



Supplement of

Diverging trends in aerosol sulfate and nitrate measured in the remote North Atlantic in Barbados are attributed to clean air policies, African smoke, and anthropogenic emissions

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Table S1: Normalized mean bias (NMB), mean bias (MB), root-mean-square error (RMSE), and the Pearson correlation coefficient (r) for EQUATES model runs for nitrate and nss-sulfate compared to observations for each year from Ragged Point.

Year	Compound	NMB (%)	MB (ug/m3)	RMSE (ug/m3)	r (pearson correlation coefficient)
2002	Nitrate	-10.18	-0.06	0.19	-0.22
	NSS-Sulfate	81.45	0.54	0.60	0.79
2003	Nitrate	-6.18	-0.03	0.15	0.60
	NSS-Sulfate	57.70	0.39	0.51	0.65
2004	Nitrate	-5.99	-0.03	0.16	0.59
	NSS-Sulfate	41.29	0.27	0.37	0.65
2005	Nitrate	-24.18	-0.15	0.24	0.84
	NSS-Sulfate	3.13	0.02	0.29	0.72
2006	Nitrate	-17.21	-0.10	0.28	0.45
	NSS-Sulfate	28.15	0.21	0.42	0.38
2007	Nitrate	-6.51	-0.04	0.14	0.62
	NSS-Sulfate	40.13	0.31	0.40	0.64
2008	Nitrate	-35.06	-0.26	0.35	0.52
	NSS-Sulfate	3.97	0.03	0.30	0.55
2009	Nitrate	-9.51	-0.06	0.20	0.28
	NSS-Sulfate	23.89	0.16	0.35	0.23
2010	Nitrate	-28.72	-0.23	0.31	0.83
	NSS-Sulfate	-14.42	-0.13	0.29	0.41
2011	Nitrate	-11.57	-0.07	0.12	0.88
	NSS-Sulfate	-8.80	-0.08	0.24	0.82

