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Long-term declines in atmospheric nitrogen and sulfur deposition reduce critical loads exceedances at multiple Canadian rural sites, 2000–2018

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S1 Daily average dry deposition velocities (V_d) of N and S species

S1.1 Spatial patterns

Daily average V_d for gas-phase compounds (SO_2, HNO_3) and particulate-phase compounds (pSO_4^{2-}, pNH_4^+ and pNO_3^-) for the 2000-2018 period are summarized in Table S2. For gaseous compounds, the mean daily V_d (cm/s) among 15 CAPMoN sites were 1.2 for HNO_3 and 0.46 for SO_2. For particulate sulfate (pSO_4^{2-}), ammonium (pNH_4^+) and nitrate (pNO_3^-), the mean daily V_d were 0.16, 0.15 and 0.21 cm/s, respectively. V_d of N and S compounds exhibited strong variability between sites. The regions with higher V_d for N and S compounds include the west coast, southeast and Atlantic (Table S2). According to the land use data surrounding a CAPMoN site (Table S1), the west coast and Atlantic sites have higher V_d likely because of nearby land use coverage that is associated with higher V_d. For example, water surfaces and forests. In particular, evergreen needleleaf or broadleaf trees are typically associated with larger leaf area index (LAI) and hence larger V_d. Meteorological conditions also differ substantially across Canadian sites, which can drive the spatial variability in V_d.

S1.2 Cold vs. warm seasonal patterns

V_d of gaseous N and S compounds were slightly greater in the warm season than cold season at most of the sites based on the monthly variations in Fig. S1. This was also the case for particulate nitrate due to the higher fraction of nitrate in coarse PM during the warm season, which is based on size-fractionated measurements previously conducted at CAPMoN sites (Zhang et al., 2008). Given that V_d of coarse PM ($PM_{2.5-10}$) is larger than that of fine PM ($PM_{2.5}$), a higher fraction in coarse PM results in higher V_d. During the cold season, nitrate is predominantly associated with fine PM at CAPMoN sites (Zhang et al., 2008). V_d of sulfate and ammonium were slightly higher in the cold season than warm season. This pattern is likely attributed to meteorology perhaps higher wind speeds in the cold season.

S1.3 Long-term annual trends

Long-term annual trends in V_d were estimated using Theil-Sen slopes of the seasonal average V_d, which have been seasonally adjusted using LOESS (locally estimated scatterplot smoothing). Statistically significant trends in V_d ($p<0.05$) are shown in Fig. S2. Among the sites, annual trends in V_d (cm/s of change per year) ranged from 2.4-8.8 x 10^{-3} for HNO_3, 9.0 x 10^{-4} to 5.1 x 10^{-3} for SO_2, 3.0 x 10^{-4} to 1.0 x 10^{-3} for sulfate, 3.0-9.0 x 10^{-4} for ammonium and 2.7 x 10^{-4} to 1.5 x 10^{-3} for nitrate. Overall, the increasing annual trends in V_d were very small indicating that V_d of a chemical specie stays constant over time. Small fluctuations in V_d over time reflect meteorological variability. Thus, the main factor driving the long-term annual dry deposition trends are the ambient air concentrations.

S2 Soil critical loads (CL) calculations

Soil CL were extracted from Canada-wide maps produced following the manual on methodologies and criteria for modelling and mapping critical loads (CLRTAP, 2004), which outlines the input parameters required for the Simple Mass Balance (SMB) model. A raster-based mapping approach was taken with maps resampled and aligned to the 250 m LandGIS project grid and reprojected (where necessary) to EPSG:3347. The critical load function (Figure S9) was calculated for each map using $CL_{max}S$ (Eq. S1), $CL_{min}N$ (Eq. S3), and $CL_{max}N$ (Eq. S4).

$$CL_{max}S = BC_{dep} + BC_w - Cl_{dep} - Bc_u - ANC_{le,crit} \quad \text{(Eq. S1)}$$

where $ANC_{le,crit} = -Q^{2/3} \cdot \left(1.5 \cdot \frac{Bc_{dep} + Bc_w - Bc_u}{K_{gibb} \cdot (Bc/Al)_{crit}}\right)$ (Eq. S2)

$$CL_{min}N = N_i + N_u \quad \text{(Eq. S3)}$$

$$CL_{max}N = CL_{min}N + \left(\frac{CL_{max}S}{(1-f_{de}}\right) \quad \text{(Eq. S4)}$$

Base cation deposition (BC_{dep}, Eq. S1) was mapped for the country using a nationwide crustal materials map and a BC_{dep} map covering the Athabasca oil sands region scaled to CAPMoN precipitation ions data (Makar et al., 2018). The BC_{dep} map was divided by long-term normal (1980-2010) annual precipitation (McKenney et al., 2006) to model concentration. Logistic regression kriging using crustal material and long-term normal (1980-2010) annual temperature (McKenney et al., 2006) was used to create a predicted regional map based on the local base cation estimates in the Athabasca oil sands region. Since this process estimates non-marine BC_{dep}, chloride deposition (Cl_{dep}) was not considered. The base cation (Ca + Mg + K + Na) weathering rate (BC_w) was estimated according to the soil type-texture approximation using continuous soil characteristics maps (absolute depth (Hengl, 2017), bulk density (Hengl, 2018c), clay content (Hengl, 2018d), sand content (Hengl, 2018b), and organic carbon (Hengl & Wheeler, 2018)) from the LandGIS project and parent material acid classification from the Soil Landscapes of Canada (SLC) v2.2 (Centre for Land and Biological Resources Research & Canada, 1996) and v.3.3 (Schut et al., 2011). Root zone was limited to a maximum of 50 cm for forests and 30 cm for other landcover types; wetlands and soils that contained >30% organic matter were removed. Soil weathering rate was temperature adjusted (Hengl, 2018a). The gibbsite equilibrium constant (K_{gibb}, Eq. S2) was assigned mid-point values from ranges suggested by CLRTAP (2004) (950 m^6 eq^{-2} for mineral soils; 300 m^6 eq^{-2} for low organic matter soils; 100 m^6 eq^{-2} for soils with some organic material; 9.5 m^6 eq^{-2} for peaty and organic soils). Base cation uptake (Bc_u) and nitrogen uptake (N_u) by trees were estimated using species-specific base cation and nutrient content databases (Pardo et al., 2005; Paré et al., 2013) for bark and trunk, in combination with species and biomass maps (Beaudoin et al., 2014). Uptake maps were limited to the harvestable areas delineated in Dymond et al. (2010). A dynamic Bc/Al_{crit} (critical chemical criteria) was assigned to each raster grid using species-specific values representing a 5% growth reduction from Sverdrup and Warfvinge (1993) where available; priority was given to the species with the lowest Bc/Al_{crit}. Where species-specific information was not available or non-forested soils occurred, a value averaged to the genus or landcover type (CCRS, 2018) was used. A runoff (Q) map was interpolated from modelled point estimates produced from the meteo-hydrological model MetHyd (Bonten et al., 2016) (detailed in Reinds et al., 2015). Landcover was used to mask out wetlands, urban areas and agricultural soils. The long-term net immobilization of N (N_i, Eq. S3) was set to 35.714 eq ha^{-1} yr^{-1} (based on assumed 0.5 ha N^{-1} yr^{-1}). The combined SLC databases were used to assign the denitrification fraction (f_{de}, Eq. S4) based on drainage classes (CLRTAP, 2004).

Table S1: Description of CAPMoN sites analyzed in this study. Land use percentages within a 3 km circle of each site were estimated from MODIS.

Site (ID)	Coordinates	Province	Land use percentages	Period analyzed
Saturna (SAT)	48.78, -123.13	British Columbia (BC)	evergreen needleleaf trees (59.1%), water (27.0%), evergreen broadleaf trees (13.9%)	2000-2018
Bratt's Lake (BRA)	50.2, -104.71	Saskatchewan (SK)	crops (100%)	2002-2012
Experimental Lakes Area (ELA)	49.66, -93.73	Ontario (ON)	long grass (52.4%), evergreen needleleaf trees (47.6%)	2000-2018
Algoma (ALG)	47.03, -84.38	ON	deciduous broadleaf trees (100%)	2000-2016
Bonner Lake (BON)	49.39, -82.12	ON	deciduous broadleaf trees (35.2%), evergreen needleleaf trees (34.4%), long grass (30.3%)	2009-2018
Longwoods (LON)	42.88, -81.48	ON	crops (100%)	2000-2018
Egbert (EGB)	44.23, -79.79	ON	crops (80.2%), long grass (19.8%)	2000-2018
Sprucedale (SPR)	45.42, -79.49	ON	deciduous broadleaf trees (52%), evergreen needleleaf trees (48%)	2003-2018
Chalk River (CHA)	46.06, -77.40	ON	deciduous broadleaf trees (60.7%), evergreen needleleaf trees (39.3%)	2000-2018
Chapais (CPS)	49.82, -74.98	Quebec (QC)	long grass (64.4%), evergreen needleleaf trees (35.6%)	2000-2017
Frelighsburg (FRE)	45.05, -72.86	QC	deciduous broadleaf trees (63.9%), long grass (36.1%)	2002-2016
Lac Edouard (LED)	47.68, -72.44	QC	evergreen needleleaf trees (50.4%), deciduous broadleaf trees (49.6%)	2002-2016
Kejimkujik National Park (KEJ)	44.43, -65.20	Nova Scotia (NS)	evergreen needleleaf trees (51.2%), deciduous broadleaf trees (48.8%)	2000-2016
Mingan (MIN)	50.27, -64.23	QC	water (39.8%), evergreen needleleaf trees (37.9%), long grass (22.3%)	2009-2016
Bay d'Espoir (BAB)	47.99, -55.82	Newfoundland and Labrador (NL)	long grass (56.1%), evergreen needleleaf trees (24.4%), deciduous broadleaf trees (19.5%)	2007-2016

Table S2: Modeled dry deposition velocities of N and S compounds at CAPMoN sites during 2000-2018 (mean ± standard deviation, cm/s)

SiteID	Region	SO_2	HNO_3	pSO_4^{2-}	pNH_4^+	pNO_3^-
SAT	West coast	0.82 ± 0.44	1.72 ± 0.69	0.19 ± 0.06	0.18 ± 0.06	0.25 ± 0.05
BRA	Prairie	0.47 ± 0.18	1.04 ± 0.32	0.2 ± 0.03	0.19 ± 0.03	0.27 ± 0.08
ELA	Remote	0.37 ± 0.12	0.97 ± 0.2	0.13 ± 0.02	0.12 ± 0.02	0.17 ± 0.03
ALG	Greater Southeast	0.28 ± 0.12	0.9 ± 0.25	0.11 ± 0.02	0.1 ± 0.02	0.16 ± 0.03
BON	Remote	0.36 ± 0.15	1.02 ± 0.26	0.13 ± 0.02	0.12 ± 0.02	0.17 ± 0.04
LON	Southeast	0.57 ± 0.11	1.2 ± 0.16	0.23 ± 0.04	0.22 ± 0.03	0.3 ± 0.05
EGB	Southeast	0.5 ± 0.11	1.04 ± 0.16	0.2 ± 0.03	0.19 ± 0.03	0.26 ± 0.05
SPR	Greater Southeast	0.41 ± 0.14	1.26 ± 0.26	0.13 ± 0.02	0.12 ± 0.02	0.17 ± 0.03
CHA	Greater Southeast	0.31 ± 0.11	0.94 ± 0.21	0.11 ± 0.01	0.1 ± 0.01	0.15 ± 0.03
CPS	Remote	0.4 ± 0.15	1.01 ± 0.23	0.14 ± 0.02	0.14 ± 0.02	0.19 ± 0.04
FRE	Greater Southeast	0.3 ± 0.1	0.78 ± 0.18	0.11 ± 0.01	0.1 ± 0.01	0.15 ± 0.03
LED	Remote	0.38 ± 0.17	1.14 ± 0.31	0.12 ± 0.02	0.12 ± 0.02	0.17 ± 0.04
KEJ	Atlantic	0.5 ± 0.12	1.46 ± 0.28	0.15 ± 0.03	0.14 ± 0.03	0.19 ± 0.02
MIN	Atlantic	0.58 ± 0.16	1.26 ± 0.2	0.21 ± 0.02	0.2 ± 0.02	0.29 ± 0.06
BAB	Atlantic	0.76 ± 0.21	1.92 ± 0.24	0.27 ± 0.04	0.26 ± 0.03	0.36 ± 0.05

Table S3: Mean and range of annual dry deposition fluxes of N and S (kg N or S/ha/yr) at CAPMoN sites

