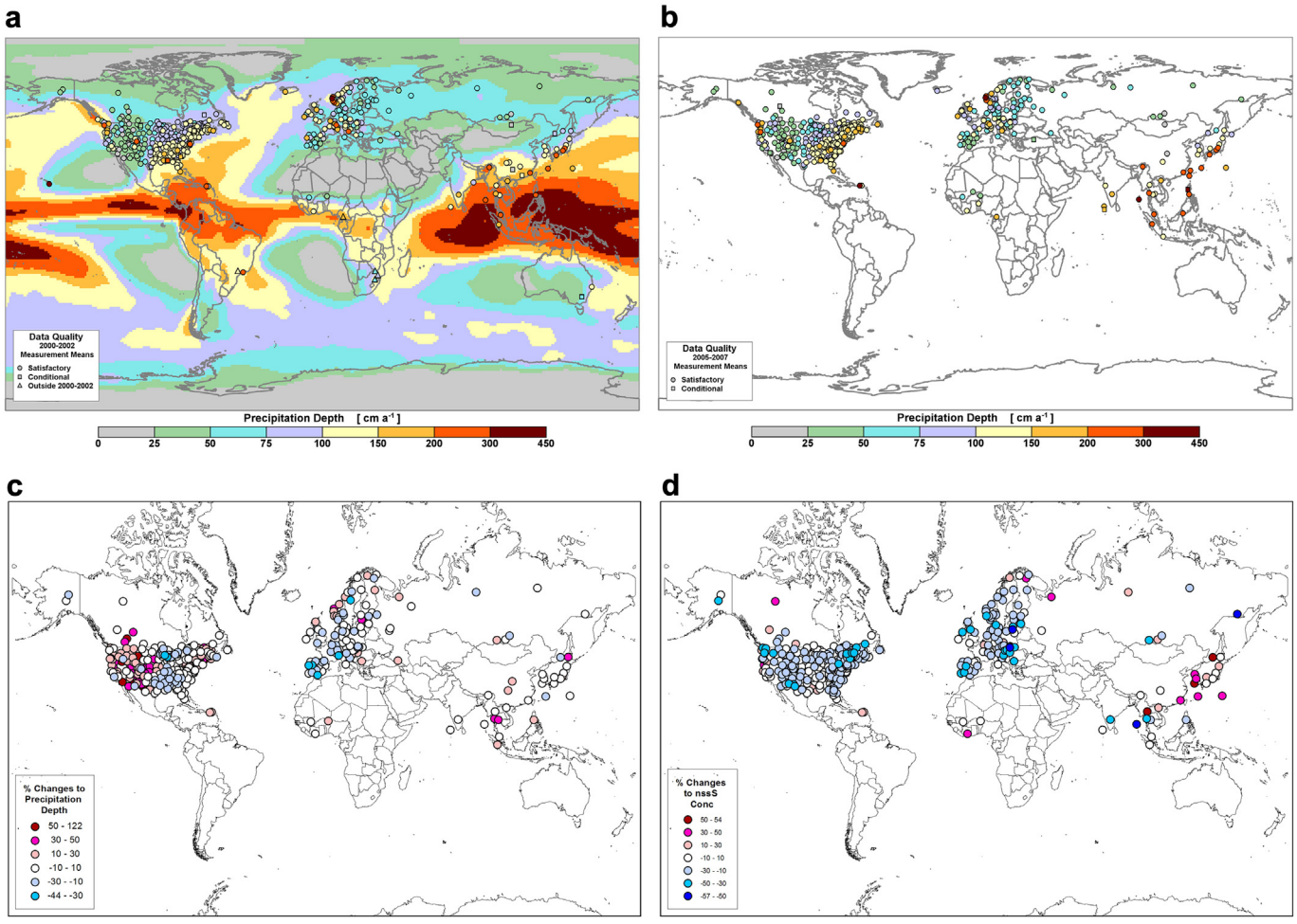

Fig. S2. (a) 2000–2002 3-year mean measurement-based and 2001 model-based precipitation depths in cm a^{-1}, (b) 2005–2007 3-year mean measured precipitation depths, (c) global % change between the 2000–2002 and 2005–2007 three-year mean precipitation depths calculated as 100 $[D_{2005-2007}-D_{2000-2002}]/[D_{2000-2002}]$ and (d) global % change between the 2000–2002 and 2005–2007 three-year mean precipitation-weighted mean concentrations of nssS (calculated as above).

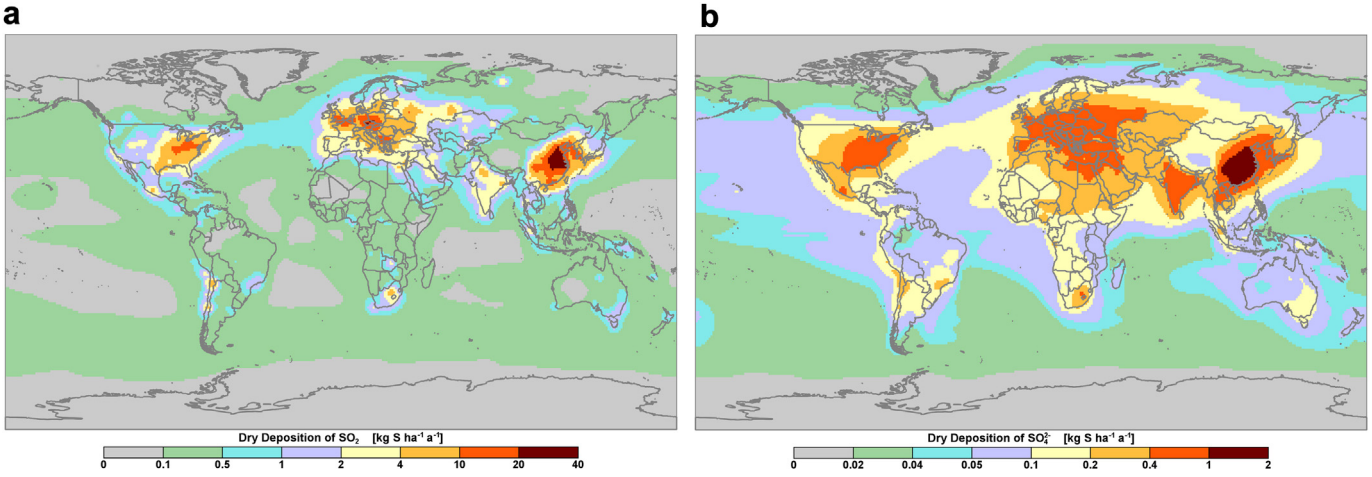

Fig. S3. 2001 ensemble-mean modeled patterns of dry deposition of: (a) SO_2-S and (b) $nssSO_4^{2-}$-S in kg S ha^{-1} a^{-1}.

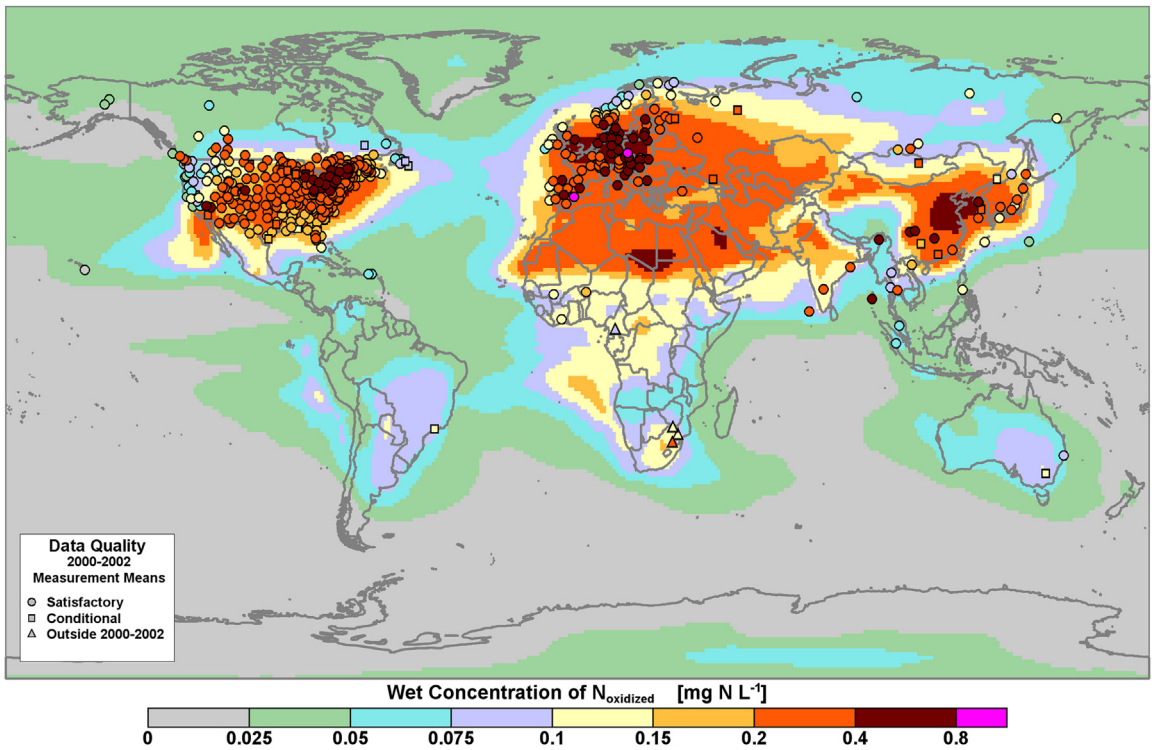

Fig. S4. Measurement-model pattern of precipitation-weighted mean concentration of $N_{oxidized}$ in mg N L^{-1}. Measurement values represent 3-year averages for 2000–2002; model results represent the 2001 model year.