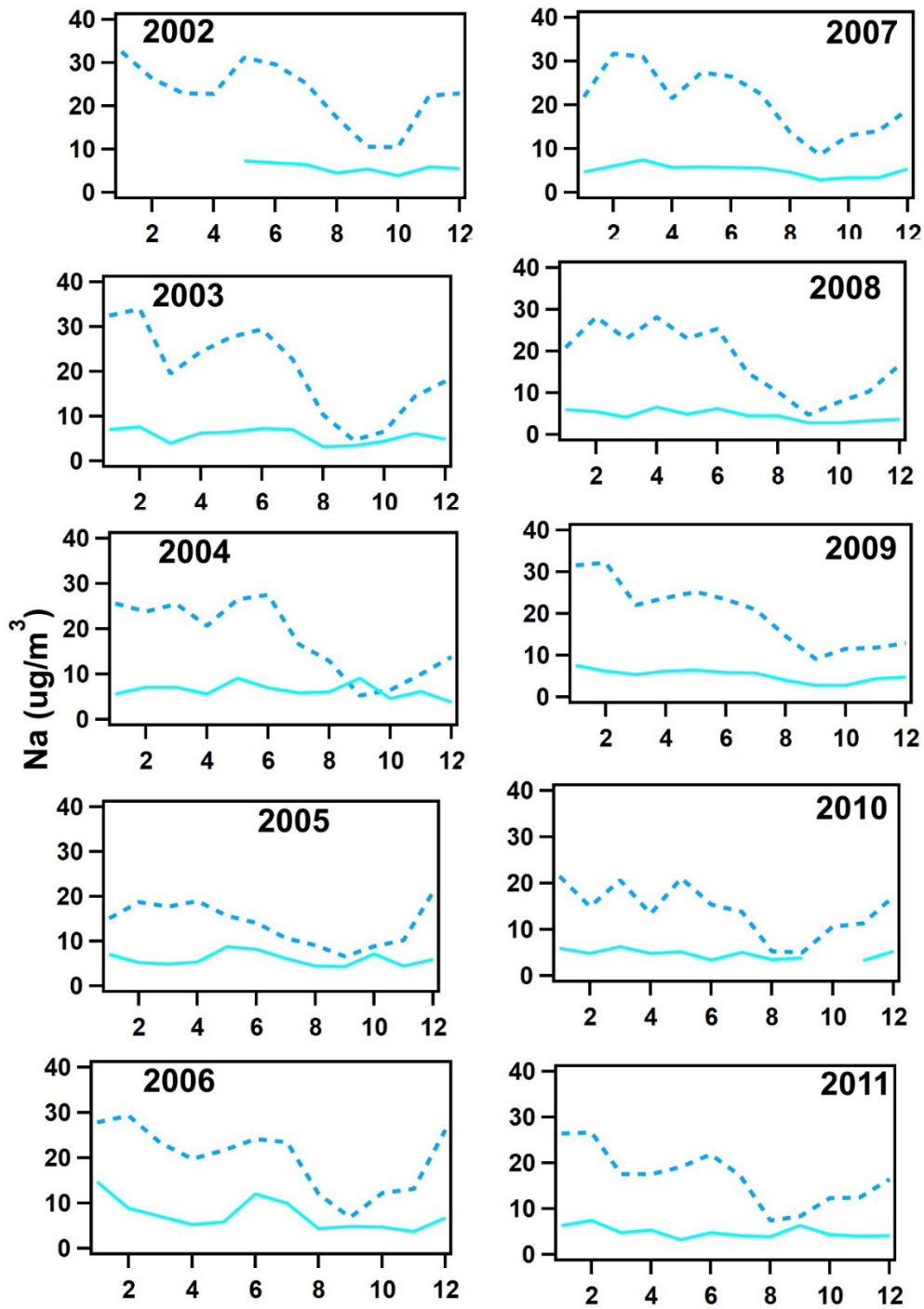


Figure S1: Monthly comparison of sodium (Na^+) mass concentrations measured at Barbados on filters (solid blue line) and predicted by the EQUATES model (dashed blue line) from 2002-2011.