Site ID	Period	∑N	∑S	HNO_3	pNO_3^-	pNH_4^+	SO_2	pSO_4^{2-}
SAT	2000-2018	0.74	1.38	0.55	0.10	0.08	1.28	0.11
		0.53 - 1.16	0.39 - 2.56	0.39 - 0.89	0.08 - 0.14	0.05 - 0.13	0.32 - 2.39	0.09 - 0.21
BRA	2002-2012	0.71	0.98	0.34	0.15	0.22	0.76	0.22
		0.54 - 0.9	0.75 - 1.23	0.26 - 0.52	0.11 - 0.17	0.17 - 0.26	0.58 - 0.98	0.18 - 0.25
ELA	2000-2018	0.31	0.28	0.16	0.05	0.10	0.17	0.11
		0.2 - 0.41	0.11 - 0.46	0.1 - 0.23	0.03 - 0.07	0.06 - 0.13	0.05 - 0.32	0.07 - 0.15
ALG	2000-2016	0.47	0.47	0.31	0.05	0.11	0.33	0.14
		0.22 - 0.82	0.13 - 0.91	0.13 - 0.58	0.03 - 0.07	0.06 - 0.18	0.07 - 0.68	0.06 - 0.23
BON	2009-2018	0.14	0.15	0.07	0.01	0.05	0.07	0.08
		0.1 - 0.18	0.08 - 0.24	0.05 - 0.09	0.01 - 0.02	0.03 - 0.07	0.02 - 0.13	0.05 - 0.11
LON	2000-2018	1.88	3.54	0.80	0.41	0.67	2.97	0.57
		1.04 - 2.87	0.74 - 6.92	0.47 - 1.39	0.25 - 0.65	0.32 - 1.05	0.49 - 6.02	0.24 - 0.9
EGB	2000-2018	1.36	2.02	0.62	0.29	0.45	1.62	0.40
		0.77 - 2.08	0.52 - 3.96	0.37 - 0.92	0.18 - 0.45	0.22 - 0.71	0.31 - 3.27	0.17 - 0.68
SPR	2003-2018	0.63	0.78	0.44	0.05	0.14	0.61	0.17
		0.33 - 1.14	0.2 - 1.46	0.22 - 0.81	0.04 - 0.08	0.07 - 0.25	0.13 - 1.2	0.07 - 0.31
CHA	2000-2018	0.35	0.57	0.22	0.03	0.09	0.45	0.13
		0.2 - 0.62	0.13 - 1.05	0.12 - 0.42	0.02 - 0.06	0.05 - 0.16	0.05 - 0.85	0.06 - 0.22
CPS	2000-2017	0.16	0.28	0.09	0.01	0.06	0.17	0.11
		0.08 - 0.25	0.1 - 0.48	0.04 - 0.14	0.01 - 0.02	0.03 - 0.09	0.05 - 0.32	0.06 - 0.15
FRE	2002-2016	0.62	0.74	0.38	0.08	0.17	0.57	0.17
		0.35 - 1.03	0.22 - 1.23	0.24 - 0.67	0.04 - 0.1	0.07 - 0.27	0.15 - 0.95	0.07 - 0.29
LED	2002-2016	0.32	0.36	0.22	0.02	0.08	0.24	0.12
		0.17 - 0.61	0.11 - 0.69	0.11 - 0.43	0.02 - 0.04	0.04 - 0.14	0.05 - 0.5	0.05 - 0.19
KEJ	2000-2016	0.34	0.51	0.21	0.04	0.09	0.34	0.16
		0.16 - 0.65	0.11 - 1.15	0.09 - 0.46	0.03 - 0.06	0.04 - 0.14	0.04 - 0.85	0.08 - 0.32
MIN	2009-2016	0.13	0.20	0.05	0.03	0.05	0.11	0.10
		0.1 - 0.15	0.12 - 0.27	0.04 - 0.06	0.03 - 0.04	0.03 - 0.06	0.05 - 0.15	0.11 - 0.17
BAB	2007-2017	0.18	0.24	0.09	0.04	0.06	0.10	0.14
		0.13 - 0.23	0.12 - 0.36	0.06 - 0.12	0.03 - 0.05	0.04 - 0.07	0.04 - 0.17	0.12 - 0.25

Table S4: Annual percent change in dry, wet and total (dry+wet) N and S deposition at CAPMoN sites estimated from Theil-Sen's trend analysis (% change/yr). ns: trend is not statistically significant; NA: not available due to incomplete data.

Site ID	Period	Dry N	Dry S	Wet N	Wet S	Total N	Total S
SAT	2000-2018	-2.6	-4.2	ns	-3.4	-1.1	-3.5
BRA	2002-2012	ns	ns	NA	NA	NA	NA
ELA	2000-2018	-2.6	-4.3	-1.3	-3.6	-1.4	-3.6
ALG	2000-2016	-3.8	-4.8	-2.4	-4.3	-2.5	-4.4
BON	2009-2018	-4.0	-7.1	-1.4	-3.6	ns	-5.4
LON	2000-2018	-3.9	-5.8	-1.8	-4.3	-2.2	-4.7
EGB	2000-2018	-3.6	-5.4	-1.4	-4.5	-2.0	-4.6
SPR	2003-2018	-4.6	-5.9	-1.9	-4.9	-2.2	-5.0
CHA	2000-2018	-4.0	-5.1	-2.2	-4.5	-2.3	-4.5
CPS	2000-2017	-3.2	-4.5	-3.1	-5.0	-3.3	-4.9
FRE	2002-2016	-4.4	-6.1	-2.1	-5.0	-2.0	-5.3
LED	2002-2016	-4.8	-5.9	-2.2	-4.2	-2.7	-4.7
KEJ	2000-2016	-5.0	-6.6	-2.1	-4.3	-2.6	-4.8
MIN	2009-2016	ns	ns	-2.9	-4.6	ns	-5.6
BAB	2007-2016	-3.7	-6.0	-2.2	-4.2	ns	-6.6

Table S5: Comparison of rates of change between annual dry and wet deposition fluxes of N and S and reduced and oxidized N (kg N or S/ha/yr) at CAPMoN sites based on Theil-Sen slopes (statistically significant at $p<0.05$). NA: not available due to incomplete data; ns: trend is not statistically significant. Note: Nitrogen and reduced N dry deposition exclude dry deposition of NH_3 and some oxidized nitrogen species.

	Reduced N		Oxidized N		Sulfur		Nitrogen		Percentage of total deposition (% increase/yr)		
Site ID	dry dep	wet dep	dry dep	wet dep	dry dep	wet dep	dry dep	wet dep	%wet N	%wet S	%reduced N
SAT	-0.004	ns	-0.02	ns	-0.09	-0.05	-0.02	ns	0.5%	0.8%	0.3%
BRA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
ELA	-0.003	ns	-0.01	-0.05	-0.02	-0.07	-0.01	-0.05	0.1%	0.3%	0.6%
ALG	-0.005	-0.067	-0.02	-0.16	-0.04	-0.25	-0.03	-0.21	0.2%	0.2%	0.8%
BON	-0.003	ns	-0.004	ns	-0.02	-0.07	-0.01	ns	0.3%	0.5%	ns
LON	-0.043	ns	-0.06	-0.15	-0.35	-0.32	-0.11	-0.15	0.6%	1.3%	0.9%
EGB	-0.027	ns	-0.04	-0.11	-0.20	-0.21	-0.07	-0.09	0.5%	0.9%	1.0%
SPR	-0.009	ns	-0.03	-0.14	-0.08	-0.29	-0.04	-0.16	0.3%	0.5%	1.0%
CHA	-0.005	ns	-0.01	-0.11	-0.05	-0.22	-0.02	-0.13	0.2%	ns	0.8%
CPS	-0.002	-0.038	-0.005	-0.08	-0.02	-0.15	-0.01	-0.12	ns	ns	0.6%
FRE	-0.011	ns	-0.03	-0.15	-0.07	-0.31	-0.04	-0.16	0.4%	0.4%	1.3%
LED	-0.004	ns	-0.02	-0.09	-0.03	-0.14	-0.02	-0.10	0.3%	0.5%	1.0%
KEJ	-0.005	ns	-0.02	-0.06	-0.05	-0.14	-0.03	-0.08	0.4%	0.8%	0.6%
MIN	-0.003	ns	ns	ns	ns	-0.10	ns	ns	ns	ns	ns
BAB	-0.003	ns	-0.01	-0.04	-0.02	-0.10	-0.01	-0.05	ns	ns	ns

Table S6: Ratio of % change in oxidized N deposition to % change in NO_x emissions in back trajectory determined source regions. Slope % based on Theil-Sen's analysis of long term trends (% change per year). ns: trend is not significant; NA: not available.

		Emissions →	NO_x Can east	NO_x Can west	NO_x US east	NO_x US west
Site ID	Deposition	Slope %	*-3.80*	*-1.20*	*-3.92*	*-3.24*
SAT	dry	*-2.6*	69%	217%	66%	0.80
BRA	dry	*ns*	NA	NA	NA	NA
ELA	dry	*-2.6*	69%	218%	67%	81%
ALG	dry	*-3.9*	103%	326%	100%	121%
BON	dry	*-3.9*	102%	323%	99%	119%
LON	dry	*-3.6*	96%	303%	93%	112%
EGB	dry	*-3.5*	92%	291%	89%	108%
SPR	dry	*-4.6*	122%	387%	118%	143%
CHA	dry	*-4.1*	107%	338%	103%	125%
CPS	dry	*-3.4*	91%	288%	88%	106%
FRE	dry	*-4.4*	116%	367%	112%	136%
LED	dry	*-5.1*	133%	421%	129%	156%
KEJ	dry	*-5.4*	142%	450%	138%	166%
MIN	dry	*ns*	NA	NA	NA	NA
BAB	dry	*-3.7*	97%	307%	94%	114%
SAT	wet	*ns*	NA	NA	NA	NA
BRA	wet	*ns*	NA	NA	NA	NA
ELA	wet	*-2.5*	67%	213%	65%	79%
ALG	wet	*-3.4*	90%	284%	87%	105%
BON	wet	*ns*	NA	NA	NA	NA
LON	wet	*-3.2*	84%	267%	82%	99%
EGB	wet	*-3.1*	82%	260%	79%	96%
SPR	wet	*-3.4*	89%	283%	87%	105%
CHA	wet	*-3.1*	82%	261%	80%	96%
CPS	wet	*-3.8*	100%	318%	97%	118%
FRE	wet	*-3.7*	96%	305%	93%	113%
LED	wet	*-3.6*	94%	298%	91%	110%
KEJ	wet	*-2.8*	72%	230%	70%	85%
MIN	wet	*ns*	NA	NA	NA	NA
BAB	wet	*ns*	NA	NA	NA	NA
SAT	Total	*-1.4*	36%	113%	35%	42%
BRA	Total	*ns*	NA	NA	NA	NA
ELA	Total	*-2.5*	66%	208%	64%	77%
ALG	Total	*-3.5*	92%	292%	89%	108%
BON	Total	*ns*	NA	NA	NA	NA
LON	Total	*-3.3*	87%	275%	84%	101%
EGB	Total	*-3.2*	85%	270%	83%	100%
SPR	Total	*-3.6*	93%	296%	91%	109%
CHA	Total	*-3.1*	83%	263%	80%	97%
CPS	Total	*-4.0*	105%	334%	102%	123%
FRE	Total	*-3.4*	89%	283%	87%	105%
LED	Total	*-3.8*	101%	320%	98%	118%
KEJ	Total	*-3.3*	87%	275%	84%	102%
MIN	Total	*ns*	NA	NA	NA	NA
BAB	Total	*ns*	NA	NA	NA	NA

Table S7: Ratio of % change in reduced N deposition to % change in NH_3 emissions in back trajectory determined source regions. Slope % based on Theil-Sen's analysis of long term trends (% change per year). ns: trend is not significant; NA: not available.