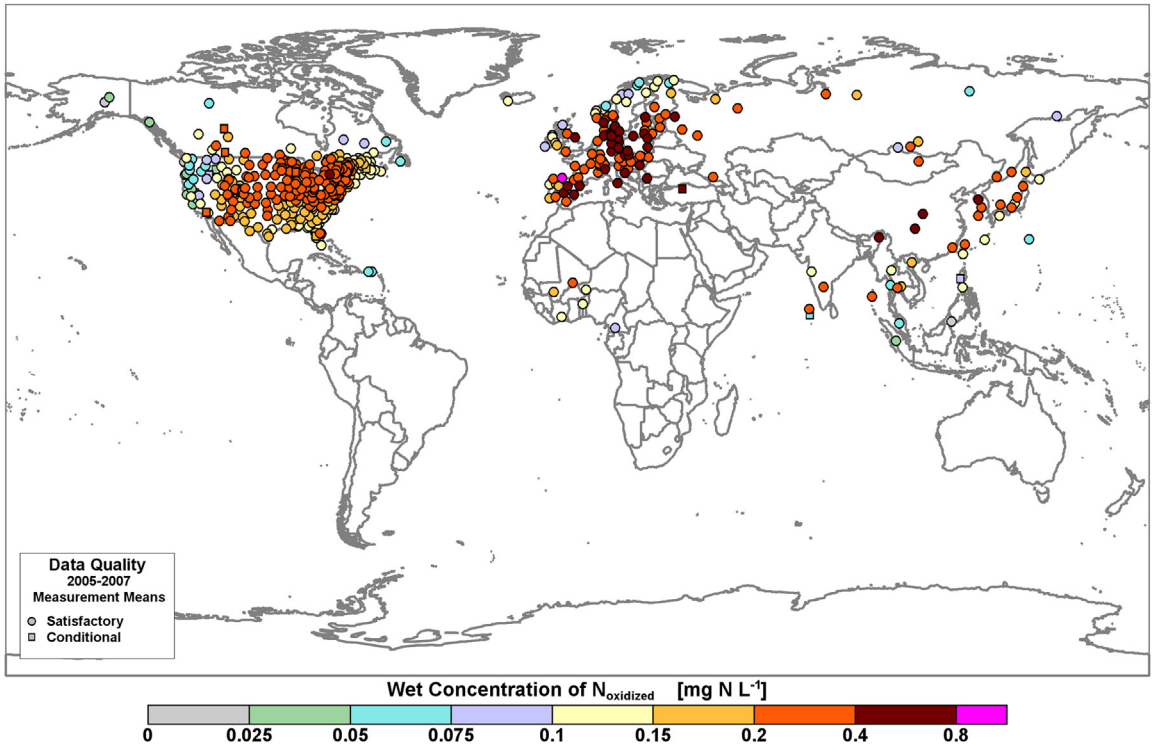

Fig. S5. 2005–2007 measurement-based spatial pattern of precipitation-weighted mean concentration of $N_{oxidized}$ in mg N L^{-1}.

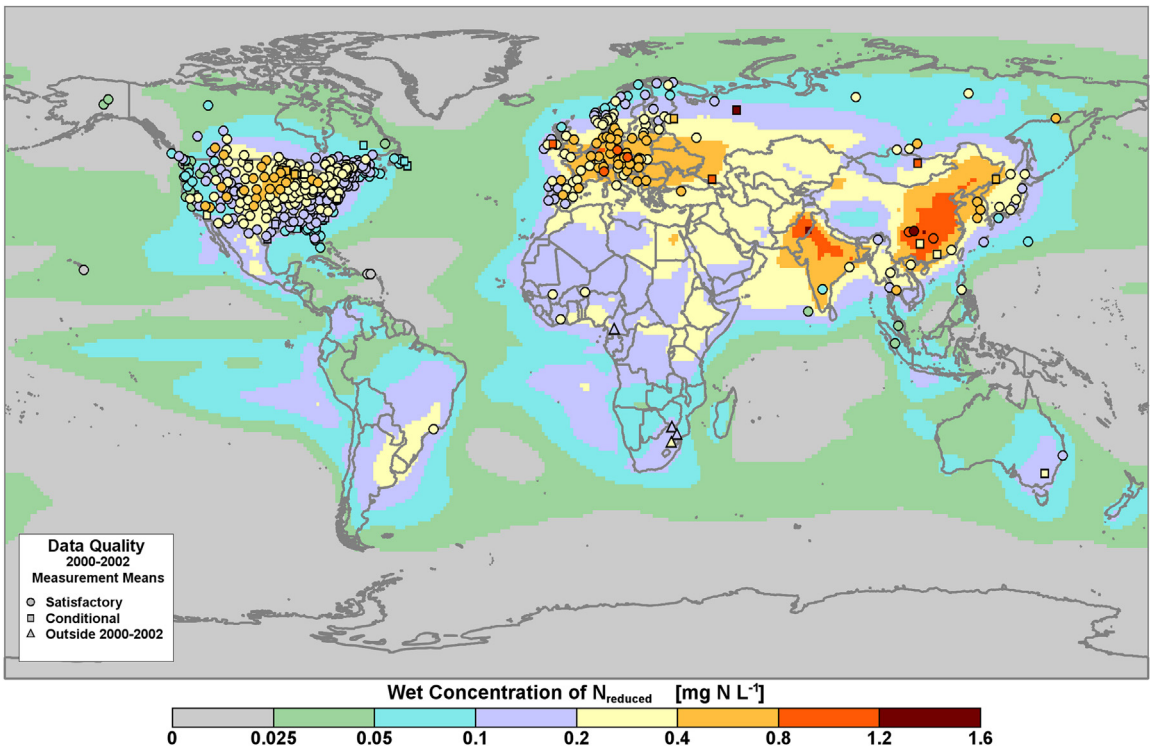

Fig. S6. Measurement-model pattern of precipitation-weighted mean concentration of $N_{reduced}$ in mg N L^{-1}. Measurement values represent 3-year averages for 2000–2002; model results represent the 2001 model year.

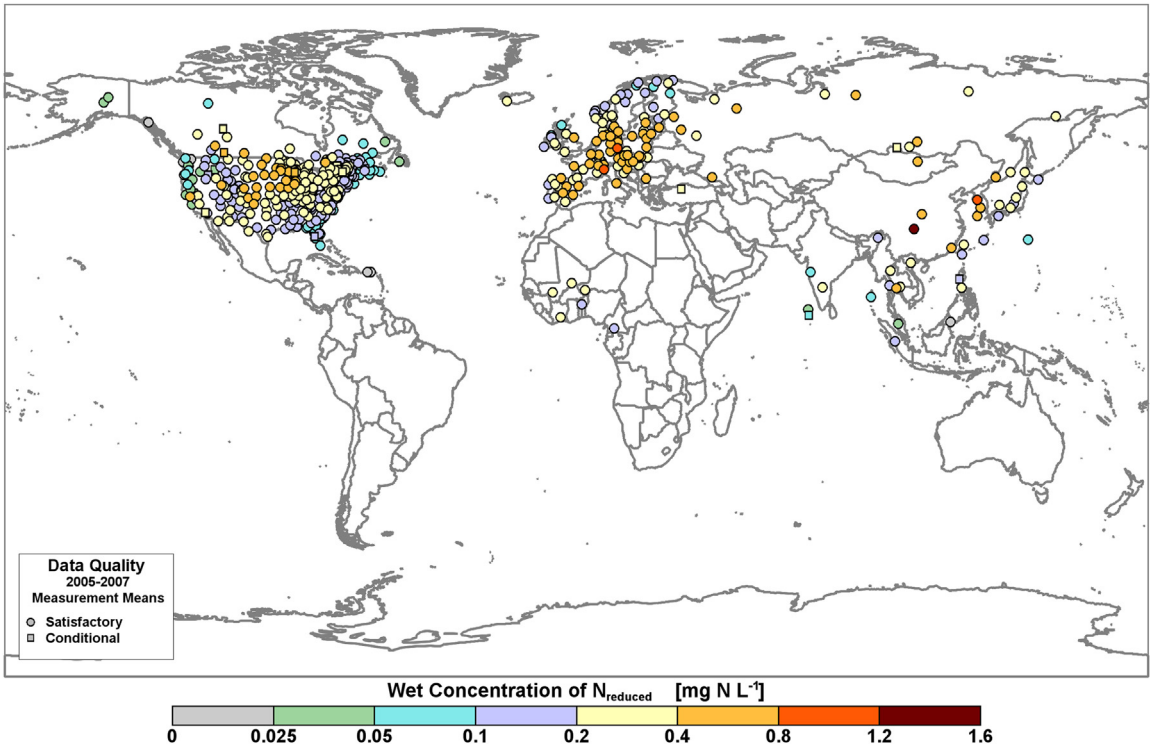

Fig. S7. 2005–2007 measurement-based pattern of precipitation-weighted mean concentration of $N_{reduced}$ in mg N L^{-1}.