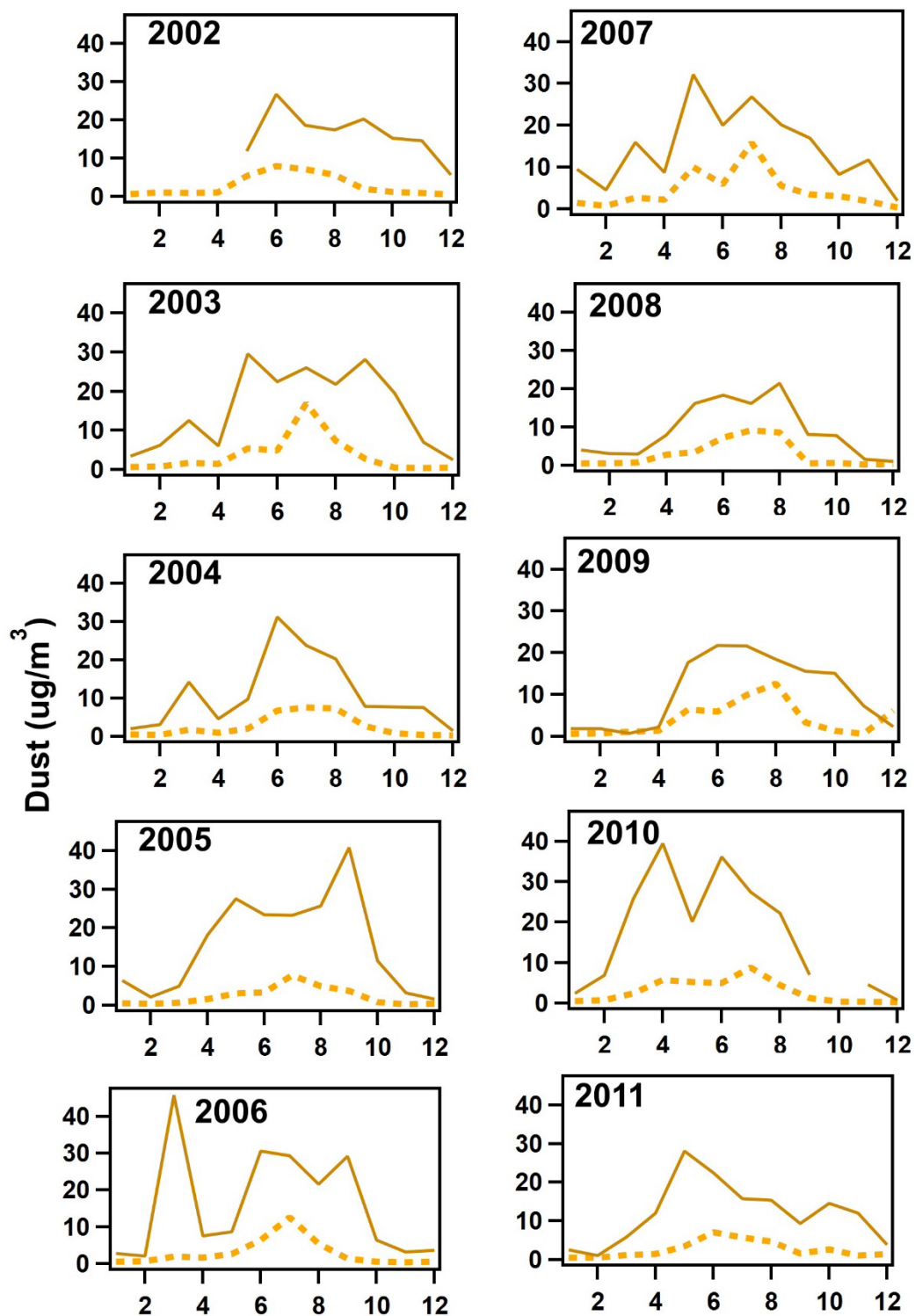


Figure S2: Monthly comparison of dust mass concentrations measured at Barbados on filters (solid brown line) and dust predicted by the EQUATES model (dashed brown line) using nss-Ca^{2+} concentrations scaled to the upper crustal abundance of calcium for 2002-2011.