		Emissions →	NH_3 Can east	NH_3 Can west	NH_3 US east	NH_3 US west
Site ID	Deposition	*Slope %*	*-1.00*	*0.43*	*ns*	*3.40*
SAT	dry	*-3.4*	337%	-787%	NA	-99%
BRA	dry	*ns*	NA	NA	NA	NA
ELA	dry	*-2.7*	266%	-622%	NA	-79%
ALG	dry	*-3.3*	329%	-767%	NA	-97%
BON	dry	*-5.1*	508%	-1186%	NA	-150%
LON	dry	*-4.1*	410%	-957%	NA	-121%
EGB	dry	*-3.9*	387%	-902%	NA	-114%
SPR	dry	*-4.3*	431%	-1006%	NA	-127%
CHA	dry	*-3.7*	372%	-868%	NA	-110%
CPS	dry	*-2.8*	276%	-644%	NA	-81%
FRE	dry	*-4.7*	473%	-1104%	NA	-139%
LED	dry	*-3.8*	380%	-887%	NA	-112%
KEJ	dry	*-4.2*	414%	-967%	NA	-122%
MIN	dry	*-4.9*	491%	-1145%	NA	-145%
BAB	dry	*-4.4*	440%	-1026%	NA	-130%
SAT	wet	*ns*	NA	NA	NA	NA
BRA	wet	*ns*	NA	NA	NA	NA
ELA	wet	*ns*	NA	NA	NA	NA
ALG	wet	*-1.5*	153%	-358%	NA	-45%
BON	wet	*ns*	NA	NA	NA	NA
LON	wet	*ns*	NA	NA	NA	NA
EGB	wet	*ns*	NA	NA	NA	NA
SPR	wet	*ns*	NA	NA	NA	NA
CHA	wet	*ns*	NA	NA	NA	NA
CPS	wet	*-2.3*	225%	-524%	NA	-66%
FRE	wet	*ns*	NA	NA	NA	NA
LED	wet	*ns*	NA	NA	NA	NA
KEJ	wet	*ns*	NA	NA	NA	NA
MIN	wet	*ns*	NA	NA	NA	NA
BAB	wet	*ns*	NA	NA	NA	NA
SAT	Total	*ns*	NA	NA	NA	NA
BRA	Total	*ns*	NA	NA	NA	NA
ELA	Total	*ns*	NA	NA	NA	NA
ALG	Total	*-1.6*	158%	-370%	NA	-47%
BON	Total	*ns*	NA	NA	NA	NA
LON	Total	*ns*	NA	NA	NA	NA
EGB	Total	*ns*	NA	NA	NA	NA
SPR	Total	*ns*	NA	NA	NA	NA
CHA	Total	*-1.3*	132%	-309%	NA	-39%
CPS	Total	*-2.4*	237%	-553%	NA	-70%
FRE	Total	*ns*	NA	NA	NA	NA
LED	Total	*ns*	NA	NA	NA	NA
KEJ	Total	*ns*	NA	NA	NA	NA
MIN	Total	*ns*	NA	NA	NA	NA
BAB	Total	*ns*	NA	NA	NA	NA

Table S8: Ratio of % change in S deposition to % change in SO_2 or SOx emissions in back trajectory determined source regions. Slope % based on Theil-Sen's analysis of long term trends (% change per year). ns: trend is not significant; NA: not available.

		Emissions →	SO_2 US east	SO_2 US west	SOx Can east	SOx Can west
Site ID	Deposition	*Slope %*	*-5.27*	*-4.84*	*-4.95*	*-2.36*
SAT	dry	*-4.2*	79%	87%	85%	177%
BRA	dry	*ns*	NA	NA	NA	NA
ELA	dry	*-4.3*	81%	89%	87%	182%
ALG	dry	*-4.8*	91%	99%	97%	203%
BON	dry	*-7.1*	134%	146%	143%	300%
LON	dry	*-5.8*	111%	121%	118%	247%
EGB	dry	*-5.4*	103%	113%	110%	231%
SPR	dry	*-5.9*	111%	121%	118%	248%
CHA	dry	*-5.1*	97%	106%	104%	218%
CPS	dry	*-4.5*	86%	94%	92%	192%
FRE	dry	*-6.1*	116%	126%	123%	258%
LED	dry	*-5.9*	113%	123%	120%	252%
KEJ	dry	*-6.6*	124%	136%	133%	278%
MIN	dry	*ns*	NA	NA	NA	NA
BAB	dry	*-6.0*	114%	125%	122%	256%
SAT	wet	*-3.4*	64%	69%	68%	142%
BRA	wet	*ns*	NA	NA	NA	NA
ELA	wet	*-3.6*	69%	75%	73%	154%
ALG	wet	*-4.3*	82%	90%	88%	184%
BON	wet	*-4.8*	91%	99%	97%	202%
LON	wet	*-4.3*	81%	89%	87%	181%
EGB	wet	*-4.5*	85%	93%	91%	190%
SPR	wet	*-4.9*	92%	101%	98%	206%
CHA	wet	*-4.5*	84%	92%	90%	189%
CPS	wet	*-5.0*	95%	104%	102%	213%
FRE	wet	*-5.0*	95%	103%	101%	212%
LED	wet	*-4.2*	81%	88%	86%	180%
KEJ	wet	*-4.3*	82%	89%	88%	183%
MIN	wet	*-5.6*	106%	116%	113%	238%
BAB	wet	*-6.1*	115%	126%	123%	257%
SAT	Total	*-3.5*	67%	73%	72%	150%
BRA	Total	*ns*	NA	NA	NA	NA
ELA	Total	*-3.6*	68%	74%	72%	151%
ALG	Total	*-4.4*	84%	91%	89%	187%
BON	Total	*-5.4*	102%	111%	109%	228%
LON	Total	*-4.7*	88%	96%	94%	197%
EGB	Total	*-4.6*	87%	95%	93%	195%
SPR	Total	*-5.0*	96%	104%	102%	214%
CHA	Total	*-4.5*	86%	94%	91%	192%
CPS	Total	*-4.9*	92%	100%	98%	206%
FRE	Total	*-5.3*	100%	110%	107%	224%
LED	Total	*-4.7*	89%	97%	94%	198%
KEJ	Total	*-4.8*	91%	99%	97%	203%
MIN	Total	*-5.6*	107%	117%	114%	239%
BAB	Total	*-6.6*	125%	136%	133%	279%

Table S9: Critical loads (CL) of acidity for a subset of lakes near CAPMoN sites during 2000-2018. Ions are water chemistry mean concentrations. DOC: mean dissolved organic carbon concentration in lakes; Q: mean runoff rate.

SiteID	Number of lakes	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Na^{+} (mg/L)	K^{+} (mg/L)	SO_4^{2-} (mg/L)	Cl^{-} (mg/L)	DOC (mg/L)	Q (m/yr)	CL (eq/ha/yr)
KEJ[1]	8	0.69	0.40	3.15	0.29	1.47	4.52	8.75	1.00	308
ELA[2]	5	2.08	0.62	0.73	0.35	1.94	0.20	6.90	0.25	388
BAB[1]	7	1.35	0.39	2.48	0.34	0.76	3.33	9.27	0.82	576
LED[3]	6	3.64	0.40	0.53	0.19	3.95	0.24	4.05	0.70	1357
ALG[4]	5	3.42	0.38	0.57	0.17	3.78	0.23	6.80	0.85	1460

[1]Maritime Lakes: https://data-donnees.ec.gc.ca/data/substances/monitor/national-long-term-water-quality-monitoring-data/maritime-coastal-basin-long-term-water-quality-monitoring-data/?lang=en

[2]Experimental Lakes Area: https://www.iisd.org/ela/science-data/our-data/data-requests/

[3]Quebec Lakes: data available upon request to Daniel.Houle@ec.gc.ca

[4]Turkey Lakes Watershed Study: https://open.canada.ca/data/en/dataset/f9e1595d-27bb-4a6a-b94d-6b9bed7db263

Table S10: Critical loads (CL) of acidity for soils near CAPMoN sites (eq/ha/yr), during 2000-2018. Source: refer to Section S2 Soil critical loads (CL) calculations.

SiteID	$CL_{max}S$	$CL_{max}N$	$CL_{min}N$
BAB	209	398	74
MIN	371	987	75
CPS	416	579	86
LED	459	578	89
ELA	540	829	85
CHA	593	760	92
ALG	598	998	127
SPR	603	799	101
KEJ	717	928	36
SAT	839	1076	36
BON	909	2589	74
EGB	1287	1719	36
FRE	1744	1314	36
LON	2936	1612	36

Table S11: Soil and lake CL exceedances (eq/ha/yr) near CAPMoN sites after including dry deposition of non-routine N species (NH_3, NO_2 and unknown NO_y). Positive exceedances indicate total acidic deposition exceeded CL, whereas negative exceedances indicate total acidic deposition was below CL. Routine N refers to the exceedance estimated using the total deposition from routinely-monitored N species (dry deposition of HNO_3, pNO_3^- and pNH_4^+; wet deposition of NO_3^- and NH_4^+). Non-routine N species were measured at selected CAPMoN sites during 2002-2005 (Zhang et al., 2009) and at EGB in 2010 (this study). Parameters for deriving soil CL are detailed in Section S2 Soil critical loads (CL) calculations. The sources of the water quality data are available in the Table S9 footnotes.

		Soil		Lake	
SiteID	Year	Non-routine and routine N	Routine N	Non-routine and routine N	Routine N
ALG	2003	178.5	158.6	-412.8	-438.0
CHA	2004	2.7	-58.3	NA	NA
EGB	2002	-203.0	-369.6	NA	NA
EGB	2010	-695.2	-885.5	NA	NA
FRE	2002	-275.0	-353.5	NA	NA
KEJ	2002	-175.8	-223.2	320.9	297.1
LED	2003	-1.6	-32.0	-795.3	-826.7
SPR	2004	230.0	205.8	NA	NA