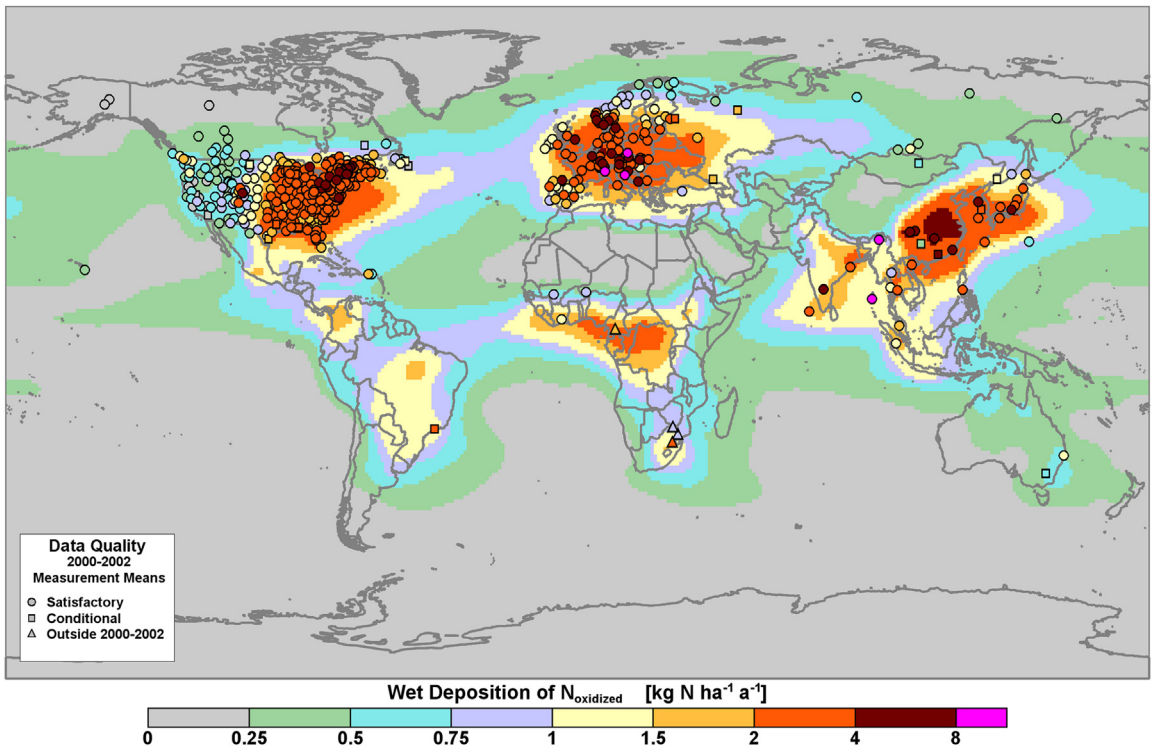

Fig. S8. Measurement-model pattern of wet deposition of $N_{oxidized}$ in kg N ha^{-1} a^{-1}. Measurement values represent 3-year averages for 2000–2002; model results represent the 2001 model year.

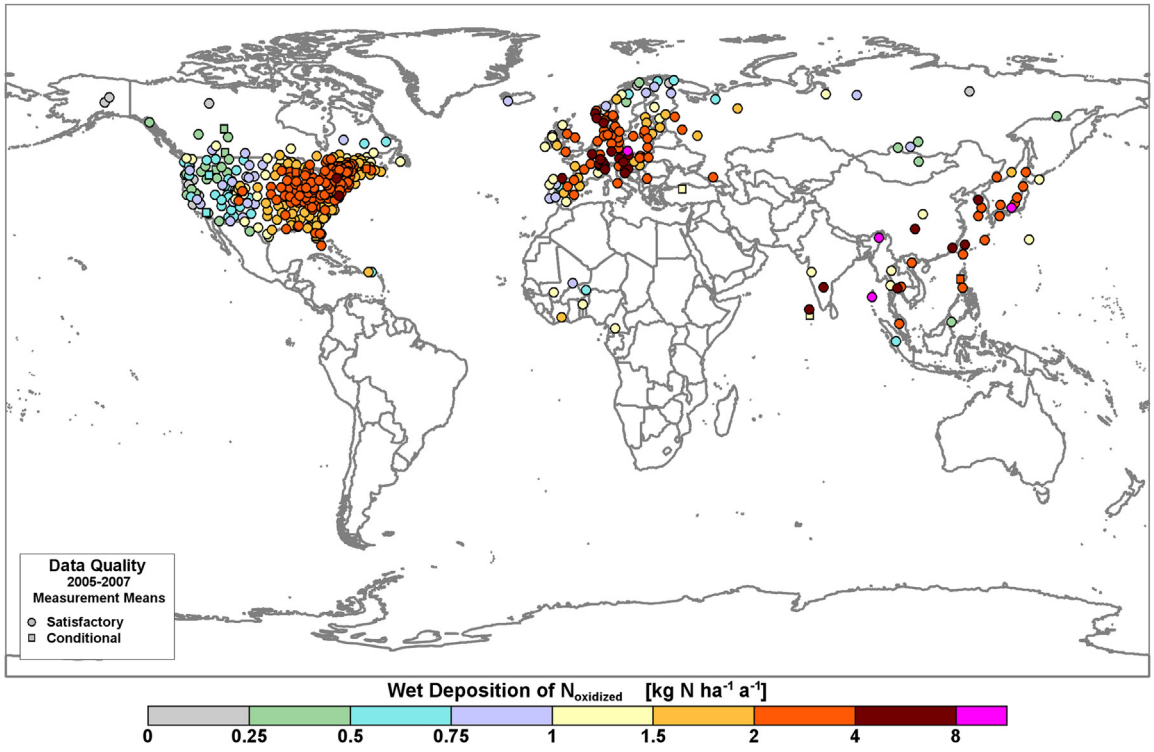

Fig. S9. 2005–2007 measurement-based pattern of 3-year average wet deposition of $N_{oxidized}$ in kg N ha^{-1} a^{-1}.

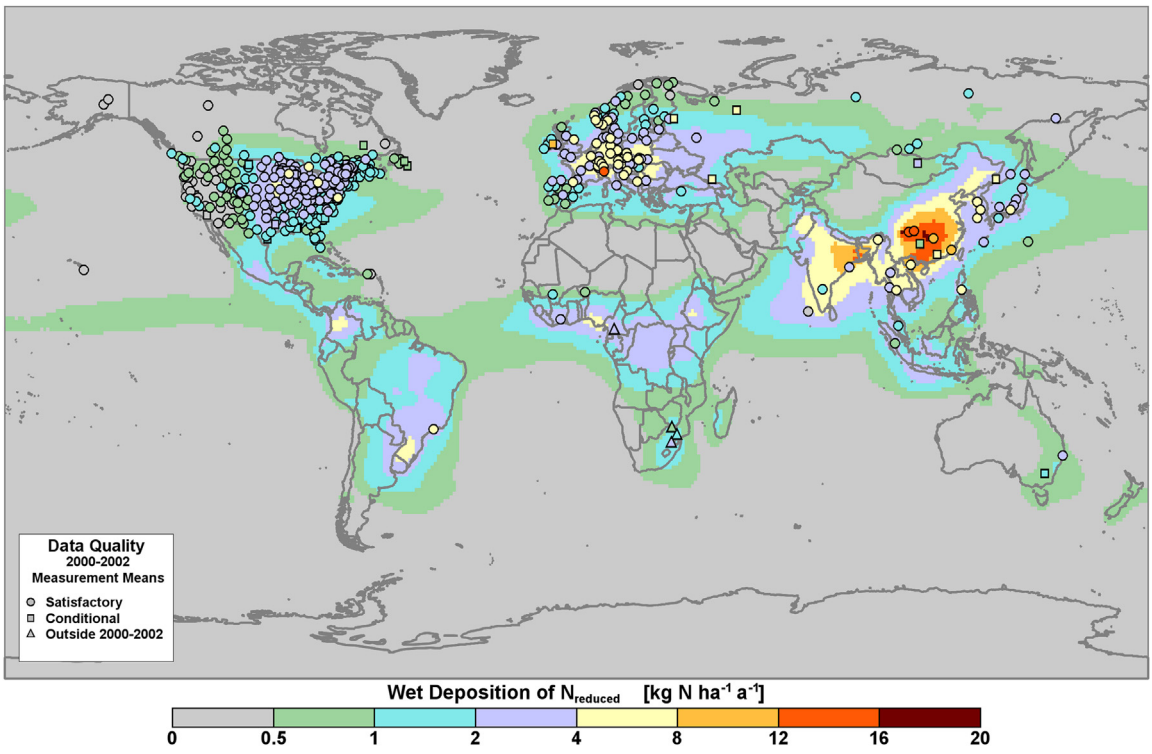

Fig. S10. Measurement-model pattern of wet deposition of $N_{reduced}$ in kg N ha^{-1} a^{-1}. Measurement values represent 3-year averages for 2000–2002; model results represent the 2001 model year.

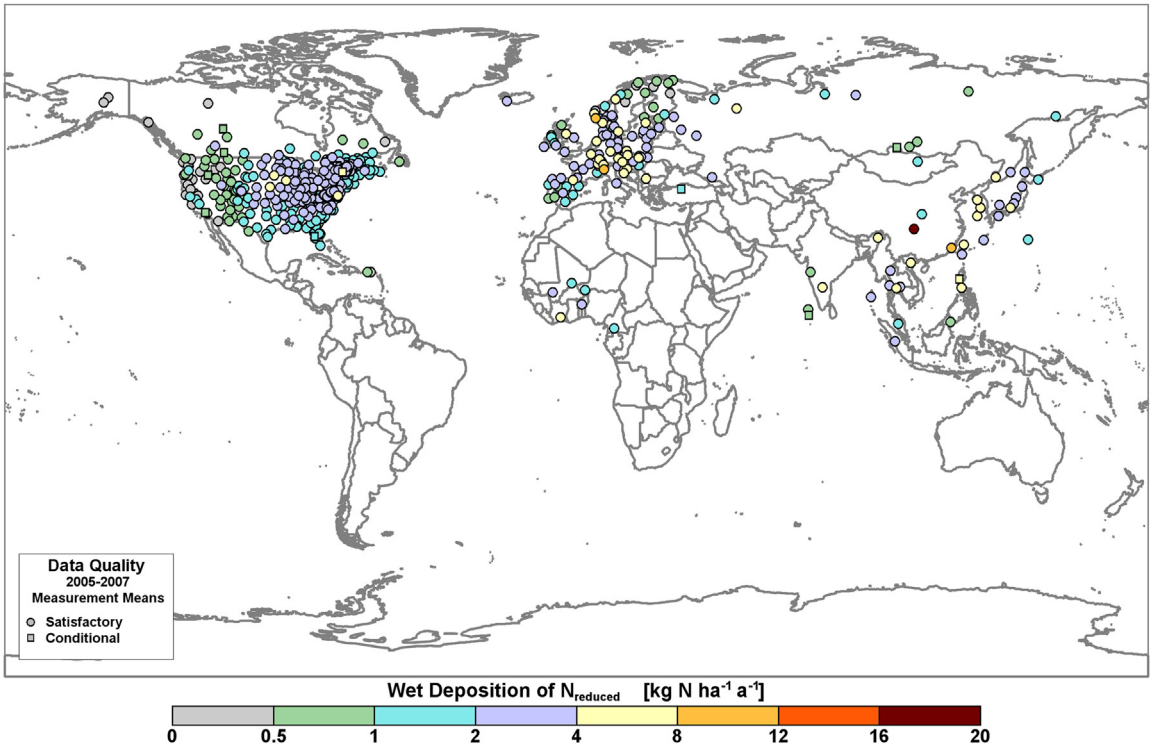

Fig. S11. 2005–2007 measurement-based pattern of 3-year average wet deposition of $N_{reduced}$ in kg N ha^{-1} a^{-1}.