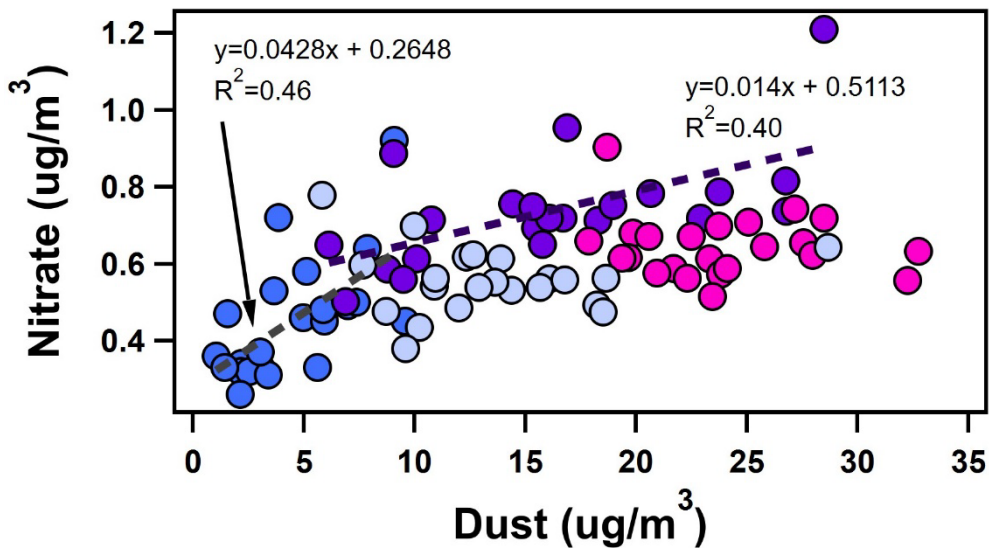
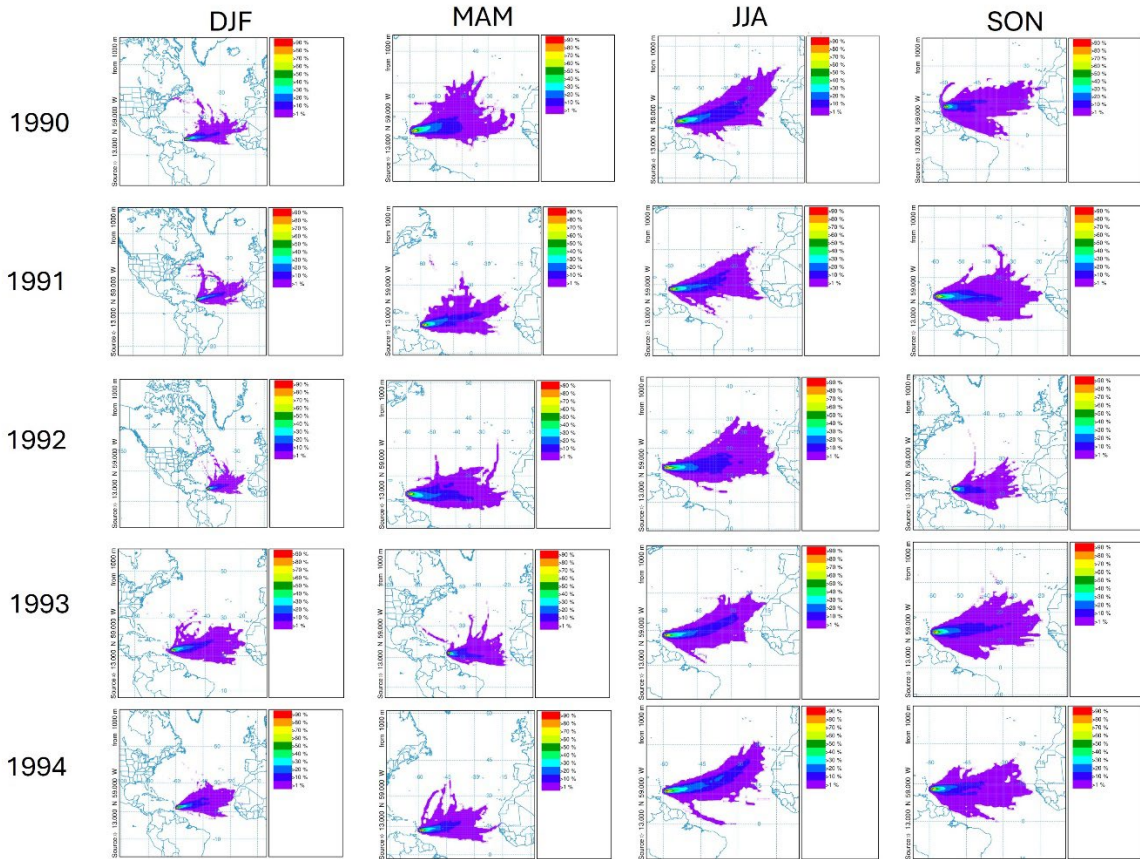


Figure S3: Comparison of nitrate and dust mass concentrations as a function of season with DJF in blue, MAM in purple, JJA in pink, and SON in light blue.