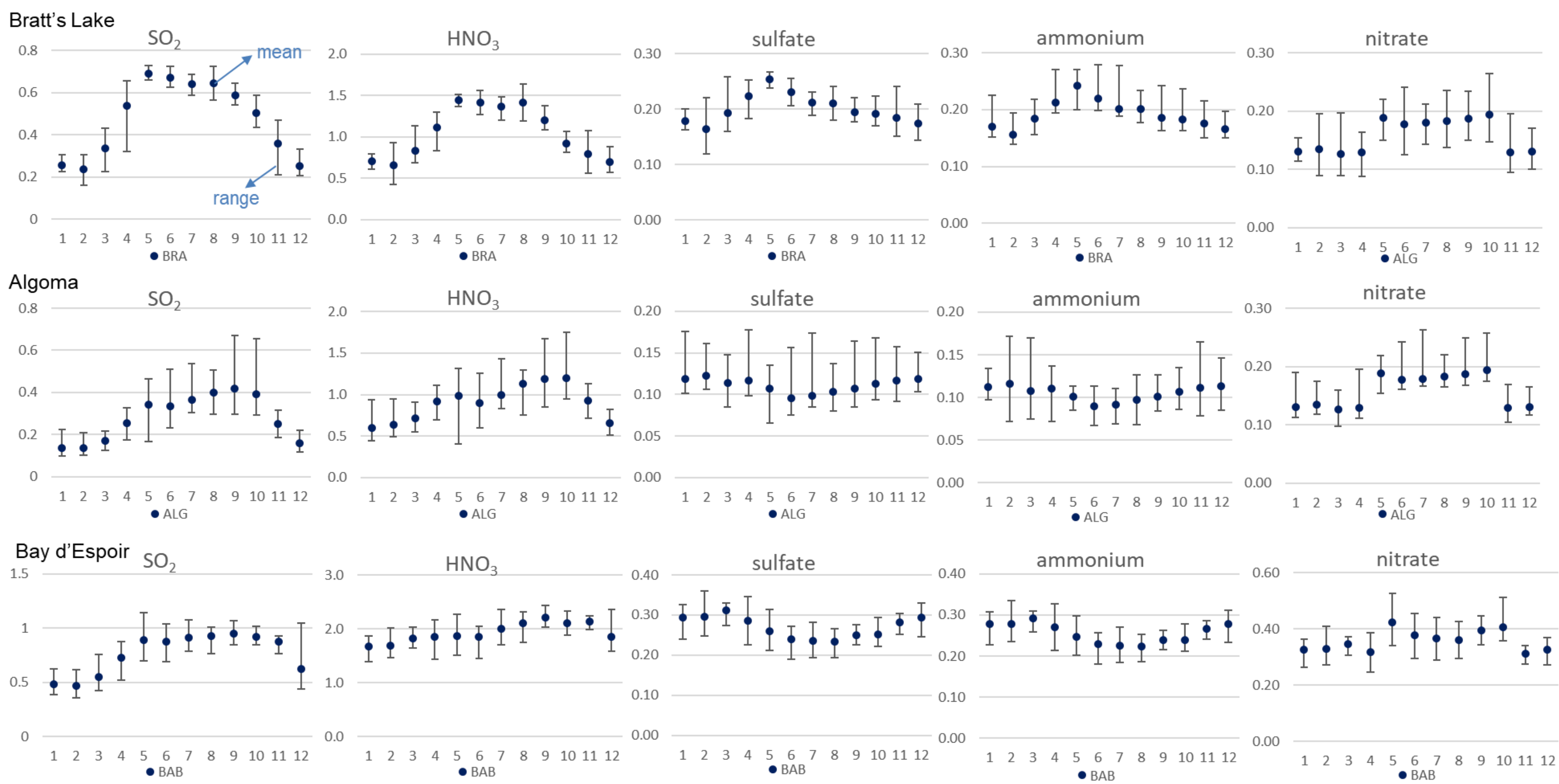
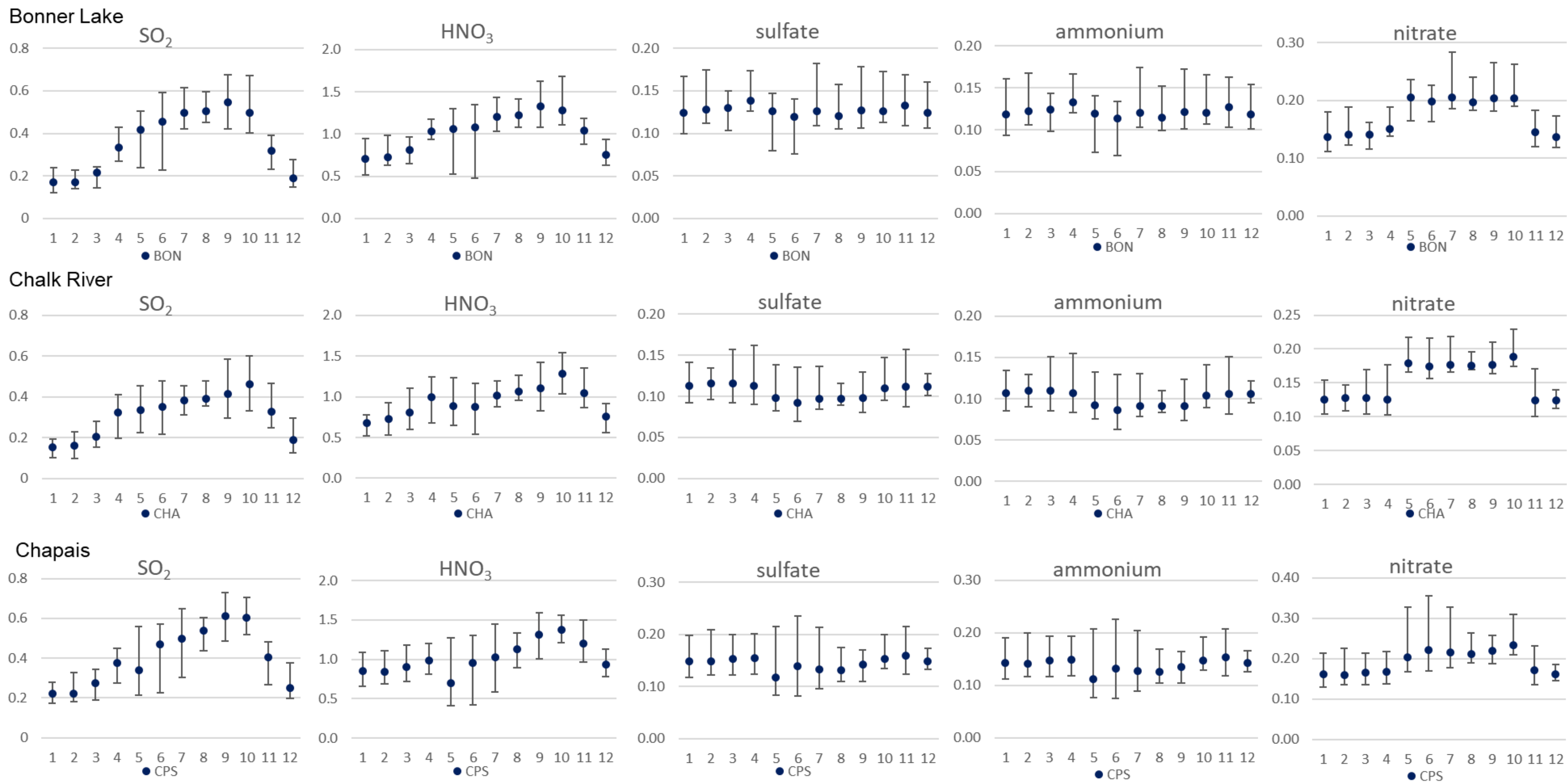


Figure S1: Monthly variation in dry deposition velocities (V_d, cm/s) of N and S species at CAPMoN sites during 2000-2018 period. Range is the annual variability in monthly V_d.



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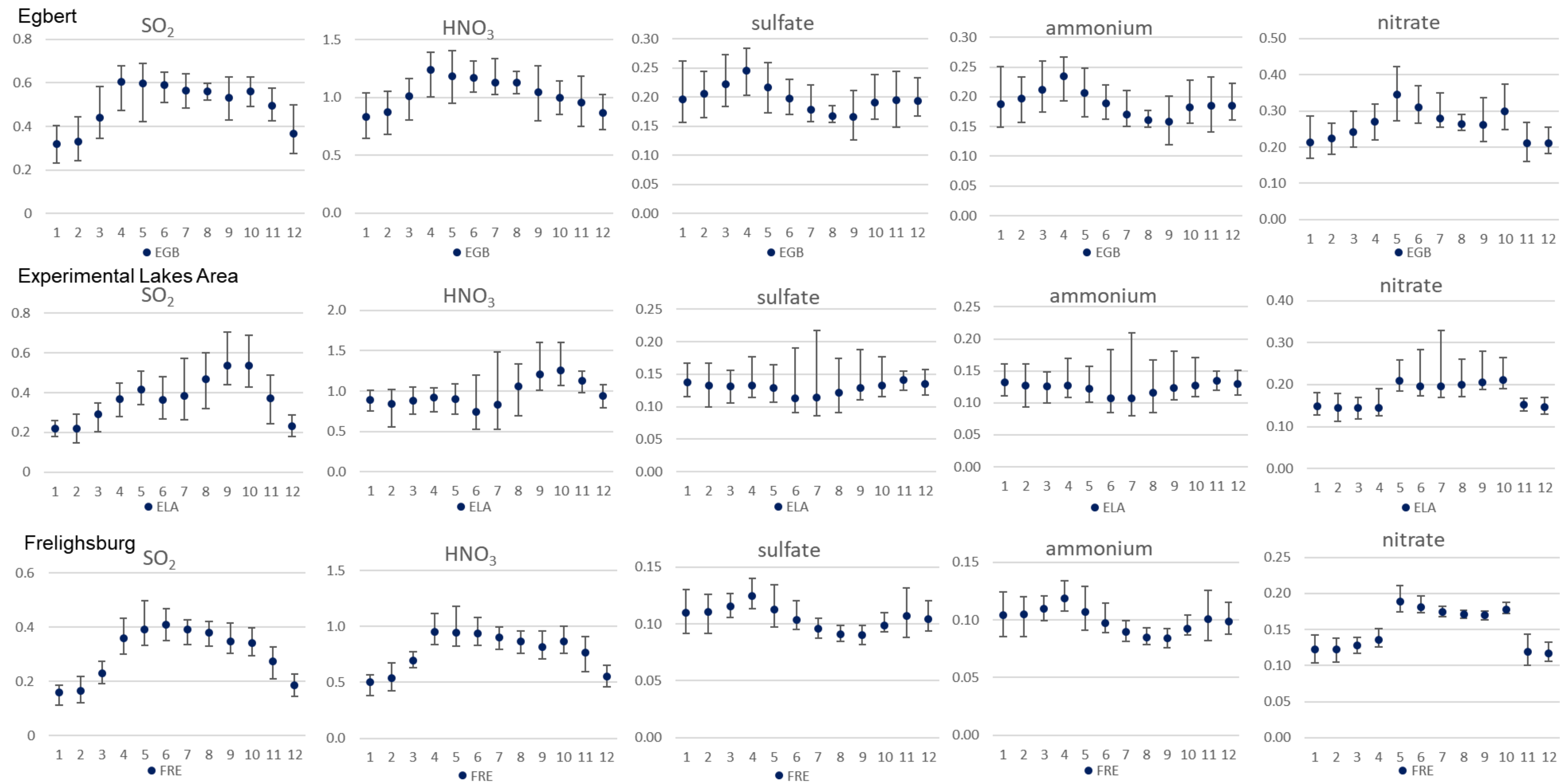

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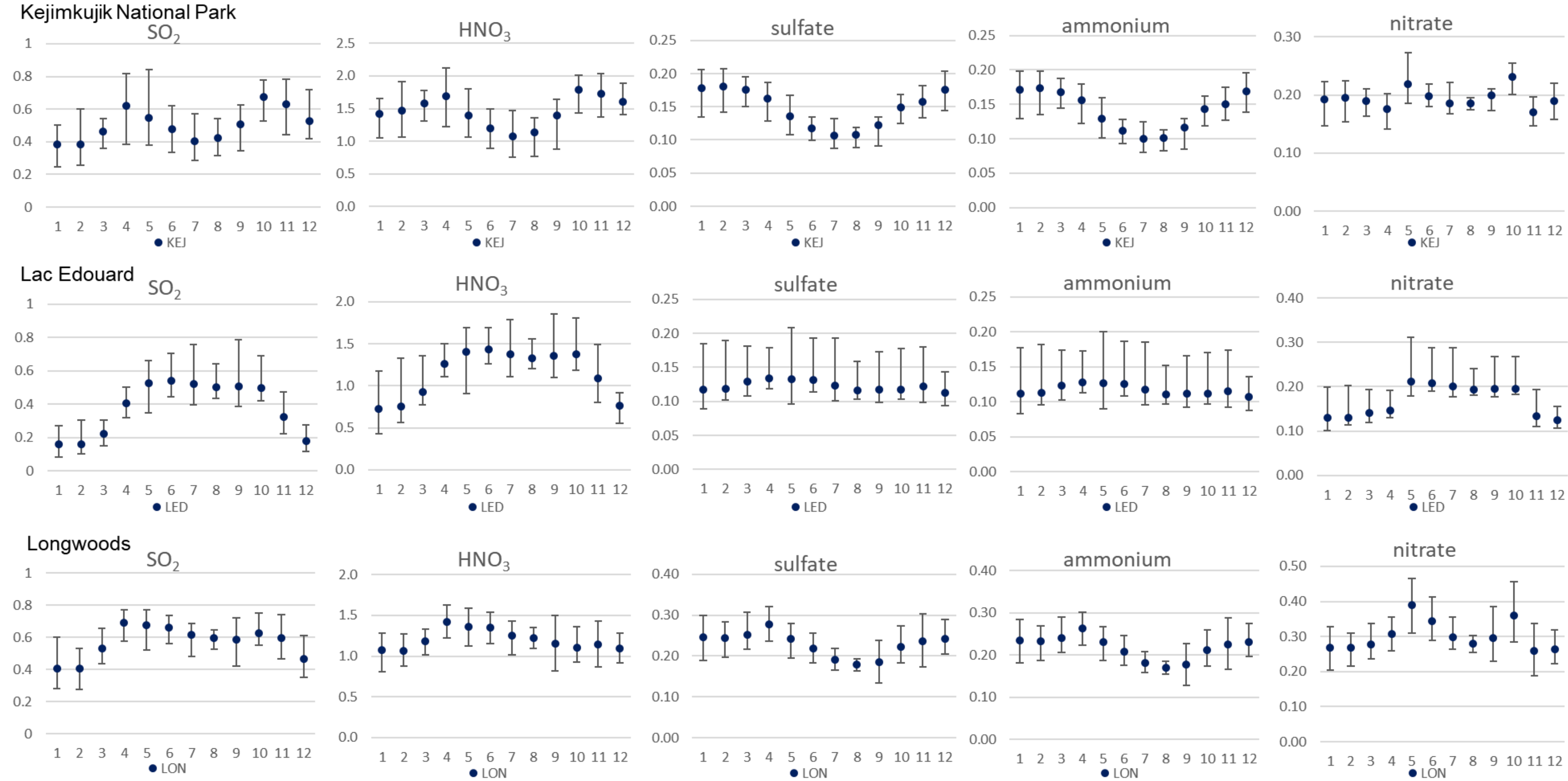