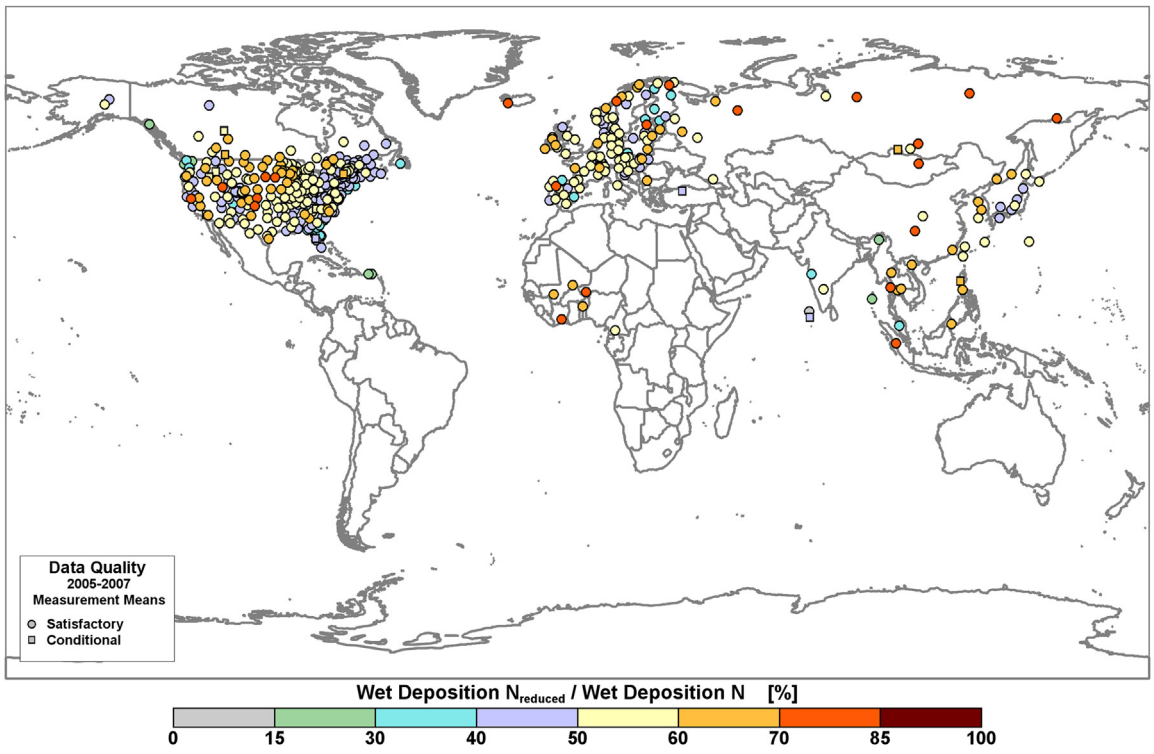

Fig. S12. 2005–2007 measurement-based % ratios of 3-year average wet deposition of $N_{reduced}/N$.

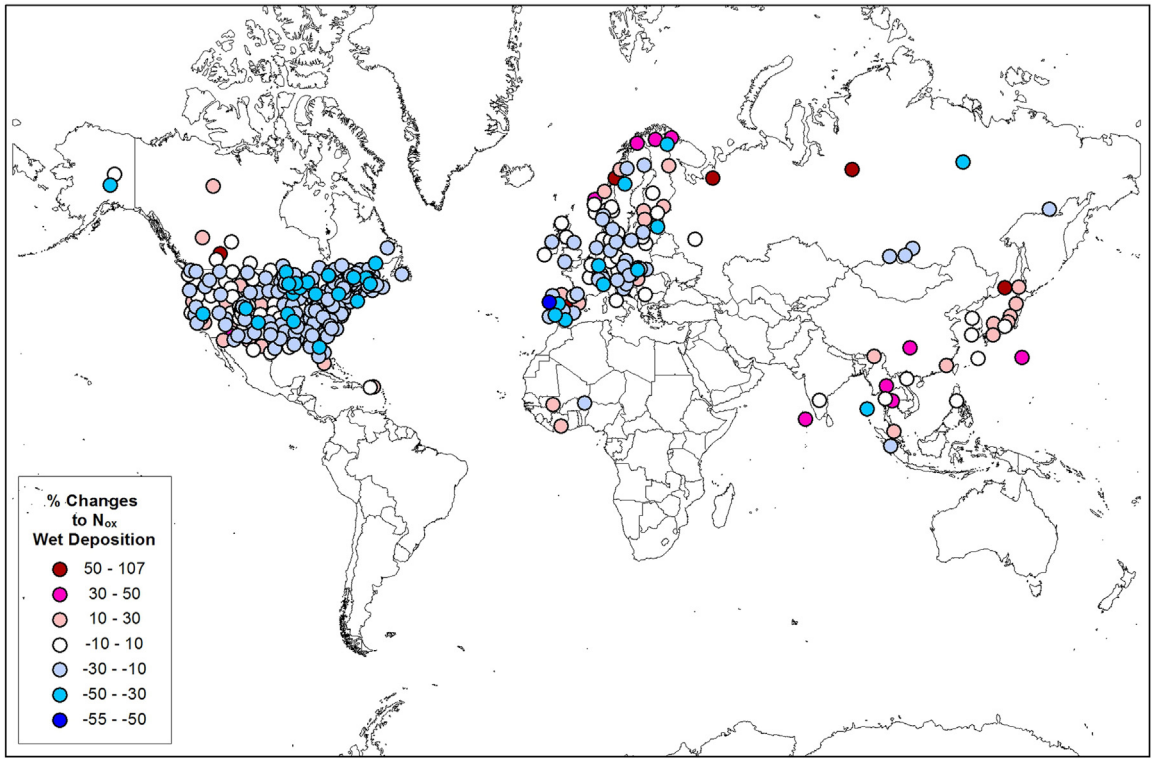

Fig. S13. % Change from 2000–2002 to 2005–2007 of measured three-year average wet deposition values of $N_{oxidized}$ calculated as 100 $[C_{2000-2005}-C_{2000-2002}]/[C_{2000-2002}]$ where C = 3-year average wet deposition in kg N ha^{-1} a^{-1}.

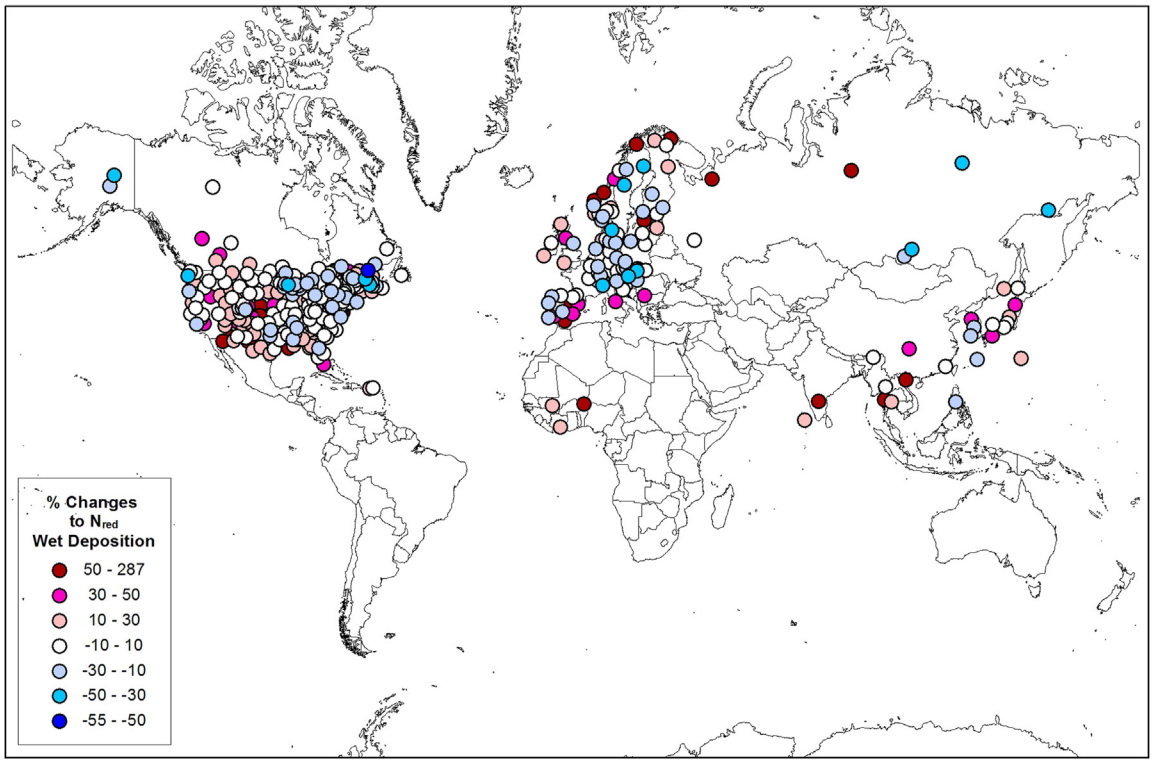

Fig. S14. % Change from 2000–2002 to 2005–2007 of measured three-year average wet deposition values of $N_{reduced}$ calculated as 100 $[C_{2000-2005}-C_{2000-2002}]/[C_{2000-2002}]$ where C = 3-year average wet deposition in kg N ha^{-1} a^{-1}.