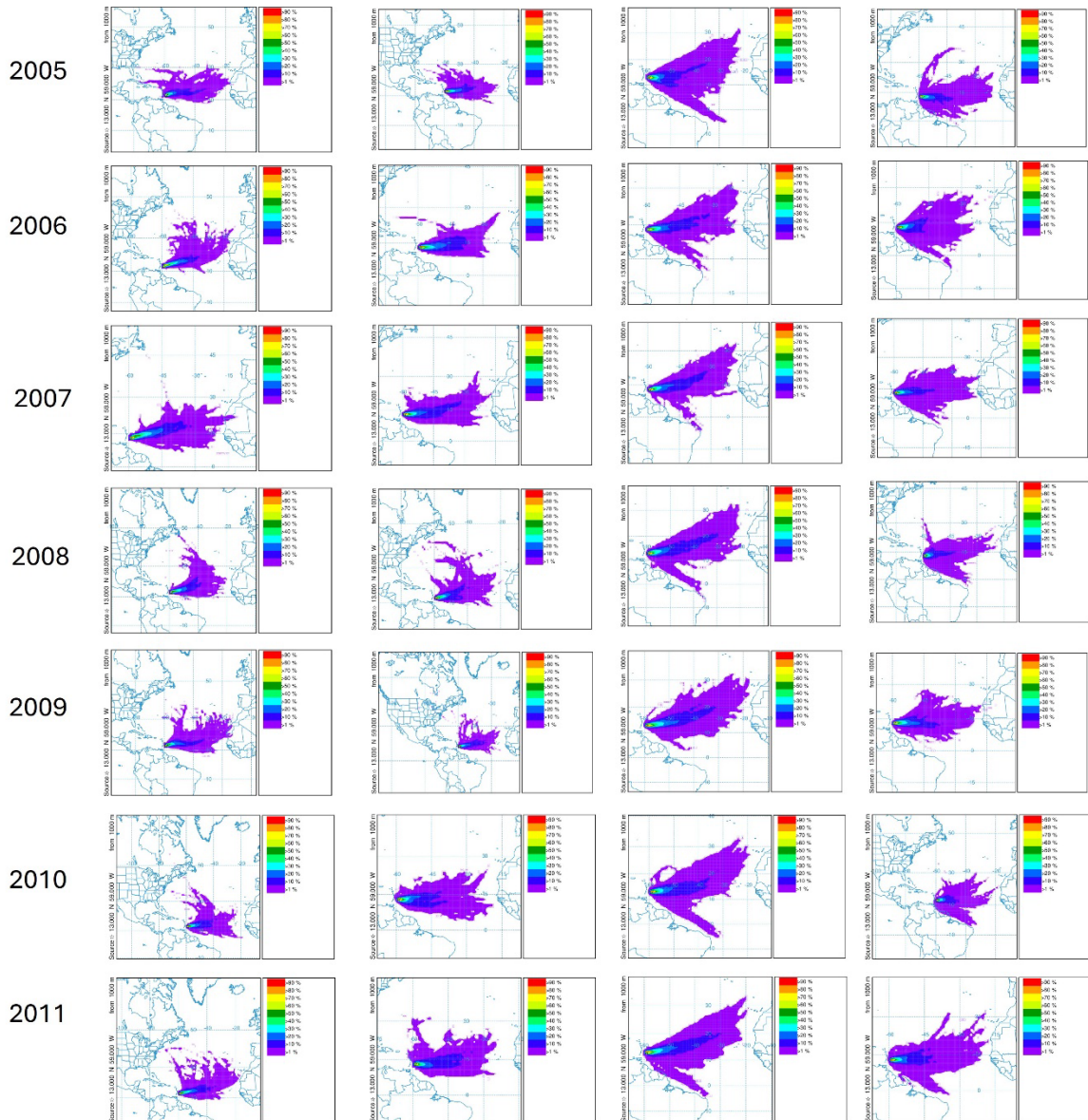


Figure S4: Seasonal HYSPLIT back trajectory frequency plots for DJF (Winter), MAM (Spring), JJA (summer), and SON (Fall). 5-day air mass back-trajectories were initiated at a height of 1000 m.