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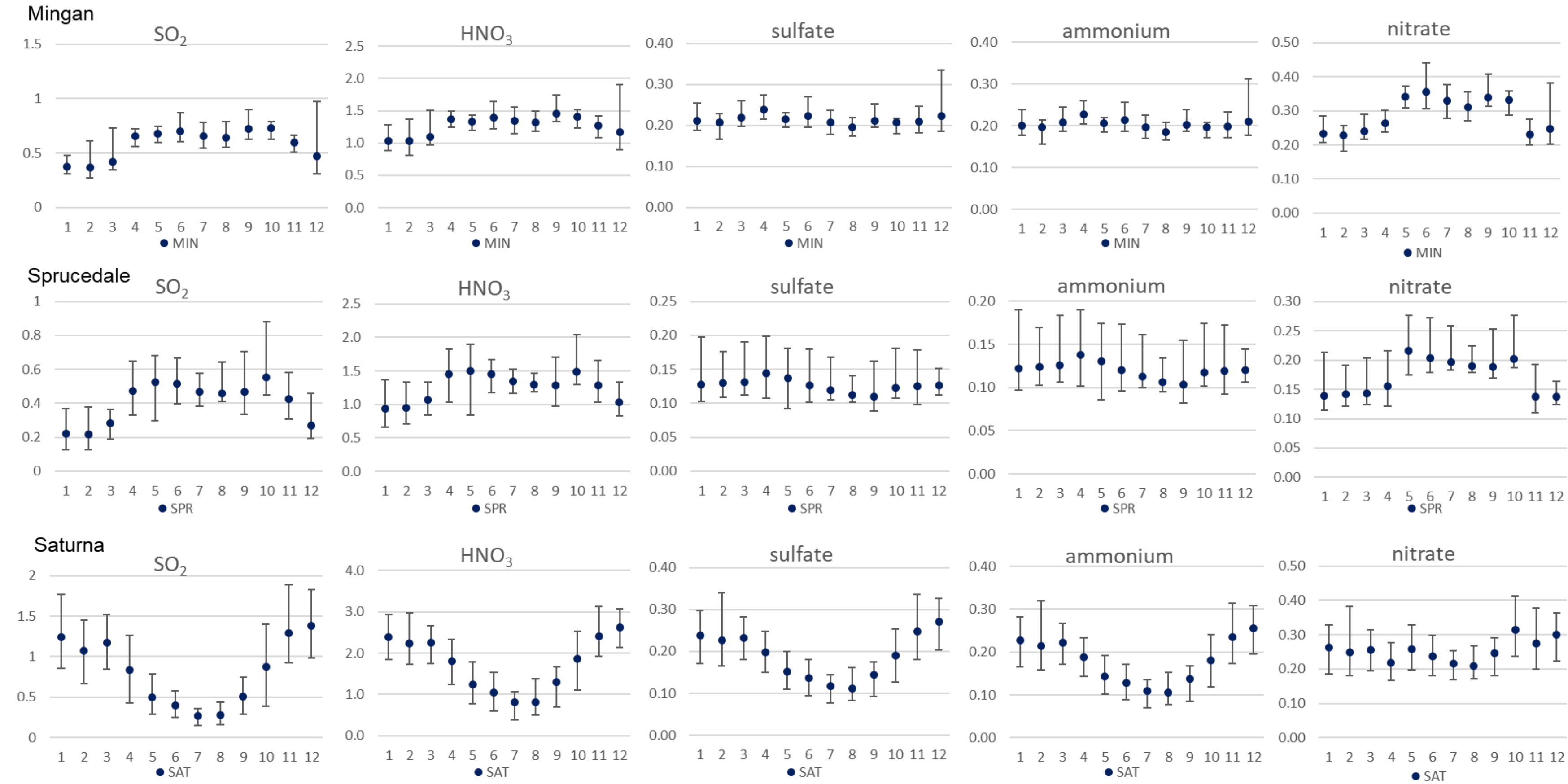

Figure S1 con't.

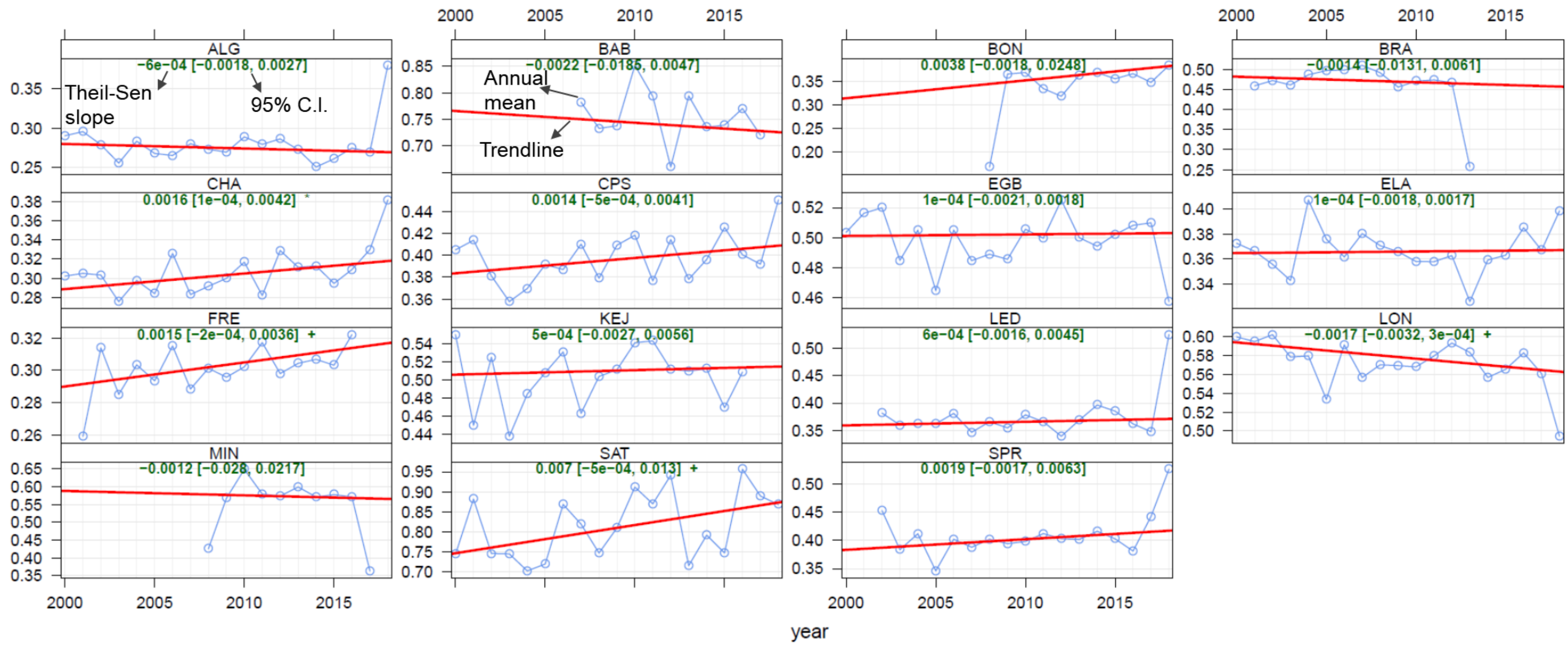

Figure S2: Long term trends in annual V_d of N and S species at CAPMoN sites. V_d of SO_2 shown above. Theil-Sen slope: change in annual Vd (cm/s per year). 95% C.I.: confidence interval of the slope; + symbol indicates p<0.1; * symbol indicates p<0.05 or statistically significant trend; no symbol indicates trend is not statistically significant.

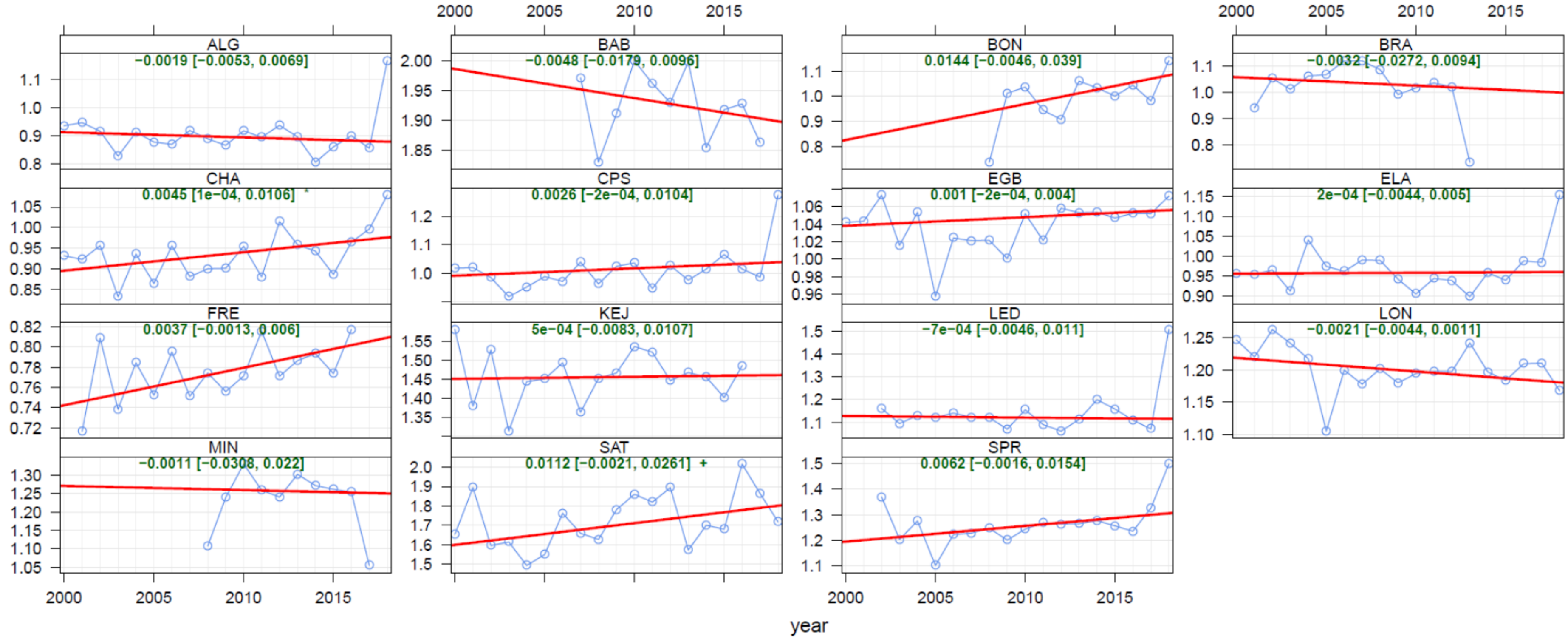

Figure S2 con't - V_d of HNO_3 at CAPMoN sites.

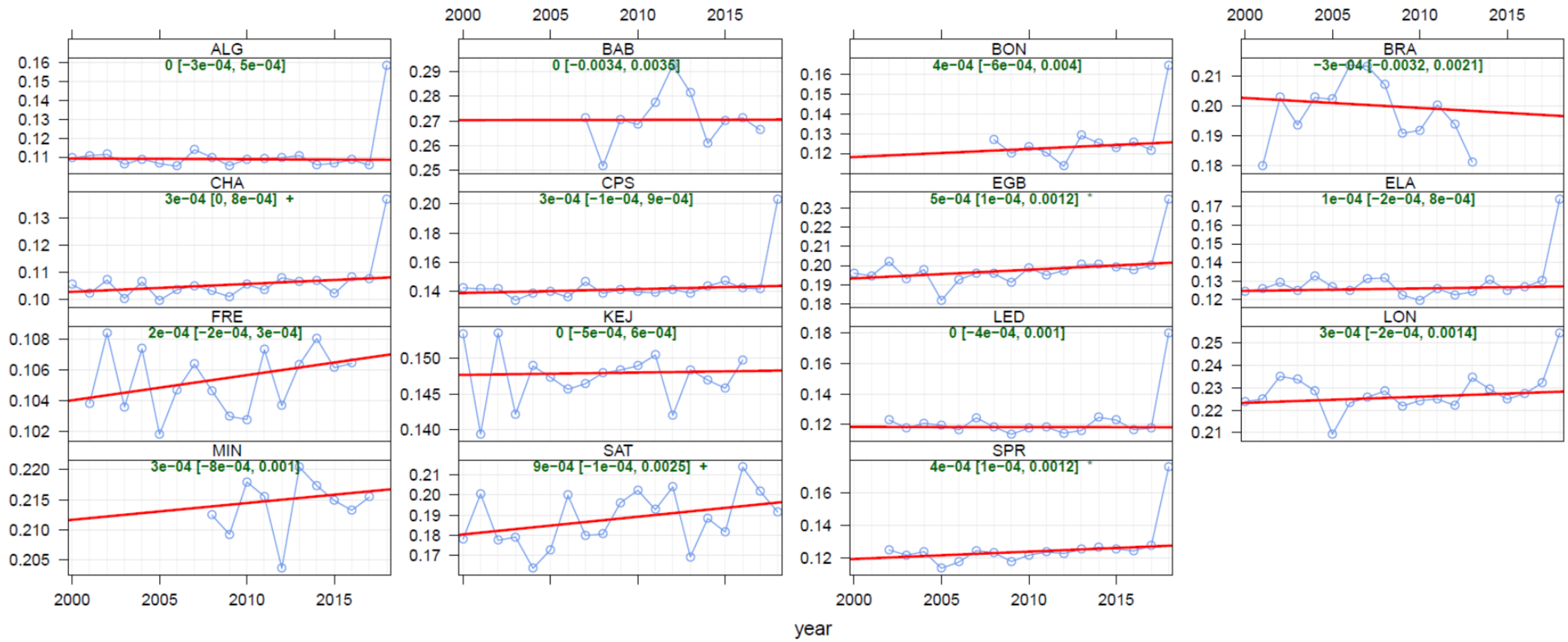

Figure S2 con't - V_d of pSO_4^{2-} at CAPMoN sites.