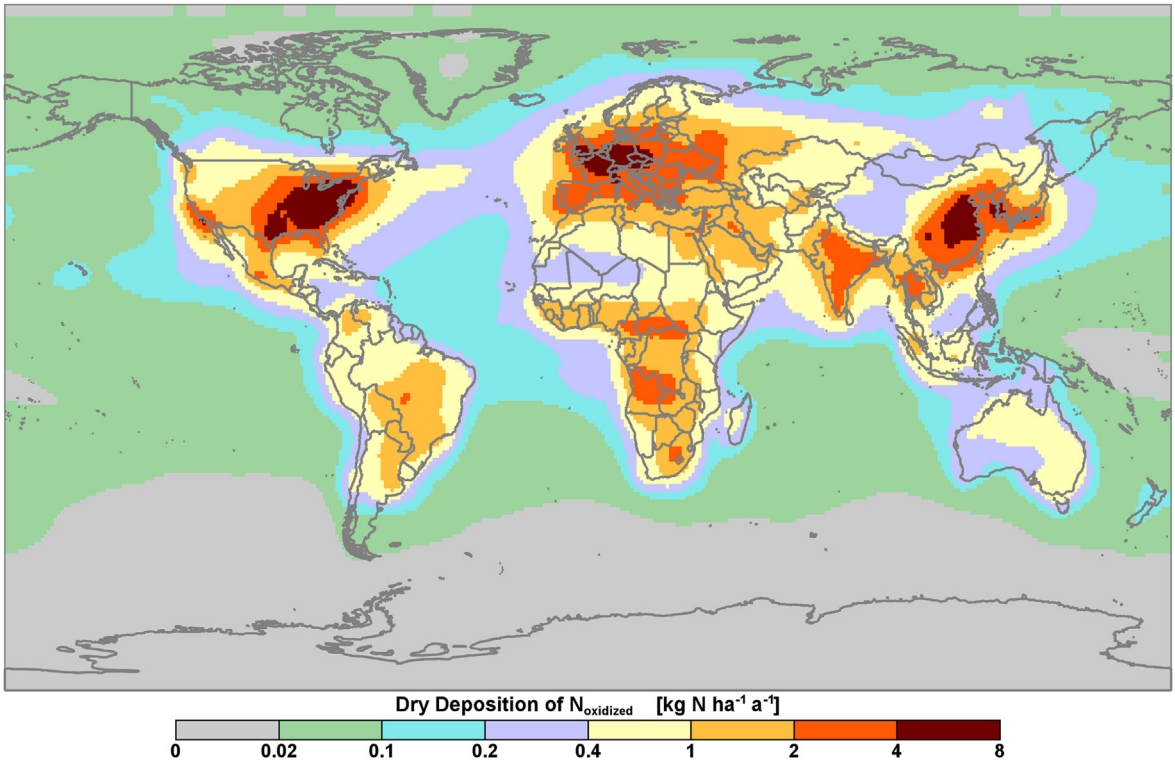

Fig. S15. 2001 ensemble-mean pattern of dry deposition of $N_{oxidized}$ in kg N ha^{-1} a^{-1}.

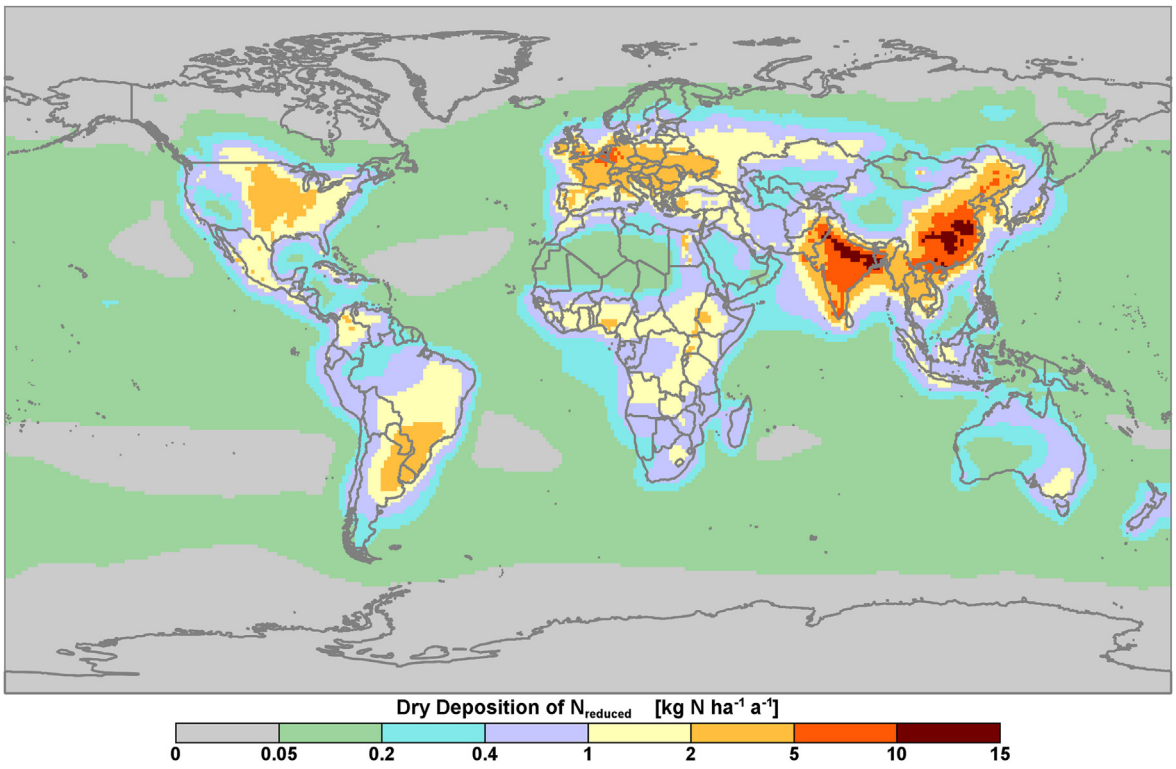

Fig. S16. 2001 ensemble-mean pattern of dry deposition of $N_{reduced}$ in kg N ha^{-1} a^{-1}.

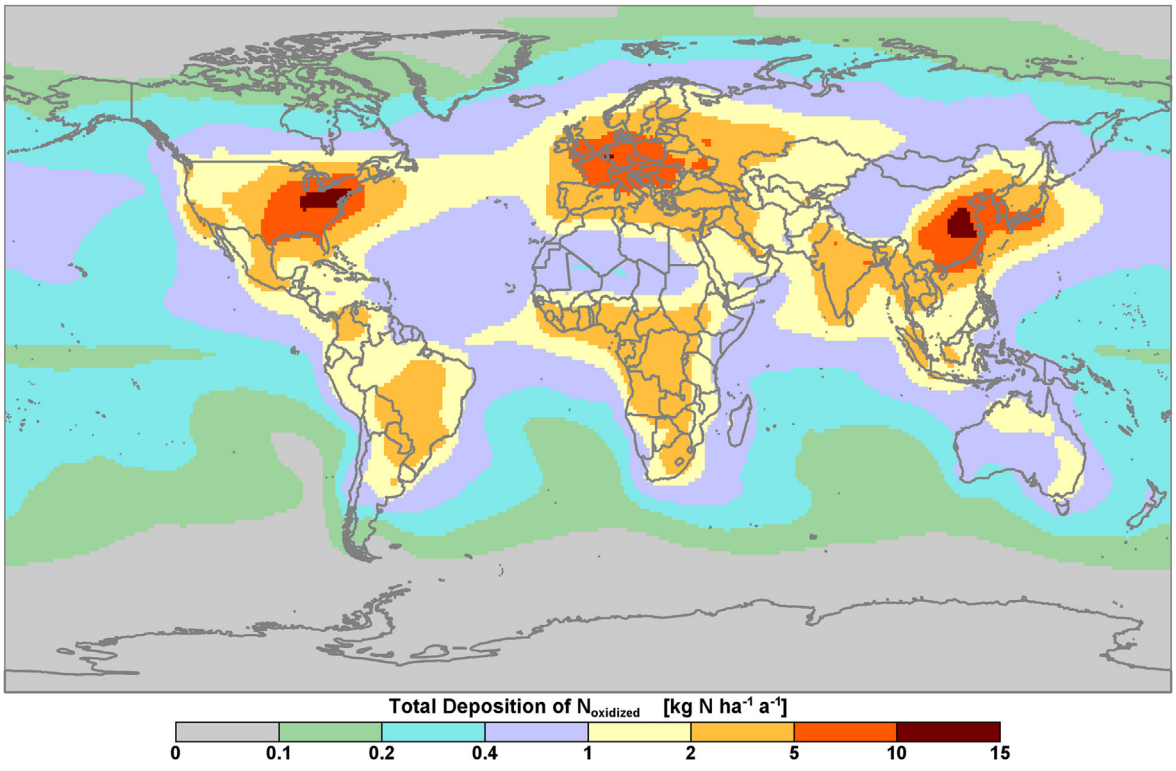

Fig. S17. 2001 ensemble-mean pattern of total deposition of $N_{oxidized}$ in kg N ha^{-1} a^{-1}.

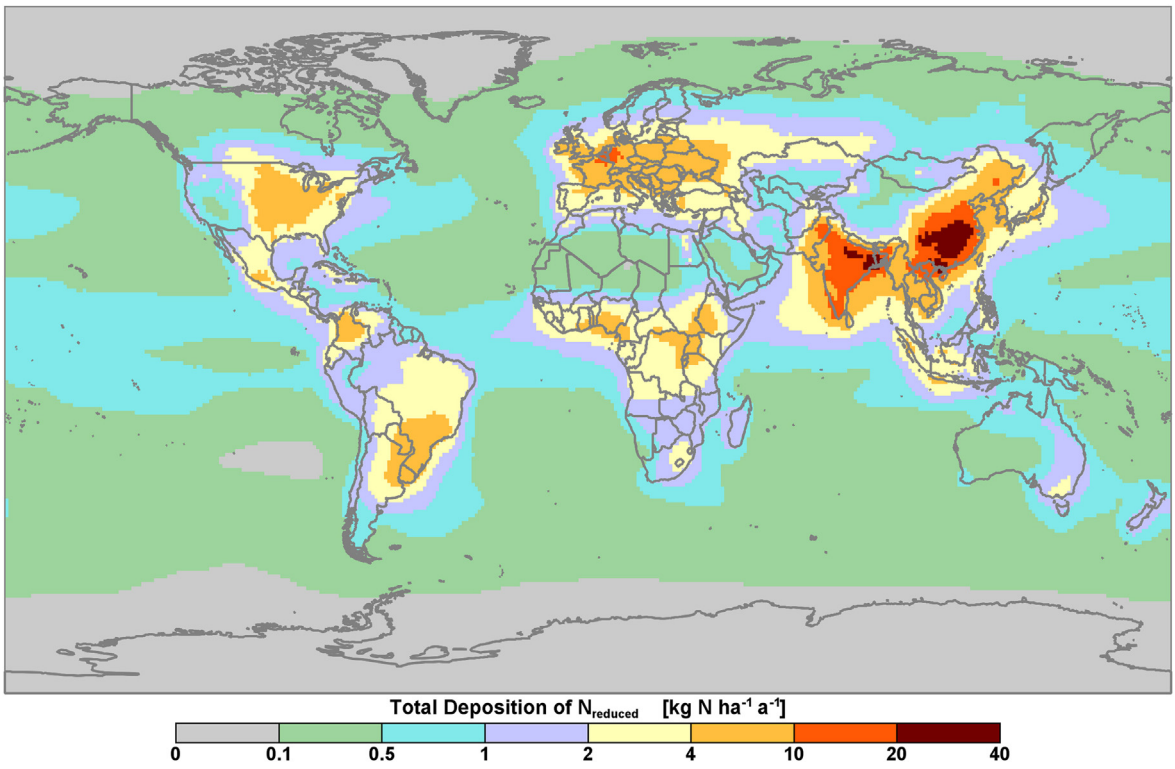

Fig. S18. 2001 ensemble-mean pattern of total deposition of $N_{reduced}$ in kg N ha^{-1} a^{-1}.

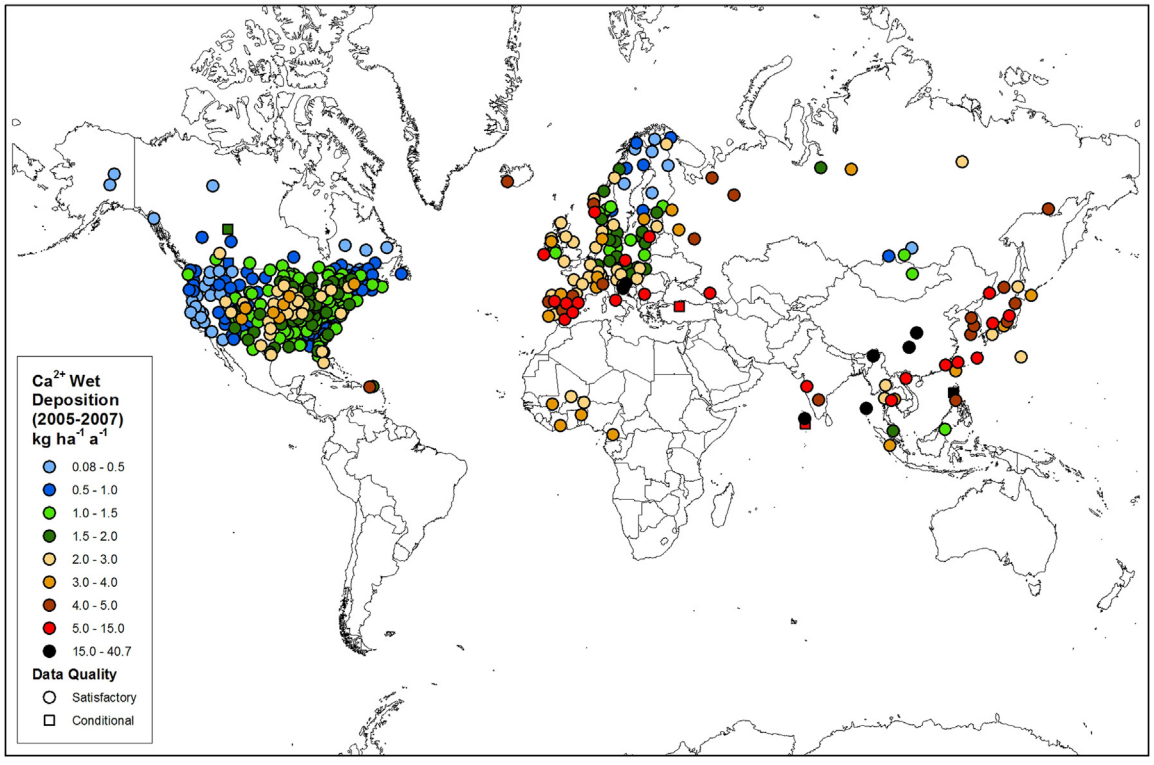

Fig. S19. Global 3-year mean annual wet deposition of Ca^{2+} in kg ha^{-1} a^{-1} for 2005–2007.

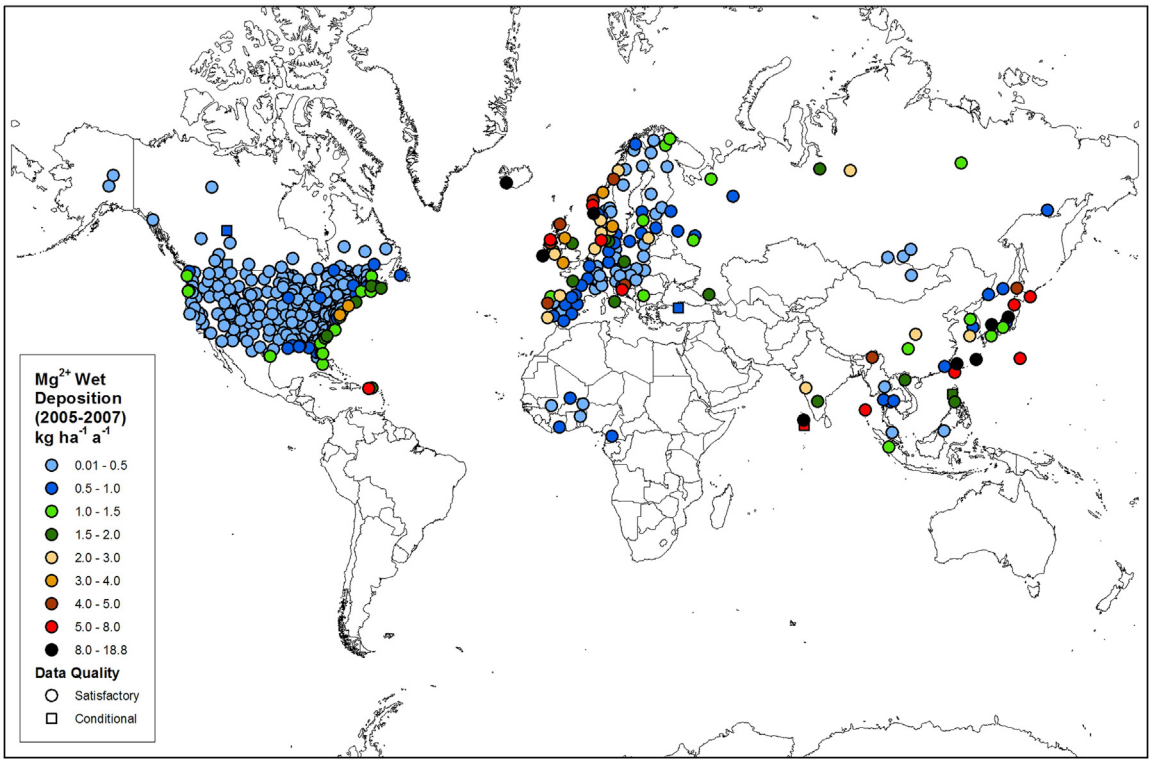

Fig. S20. Global 3-year mean annual wet deposition of Mg^{2+} in kg ha^{-1} a^{-1} for 2005–2007.