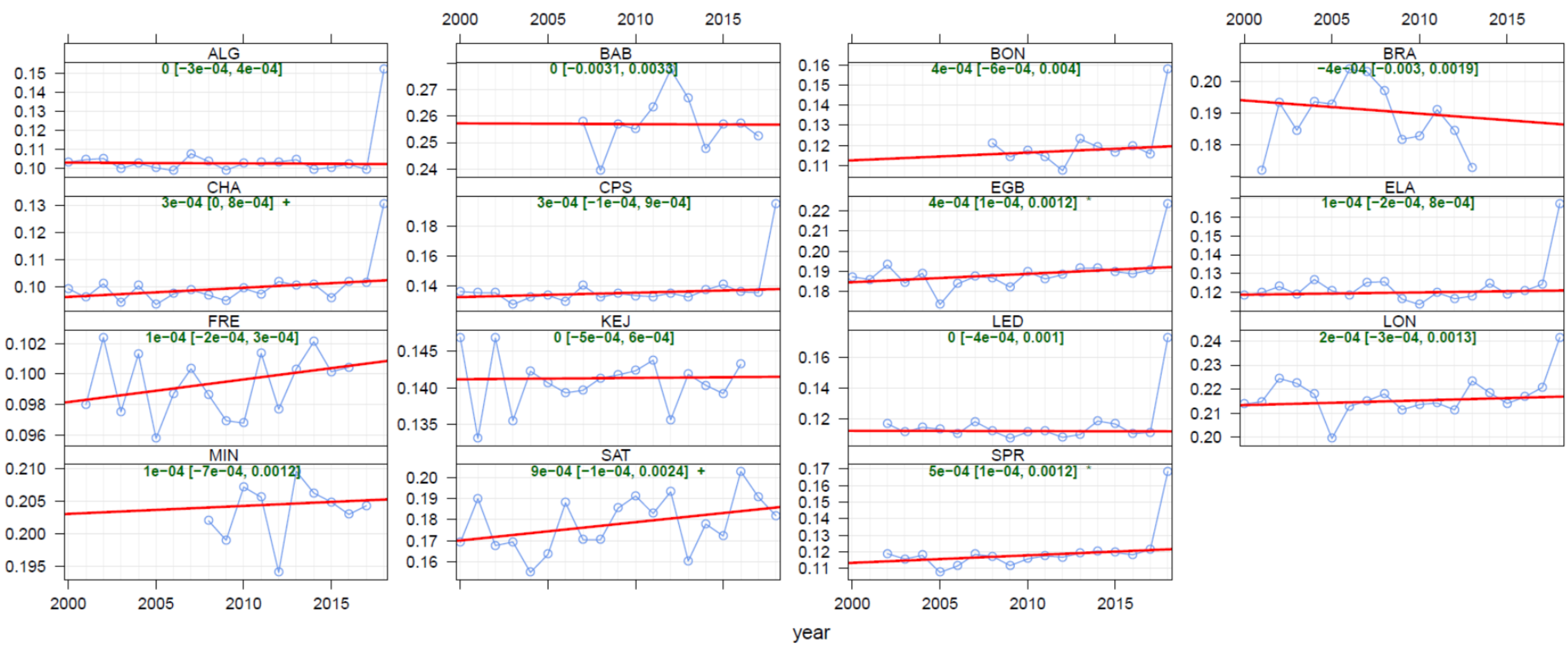

Figure S2 con't - V_d of pNH_4^+ at CAPMoN sites.

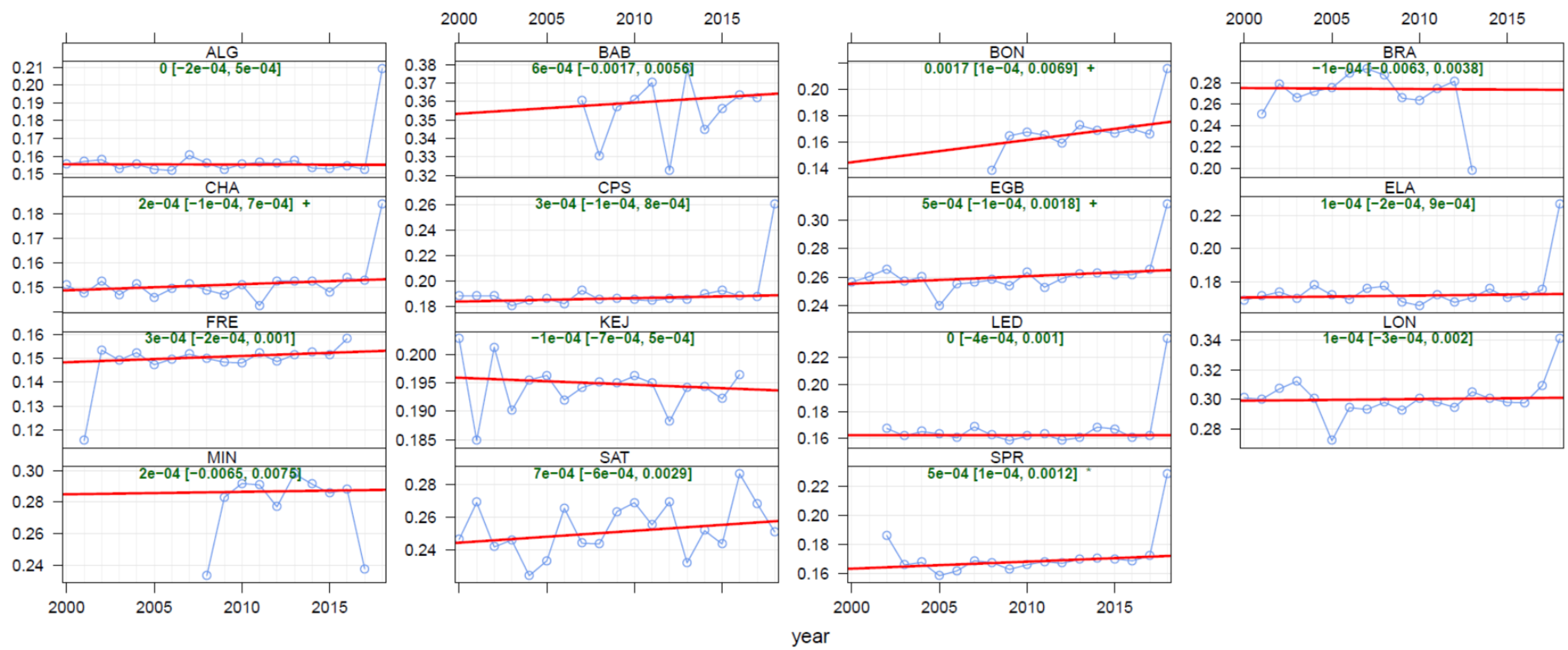

Figure S2 con't - V_d of pNO_3^- at CAPMoN sites.

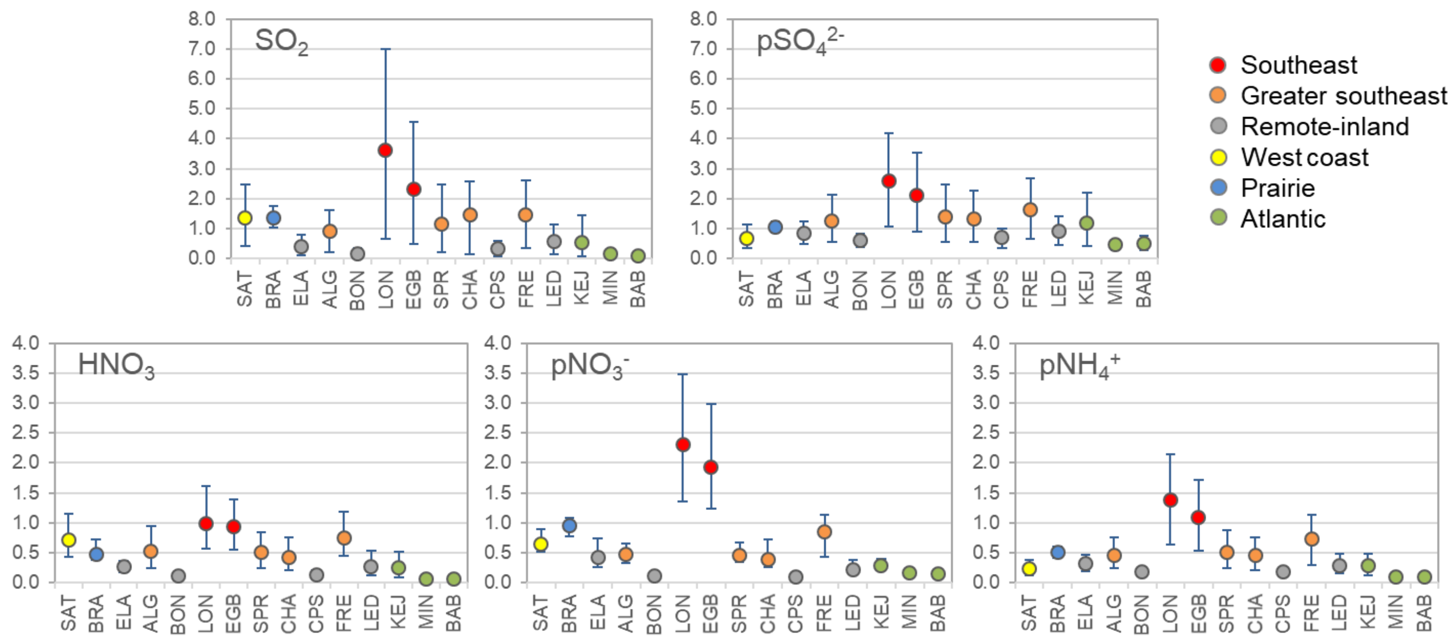

Figure S3: Annual mean of the 24-h integrated atmospheric S and N species concentrations (μg of ion/m^3) at CAPMoN sites during the 2000-2018 period. Error bars indicate the range of the annual means. Sites are color-coded by region and arranged in order longitudinally from west to east. Source: ECCC (2021a).

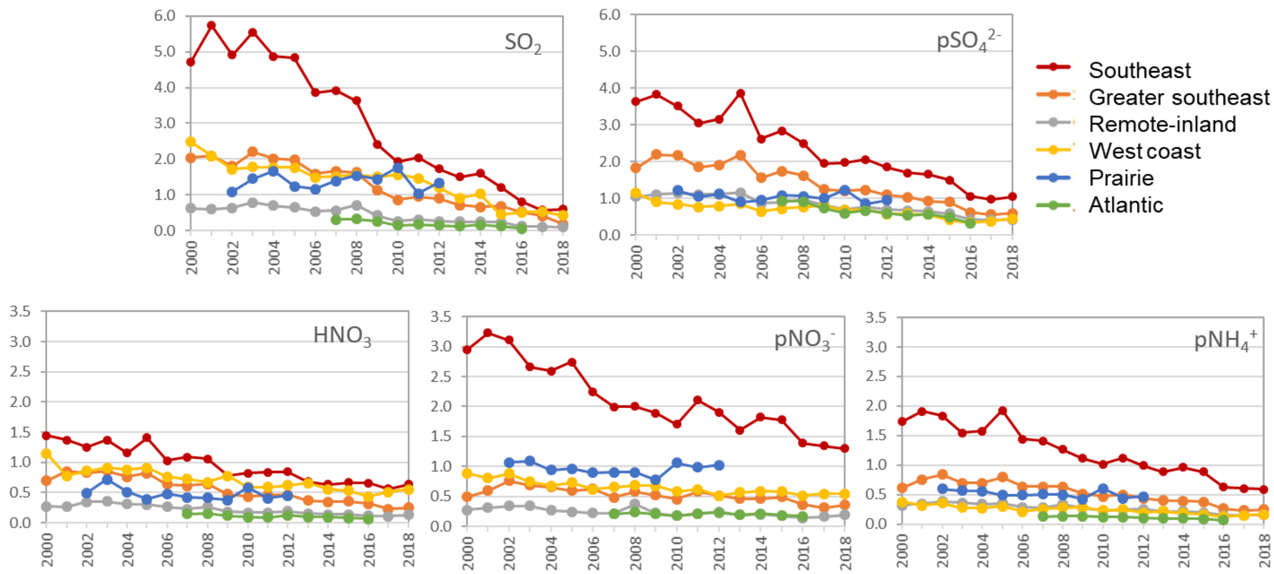

Figure S4: Annual trends in atmospheric S and N species concentrations (µg of ion/m^3) at CAPMoN sites grouped by region. CAPMoN sites belonging to each region are listed as follows. Southeast: EGB, LON; Greater southeast: ALG, SPR, CHA, FRE; Remote-inland: ELA, BON, CPS, LED; West coast: SAT; Prairie: BRA; Atlantic: KEJ, MIN, BAB. Source: ECCC (2021a).