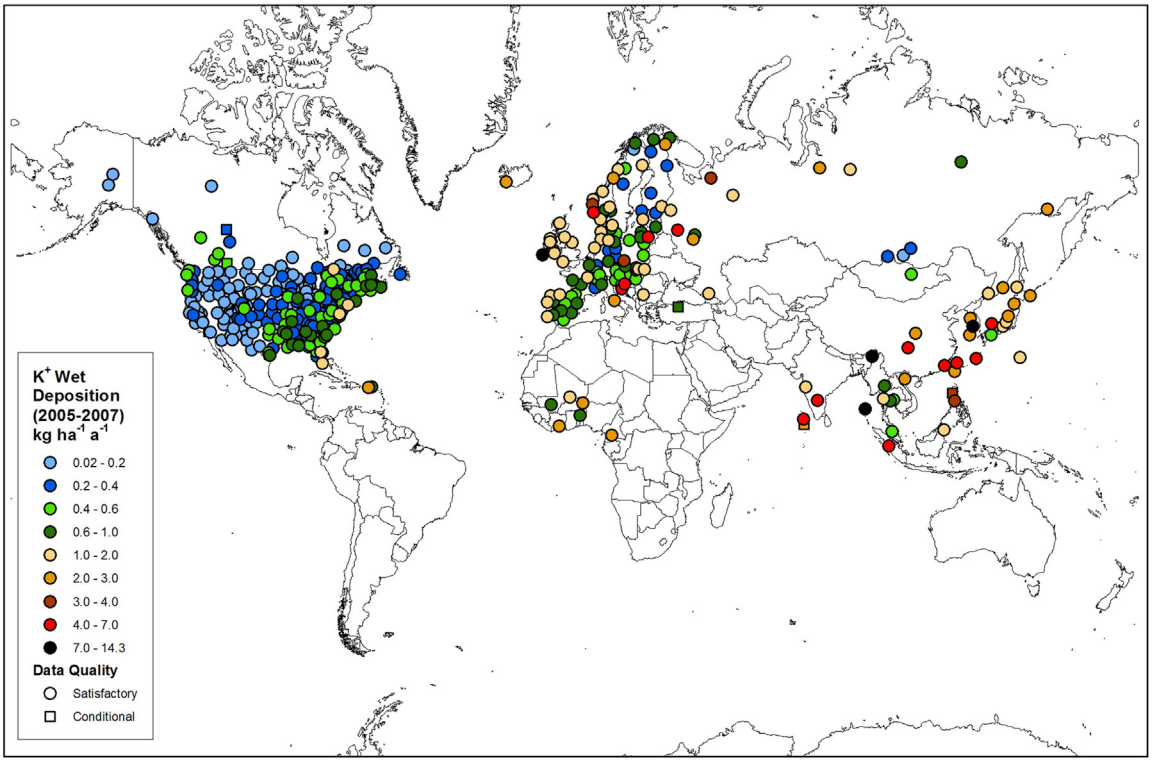

Fig. S21. Global 3-year mean annual wet deposition of K^{+} in kg ha^{-1} a^{-1} for 2005–2007.

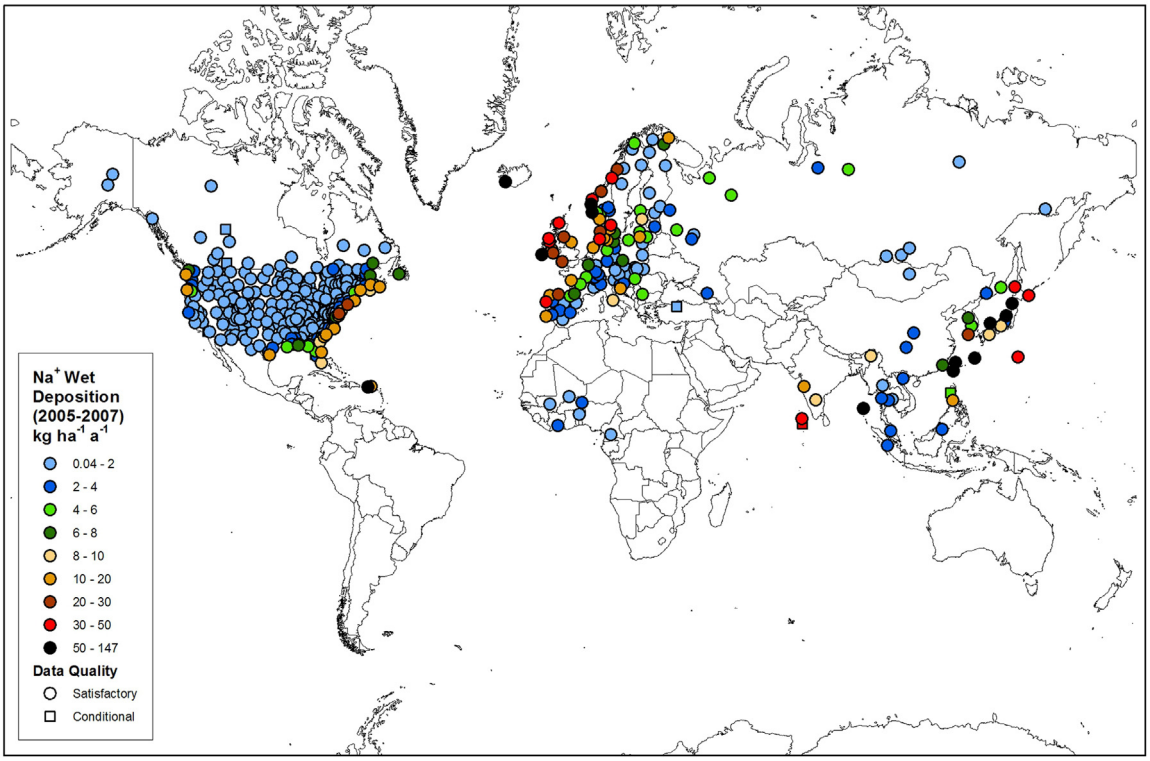

Fig. S22. Global 3-year mean annual wet deposition of Na^+ in kg ha^{-1} a^{-1} for 2005–2007.

Table S1
Network and other sources of wet and dry deposition data used in this assessment, including the numbers of sites reported by each and web-based links and/or citations. Data from Global Atmosphere Watch (GAW) stations are dealt with under the regional and national measurement programs.

Network	Network name	Reference/link
Wet deposition (533 sites)		
Africa (9 sites)		
IDAF (9)	International Global Atmospheric Chemistry/Deposition of Biogeochemically Important Trace Species/Africa	http://idaf.sedoo.fr
Asia (49 sites)		
EANET (30)	Acid Deposition Monitoring Network in East Asia	http://www.eanet.cc/jpn/
IND (4)	WMO/GAW Network of the India Meteorological Department	http://www.imdpune.gov.in/research/reinfo_index.html
ABC (3)	Atmospheric Brown Cloud Network	http://www.rrcap.unep.org/abc/
IMPACTS (4)	Integrated Monitoring Programme on Acidification of Chinese Terrestrial Systems	Larssen et al., 2006
Rus PCCnet (5)	Russian Federation Precipitation Chemistry Composition Network	Ryaboshapko and Gromov, 1999
CAD (1)	Composition of Asian Deposition	http://www.sei-international.org/rapidc/networks-cad.htm
TADN (2)	Taiwan Acid Deposition Network	http://www.epa.gov.tw/en/index.aspx
Australia (2 sites)		
AUST (2)	Australia Regional GAW Precipitation Chemistry Network	Gillett et al., 2007
Europe (126 sites)		
EMEP (107)	European Monitoring and Evaluation Programm	http://www.emep.int/; http://ebas.nilu.no/
NO (12)	Norwegian Monitoring Programme on Atmospheric Deposition	http://ebas.nilu.no/
Rus PCCnet (4)	Russian Federation Precipitation Chemistry Composition Network	Ryaboshapko and Gromov, 1999
SE (3)	Swedish National Programme on Precipitation Chemistry (PMK)	http://www.ivl.se/
South America (1 site)		
CCST/INPE (1)	Environmental Biogeochemistry Research Laboratory	http://www.ccst.inpe.br/
North America (346 sites)		
NADP/NTN (259)	United States National Atmospheric Deposition Program/National Trends Network	http://nadp.sws.uiuc.edu/ntn/
REPQ (33)	Réseau d'échantillonnage des précipitations du Québec	http://www.ec.gc.ca/natchem/
CAPMoN (25)	Canadian Air and Precipitation Monitoring Network	http://www.ec.gc.ca/rs-mn/default.asp?lang=En&n%20=752CE271-1
NBPN (10)	New Brunswick Precipitation Monitoring Network	http://www.ec.gc.ca/natchem/
ABPM (6)	Precipitation Quality Monitoring Program in Alberta	http://www.ec.gc.ca/natchem/
NADP/AIRM (4)	NADP/Atmospheric Integrated Research Monitoring Network	http://nadp.sws.uiuc.edu/airmon/
BCPM (4)	British Columbia Precipitation Chemistry Sampling Network	http://www.ec.gc.ca/natchem/
NFPM (4)	Newfoundland Acid Precipitation Monitoring Network	http://www.ec.gc.ca/natchem/

(*continued on next page*)

Table S1 (*continued*)

Network	Network name	Reference/link
NSPM (1)	Nova Scotia Precipitation Study Network	http://www.ec.gc.ca/natchem/
Dry deposition (100 sites)		
Africa (7 sites)		
IDAF (7)	International Global Atmospheric Chemistry/Deposition of Biogeochemically Important Trace Species/Africa	http://idaf.sedoo.fr
Australia (1 site)		
BPAPS (1)	Burrup Peninsula Air Pollution Study	Gillett et al., 2012
North America (92 sites)		
CASTNET (80)	United States Clean Air Status and Trends Network	http://java.epa.gov/castnet/
CAPMoN (12)	Canadian Air and Precipitation Monitoring Network	http://www.ec.gc.ca/rs-mn/default.asp?lang=En&n%20=752CE271-1

Table S2
Summary of HTAP models and contacts used in each of the ensemble-mean model calculations of deposition, concentration and precipitation depth. X indicates that a model was used in the ensemble-mean, O indicates that a model was not used and a blank indicates that no modeled values were available. Additional model information may be found in HTAP (2010) available at www.htap.org.

Model	Contact	Sulfur	Oxidized Nitrogen	Reduced Nitrogen	Precipitation Depth	Sea Salt
CAMCHEM-3311m13	Peter Hess	X	X	X	X	X
CAMCHEM-3514	Peter Hess	X	X	O		X
CHASER-v03	Kengo Sudo	X	O		X	
ECHAM-HAMMOZ-v21	Gerd Folbert, Isabelle Bey	O	O		X	O
EMEP-rv26 (NH)	Jan-Eiof Jonson	X	X	X	X	
FRSGCUCI-v01	Oliver Wild		X		X	
GEMAQ-EC	Alex Lupu	X	O		O	O
GEMAQ-v1p0	Alex Lupu	O	X		O	
GEOSChem-v07	Rokjin Park	X	X	X	X	X
GISS-PUCCINI-ModelEaer	Drew Shindell	X	O	X	X	O
GISS-PUCCINI-modelA	Drew Shindell	X	X		X	
GISS-PUCCINI-modelE	Drew Shindell	X	X		X	
GMI-v02a	Bryan Duncan	X			X	X
GMI-v02f	Bryan Duncan		X			
GOCART-v4p1	Mian Chin	X			X	X
GOCART-v4p2	Mian Chin	X			X	X
HADGEM2-A-v01	Shekar Reddy, Bill Collins	O			O	
INCA-v2MS	Sophie Szopa, Didier Hauglustaine	X				O
INCA-vSSz	S. Szopa, D. Hauglustaine, Michael Schultz		X		O	
LLNL-IMPACT-T5a	Cindy Atherthon, Dan Bergmann	X	O		O	
MOZARTGFDL-v2	Arlene Fiore	X	X	X	X	
MOZECH-v16	Martin Schultz		O		X	
SPRINTARS-v356	Toshiko Takemura	X			O	X
STOC-HadAM3-v01	Ian Mackenzie, David Stevenson	X	X	X	X	
STOCHEM-v02	Michael Sanderson, Bill Collins	X	X	O	O	
TM5-JRC-cy2-ipcc-v1	Frank Dentener	X	X	X		X
ULAQ-v02	Veronika Montanaro	O	X		O	O
UM-CAM-v01	Guang Zeng		X		X	

Table S3
Measurement-model comparability statistics. Mean and Std Dev (Standard Deviation) Δ refer to all measurement minus model differences of the "n" grid squares in the regions indicated. The % Relative Error represents the Mean Δ divided by the mean of the observations. *R* is the correlation coefficient.

Model species	Region	N	Mean Δ	StDv Δ	Error (%)	*R*
Precipitation depth [cm a^{-1}]	Global	429	5.4	39.6	5.65	0.648
	Africa	7	−31.1	9.3	−36.13	0.986
	Asia	39	−6.4	58.6	−4.76	0.705
	Australia	2	25.3	24.8	24.81	
	Europe	106	20.4	43.0	21.24	0.708
	N. America	273	1.9	33.0	2.15	0.668
	S. America	2	20.5	64.6	12.79	
Concentration wet $N_{oxidized}$ [mg N L^{-1}]	Global	427	0.01	0.10	5.85	0.687
	Africa	7	0.05	0.08	29.18	0.622
	Asia	39	0.07	0.14	28.86	0.435
	Australia	2	0.03	0.02	27.31	
	Europe	105	0.02	0.12	6.13	0.724
	N. America	273	0.00	0.09	1.97	0.676
	S. America	1	0.04		28.52	
Concentration wet $N_{reduced}$ [mg N L^{-1}]	Global	426	0.03	0.15	11.64	0.681
	Africa	7	0.08	0.07	36.93	0.200
	Asia	38	0.04	0.24	11.17	0.670
	Australia	2	0.06	0.02	30.27	
	Europe	105	0.03	0.21	6.83	0.537
	N. America	273	0.03	0.10	13.92	0.702
	S. America	1	0.20		52.29	
Concentration wet N [mg N L^{-1}]	Global	426	0.04	0.22	8.59	0.722

Table S3 (*continued*)

Model species	Region	N	Mean Δ	StDv Δ	Error (%)	*R*
	Africa	7	0.13	0.14	33.72	0.261
	Asia	38	0.10	0.28	16.16	0.743
	Australia	2	0.09	0.04	29.25	
	Europe	105	0.05	0.29	6.51	0.669
	N. America	273	0.03	0.17	7.67	0.664
	S. America	1	0.23		46.40	
Concentration wet nssS [mg S L^{-1}]	Global	428	−0.03	0.23	−8.42	0.678
	Africa	7	0.15	0.22	54.52	0.986
	Asia	39	0.22	0.46	33.81	0.742
	Australia	2	0.02	0.04	12.98	
	Europe	106	−0.12	0.28	−25.46	0.586
	N. America	273	−0.04	0.09	−12.26	0.886
	S. America	1	0.00		−1.45	
Deposition wet $N_{oxidized}$ [kg N ha^{-1} a^{-1}]	Global	427	0.22	1.44	9.52	0.587
	Africa	7	−0.11	0.54	−9.42	0.604
	Asia	39	1.25	3.43	38.46	0.286
	Australia	2	0.48	0.46	48.21	
	Europe	105	0.63	1.52	21.82	0.635
	N. America	273	−0.08	0.64	−4.26	0.858
	S. America	1	1.56		55.40	
Deposition wet $N_{reduced}$ [kg N ha^{-1} a^{-1}]	Global	426	0.18	1.46	7.76	0.706
	Africa	7	0.14	0.63	7.52	0.785
	Asia	38	−0.40	3.13	−10.15	0.690
	Australia	2	0.91	0.72	49.33	
	Europe	105	0.73	1.93	22.09	0.555
	N. America	273	0.02	0.54	1.22	0.851
	S. America	1	5.28		67.42	
Deposition wet $N_{reduced}$/wet N [%]	Global	422	1.18	10.62	2.42	0.543
	Africa	3	5.56	7.30	8.63	0.931
	Asia	38	−3.29	21.33	−5.63	−0.077
	Australia	2	1.14	0.39	1.74	
	Europe	105	1.44	10.48	2.74	0.198
	N. America	273	1.63	8.18	3.58	0.641
	S. America	1	6.54		8.90	
Deposition wet sea salt [kg ha^{-1} a^{-1}]	Global	170	23.92	68.83	53.56	0.514
	Africa	4	1.75	0.82	42.16	0.981
	Asia	25	78.38	139.95	80.32	0.607
	Australia	1	99.56		77.91	
	Europe	61	28.84	55.87	49.10	0.737
	N. America	78	2.95	18.24	16.37	0.493
	S. America	1	11.34		57.74	
Deposition wet N [kg N ha^{-1} a^{-1}]	Global	426	0.36	2.28	7.90	0.732
	Africa	7	0.03	1.06	0.83	0.753
	Asia	38	0.47	4.14	6.89	0.703
	Australia	2	1.39	1.18	48.94	
	Europe	105	1.36	3.17	21.96	0.609
	N. America	273	−0.06	1.09	−1.78	0.861
	S. America	1	6.84		64.23	
Deposition wet nssS [kg S ha^{-1} a^{-1}]	Global	428	−0.10	2.03	−2.82	0.793
	Africa	7	0.57	1.10	29.03	0.938
	Asia	39	2.25	4.50	29.85	0.838
	Australia	2	0.51	0.68	37.58	
	Europe	106	−0.39	2.11	−10.22	0.579
	N. America	273	−0.35	0.98	−11.18	0.900
	S. America	1	0.83		30.40	

Table S4
Bulk deposition measurements of TP, PO_4^{3-} and TDP (g P ha^{-1} a^{-1}).

Country	Site name	Latitude (°N)	Longitude (°E/W)	TP unfiltered	PO_4^{3-} unfiltered	PO_4^{3-} filtered	TDP filtered	Period	Preservation technique	Source/method
Monthly sampling periods — Atmospheric Deposition and Impact on the Open Mediterranean Sea (ADIOS)										
Cyprus	Cavo Greco	34.9500	34.0833	189				06/2001–05/2002	10% HNO3	1A
						145			Thymol	2A
France	Cap Bear	43.5167	3.1500	376				06/2001–05/2002	10% HNO3	1A
						192		06/2001–05/2003	Thymol	2A
France	Corsica-Ostriconi	42.6667	9.0667	418				06/2001–05/2002	10% HNO3	1A
						139		06/2001–05/2003	Thymol	2A
Greece	Finokalia	35.3333	25.6667	125				06/2001–05/2002	10% HNO3	1A
						88		06/2001–05/2003	Thymol	2A
Greece	Lesbos-Mytilene	39.0333	26.6000	142				06/2001–05/2002	10% HNO3	1A

(*continued on next page*)

Table S4 (*continued*)

Country	Site name	Latitude (°N)	Longitude (°E/W)	TP unfiltered	PO_4^{3-} unfiltered	PO_4^{3-} filtered	TDP filtered	Period	Preservation technique	Source/method
						98			Thymol	2A
Malta	Gozo	36.0667	14.2167	110				03/2002–02/2003	Thymol	2A
Morocco	Cap Spartel	35.7833	−5.9000	332				06/2001–05/2002	10% HNO3	1A
						188			Thymol	2A
Tunisia	Mahdia	35.4167	11.0333	472				06/2001–05/2002	10% HNO3	1A
						115			Thymol	2A
Turkey	Akkuyu	36.1333	33.5333	323				06/2001–05/2002	10% HNO3	1A
						149			Thymol	2A
Variable sampling periods (<31 days)										
Canada	Harp Lake	45.3839	−79.1441					2003–2007	None	3A
Canada	Heney Lake	45.1817	−78.8246	142				2003–2007	None	3A
Canada	Plastic Lake	45.1305	−79.0979	149				2003–2007	None	3A
Weekly sampling periods										
Canada	Turkey Lakes	47.0337	−84.3790	59				2000–2005	None	4B
Event sampling periods										
Israel	Tel Shikmona	32.8194	34.9556		90			1992–3/1998	Frozen post collection	5C
Japan	Ashiu Exp. Forest	35.3333	135.7167			54	78	11/2000–11/2003	Frozen post collection	6A

Source
1. Guieu et al., 2010
2. Markaki et al., 2010
3. Yao et al., 2010
4. Semkin, R., 2010
5. Herut et al., 1999
6. Tsukuda et al., 2005

Method
A. Funnel-bottle with debris screen
B. Funnel-bag with debris screen
C. Funnel-bottle