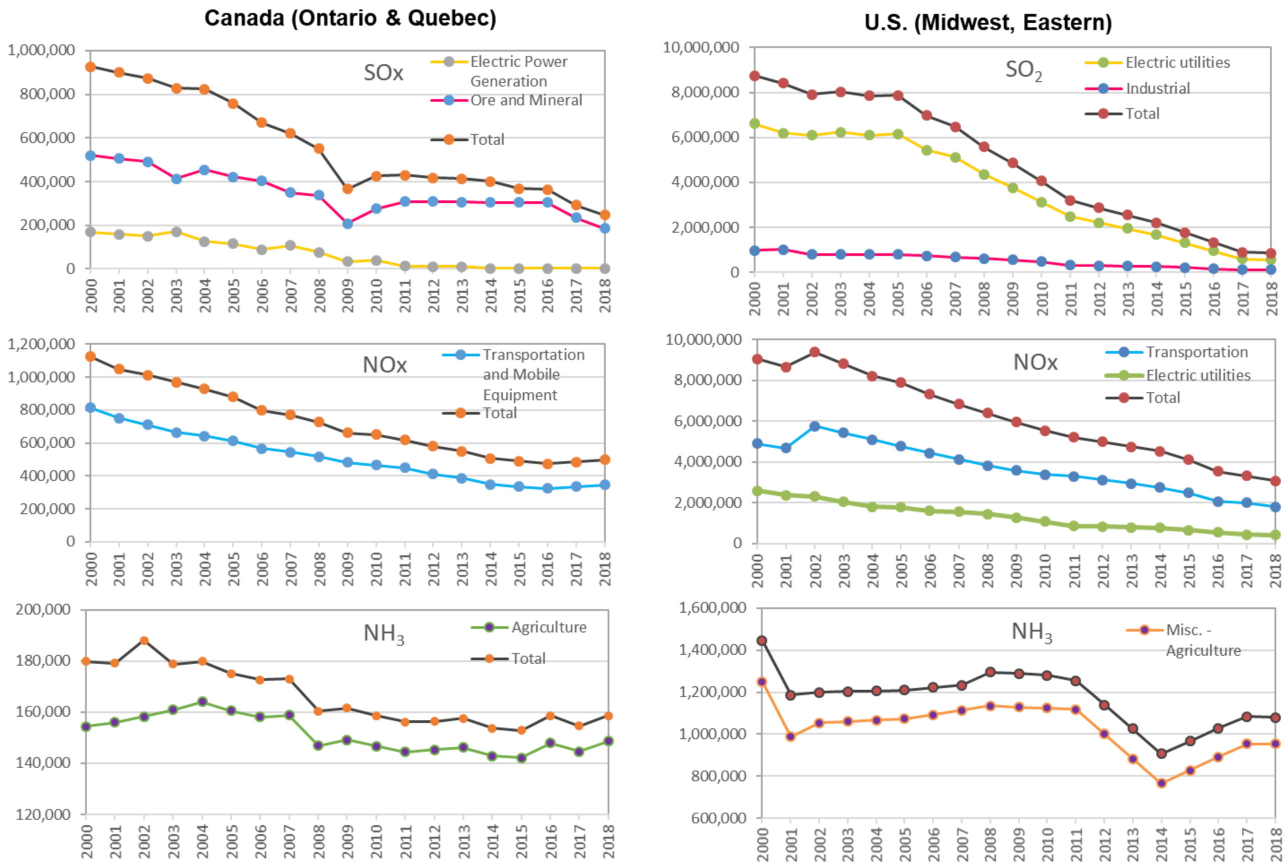

Figure S5a: Annual trends in emissions (tonnes) in provinces of Ontario and Quebec and in Midwest and Eastern U.S. Source: ECCC APEI (2021b) and USEPA (2021).

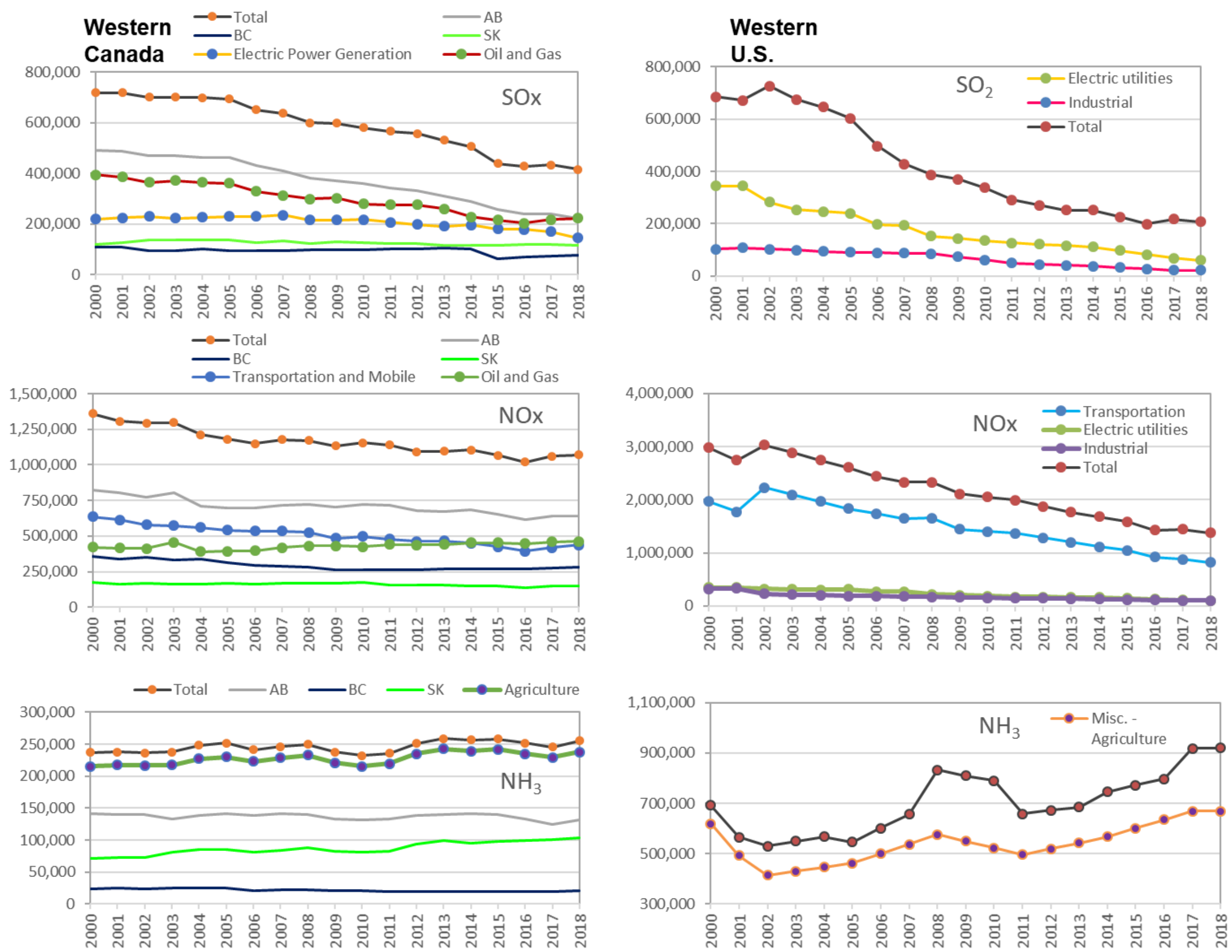

Figure S5b: Annual trends in emissions (tonnes) in provinces of British Columbia, Alberta and Saskatchewan and in Western U.S. Source: ECCC APEI (2021b) and USEPA (2021).

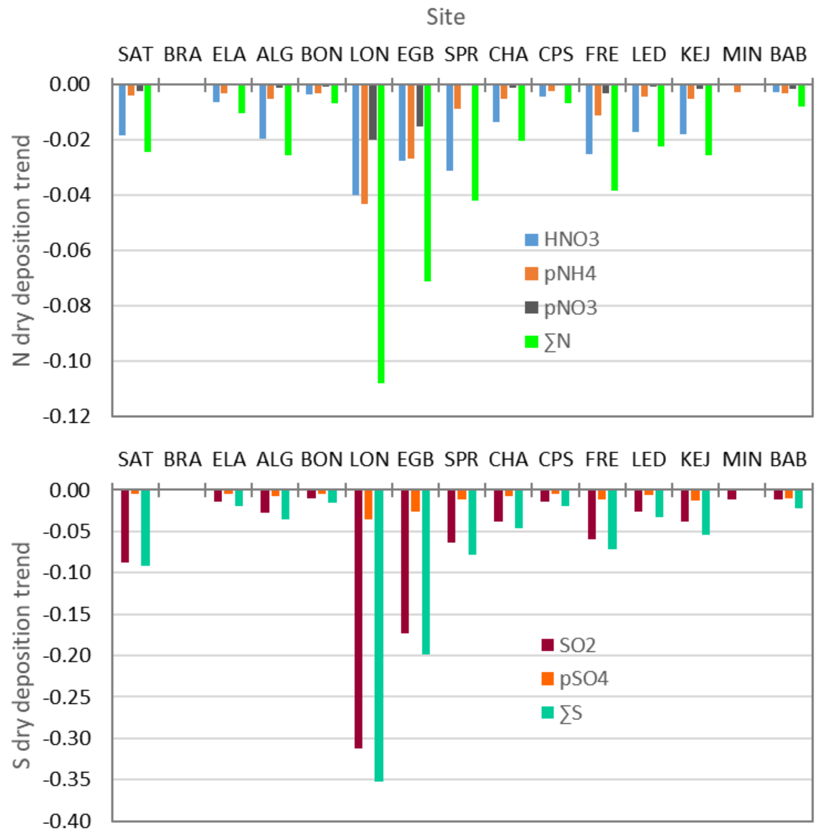

Figure S6: Trends in annual dry deposition fluxes of N and S species (kg N or S/ha/yr) at CAPMoN sites during 2000-2018 based on Theil-Sen slopes (statistically significant at $p<0.05$). Sites (denoted by site IDs) are plotted longitudinally from left (west) to right (east). Trends for some N or S species were not significant at BRA, ELA, SPR, CPS and MIN. Note: ∑N excludes dry deposition of NH_3 and some oxidized nitrogen species.

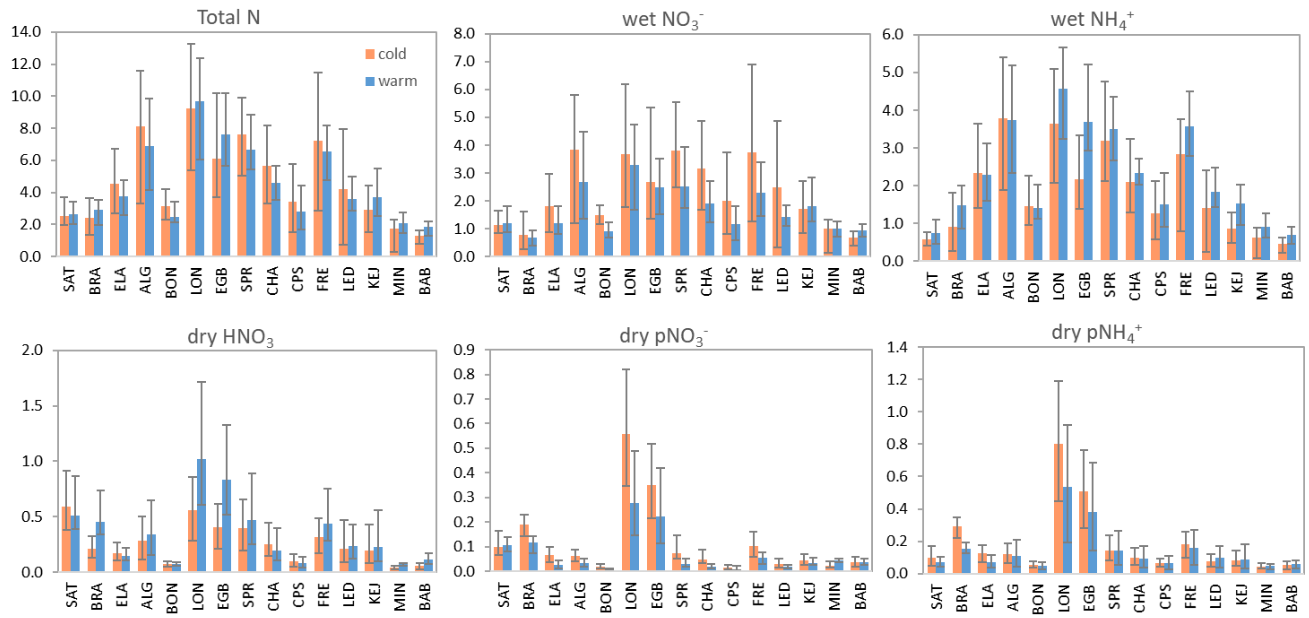

Figure S7: Mean and range of N deposition (kg N/ha/yr) during the cold (Nov-Apr) and warm (May-Oct) seasons during 2000-2018 at CAPMoN sites (denoted by site IDs) plotted longitudinally from left (west) to right (east). Error bars indicate annual variability in seasonal deposition.

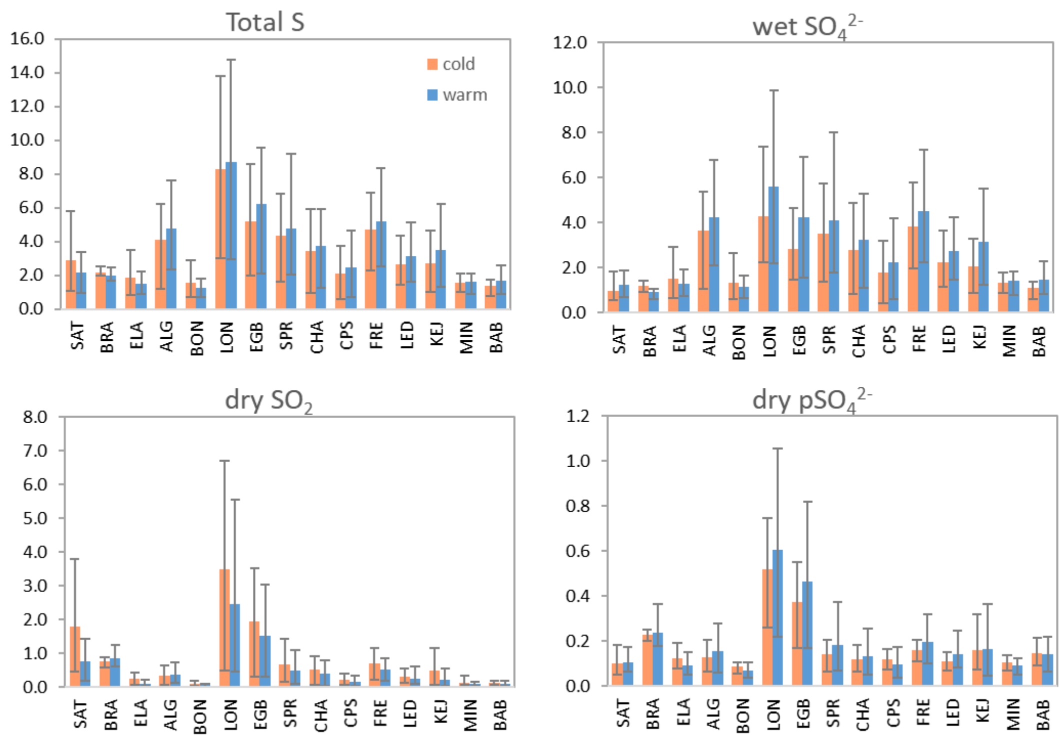

Figure S8: Mean and range of S deposition (kg S/ha/yr) during the cold (Nov-Apr) and warm (May-Oct) seasons during 2000-2018 at CAPMoN sites (denoted by site IDs) plotted longitudinally from left (west) to right (east). Error bars indicate annual variability in seasonal deposition.

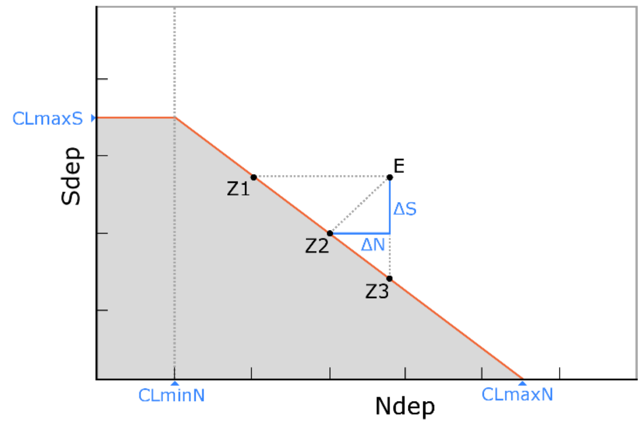

Figure S9: The critical load function for sulfur and nitrogen is defined by the maximum critical load of sulfur when nitrogen deposition (Ndep) is zero (CLmaxS), the maximum critical load of nitrogen when sulfur deposition (Sdep) is zero (CLmaxN), and a minimum critical load of nitrogen (CLminN) above which nitrogen deposition is acidifying. Sites where Sdep and Ndep are below the function line (the grey area) are protected from damage from acid deposition (e.g. not in exceedance). Exceedance (point E) is defined as the shortest path to non-exceedance, as illustrated by the points Z1, Z2, and Z3, but there is no unique path to non-exceedance. For more details see Posch et al. (2015).

Supplementary Information references

Beaudoin, A., Bernier, P. Y., Guindon, L., Villemaire, P., Guo, X. J., Stinson, G., Bergeron, T., Magnussen, S., and Hall, R. J.: Mapping attributes of Canada's forests at moderate resolution through kNN and MODIS imagery, Can. J. For. Res., 44(5), 521–532, https://doi.org/10.1139/cjfr-2013-0401, 2014.

Bonten, L. T. C., Reinds, G. J., and Posch, M.: A model to calculate effects of atmospheric deposition on soil acidification, eutrophication and carbon sequestration, Environ. Model. Softw., 79, 75–84, https://doi.org/10.1016/j.envsoft.2016.01.009, 2016.

CCRS: 2010 Land Cover of North America at 250 meters (v2.0), Canada Centre for Remote Sensing (CCRS), Earth Sciences Sector, Natural Resources Canada, http://www.cec.org/naatlas/, 2018.

Centre for Land and Biological Resources Research: Soil Landscapes of Canada, v.2.2, Research Branch, Agriculture and Agri-Food Canada, Ottawa, https://sis.agr.gc.ca/cansis/nsdb/slc/v2.2/index.html, 1996.

CLRTAP: Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends, CLRTAP, www.icpmapping.org, 2004.

Dymond, C. C., Titus, B. D., Stinson, G., and Kurz, W. A.: Future quantities and spatial distribution of harvesting residue and dead wood from natural disturbances in Canada, For. Ecol. Manag., 260(2), 181–192, https://doi.org/10.1016/j.foreco.2010.04.015, 2010.

Environment and Climate Change Canada (ECCC): Major ions and acidifying gases, Government of Canada Open Data Portal, https://open.canada.ca/data/en/dataset/10ec2a54-9b6d-4dd7-9b05-5c30b9fa4920, 2021a.

Environment and Climate Change Canada (ECCC): Canada's Air Pollutant Emissions Inventory (APEI), Government of Canada Open Data Portal, https://open.canada.ca/data/en/dataset/fa1c88a8-bf78-4fcb-9c1e-2a5534b92131, 2021b.

Hengl, T.: Absolute depth to bedrock (in cm) [Dataset], SoilGrids250m, https://data.isric.org/geonetwork/srv/api/records/f36117ea-9be5-4afd-bb7d-7a3e77bf392a, 2017.

Hengl, T.: Long-term MODIS LST day-time and night-time temperatures, sd and differences at 1 km based on the 2000–2017 time series (1.0) [Data set], Zenodo, https://doi.org/10.5281/zenodo.1435938, 2018a.

Hengl, T.: Sand content in % (kg / kg) at 6 standard depths (0, 10, 30, 60, 100 and 200 cm) at 250 m resolution (v0.2) [Data set], Zenodo, https://doi.org/10.5281/zenodo.2525662, 2018b.

Hengl, T.: Soil bulk density (fine earth) 10 x kg / m-cubic at 6 standard depths (0, 10, 30, 60, 100 and 200 cm) at 250 m resolution (v0.2) [Data set], Zenodo, https://doi.org/10.5281/zenodo.2525665, 2018c.

Hengl, T.: Clay content in % (kg / kg) at 6 standard depths (0, 10, 30, 60, 100 and 200 cm) at 250 m resolution (v0.2) [Dataset], Zenodo, https://doi.org/10.5281/zenodo.2525663, 2018d.

Hengl, T., and Wheeler, I.: Soil organic carbon content in x 5 g / kg at 6 standard depths (0, 10, 30, 60, 100 and 200 cm) at 250 m resolution (v0.2) [Data set], Zenodo, https://doi.org/10.5281/zenodo.2525553, 2018.

Makar, P. A., Akingunola, A., Aherne, J., Cole, A. S., Aklilu, Y., Zhang, J., Wong, I., Hayden, K., Li, S.-M., Kirk, J., Scott, K., Moran, M. D., Robichaud, A., Cathcart, H., Baratzedah, P., Pabla, B., Cheung, P., Zheng, Q., and Jeffries, D. S.: Estimates of exceedances of critical loads for acidifying deposition in Alberta and Saskatchewan, Atmos. Chem. Phys., 18(13), 9897–9927, https://doi.org/10.5194/acp-18-9897-2018, 2018.

McKenney, D., Papadopol, P., Campbell, K., Lawrence, K., and Hutchinson, M.: Spatial models of Canada-and North America-wide 1971/2000 minimum and maximum temperature, total precipitation and derived bioclimatic variables. Frontline, forestry research applications, Technical Note, 106, Natural Resources Canada, www.publications.gc.ca/pub?id=9.559411&sl=1, 2006.

Pardo, L. H., Duarte, N., Miller, E. K., and Robin-Abbott, M.: Tree chemistry database (version 1.0) (Vol. 324), USDA Forest Service, Northeastern Research Station, 2005.

Paré, D., Bernier, P., Lafleur, B., Titus, B. D., Thiffault, E., Maynard, D. G., and Guo, X.: Estimating stand-scale biomass, nutrient contents, and associated uncertainties for tree species of Canadian forests, Can. J. For. Res., 43(7), 599–608, https://doi.org/10.1139/cjfr-2012-0454, 2013.

Posch, M., de Vries, W., Sverdrup, H.: Mass Balance Models to Derive Critical Loads of Nitrogen and Acidity for Terrestrial and Aquatic Ecosystems. In: de Vries, W., Hettelingh, JP., Posch, M. (eds) Critical Loads and Dynamic Risk Assessments, Environ. Pollut., 25, Springer, Dordrecht, https://doi.org/10.1007/978-94-017-9508-1_6, 2015.

Reinds, G. J., Posch, M., Aherne, J., and Forsius, M.: Assessment of Critical Loads of Sulphur and Nitrogen and Their Exceedances for Terrestrial Ecosystems in the Northern Hemisphere, In W. De Vries, J.-P. Hettelingh, and M. Posch (Eds.), Critical loads and dynamic risk assessments: Nitrogen, acidity and metals in terrestrial and aquatic ecosystems (Vol. 25, pp. 403–418), Springer, 2015.

Schut, P., Smith, S., Fraser, W., Geng, X., and Kroetsch, D.: Soil Landscapes of Canada: Building a National Framework for Environmental Information, GEOMATICA, 65(3), 293–309, https://doi.org/10.5623/cig2011-045, 2011.

Sverdrup, H., and Warfvinge, P.: The effect of soil acidification effect on the growth of trees, grass and herbs, as expressed by the (Ca+ Mg+ K)/Al ratio, Reports in ecology and environmental engineering, Department of Chemical Engineering II, Lund University, 1993.

USEPA: Air pollutant emissions trends data, https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data, 2021.

Zhang, L., Vet, R., O'Brien, J. M., Mihele, C., Liang, Z., and Wiebe, A.: Dry deposition of individual nitrogen species at eight Canadian rural sites. J. Geophys. Res. Atmos., 114, D02301, https://doi.org/10.1029/2008JD010640, 2009.

Zhang, L., Vet, R., Wiebe, A., Mihele, C., Sukloff, B., Chan, E., Moran, M. D., and Iqbal, S.: Characterization of the size-segregated water-soluble inorganic ions at eight Canadian rural sites, Atmos. Chem. Phys., 8(23), 7133-7151, 2008.