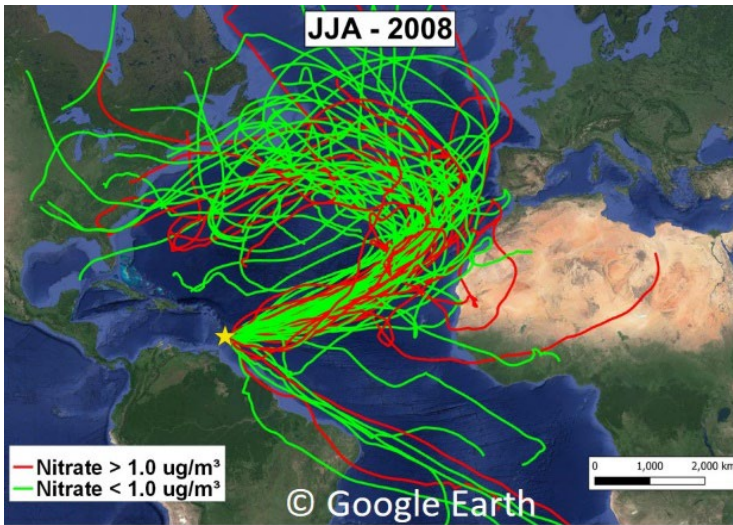


Figure S5: 315-h air mass back-trajectories initiated at 500-m for JJA 2008. Back-trajectories labeled in green are for days where nitrate concentrations measured at Ragged Point were $< 1 \text{ ug/m}^3$ while trajectories labeled in red had nitrate $> 1 \text{ ug/m}^3$. Maps are from Google Earth (©Google Earth).

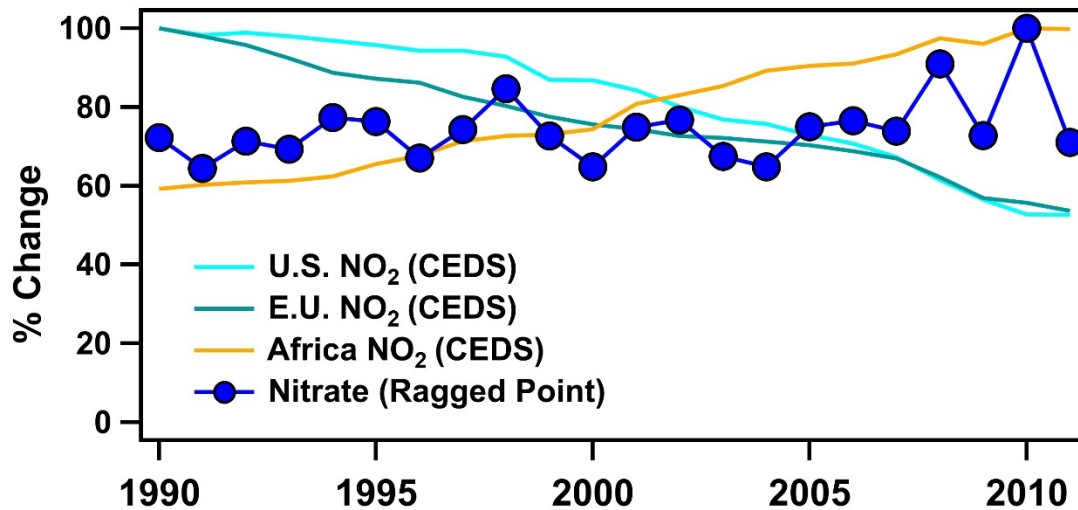


Figure S6: Nitrate mass concentrations measured at Ragged Point from 1990-2011 are shown in blue lines with markers. Emissions of nitrogen dioxide (NO₂) from McDuffie et al., 2020 are included for comparison. Decreasing emissions of NO₂ from the US and EU are shown in blue lines. Increasing emissions of NO₂ from Africa are shown in orange.

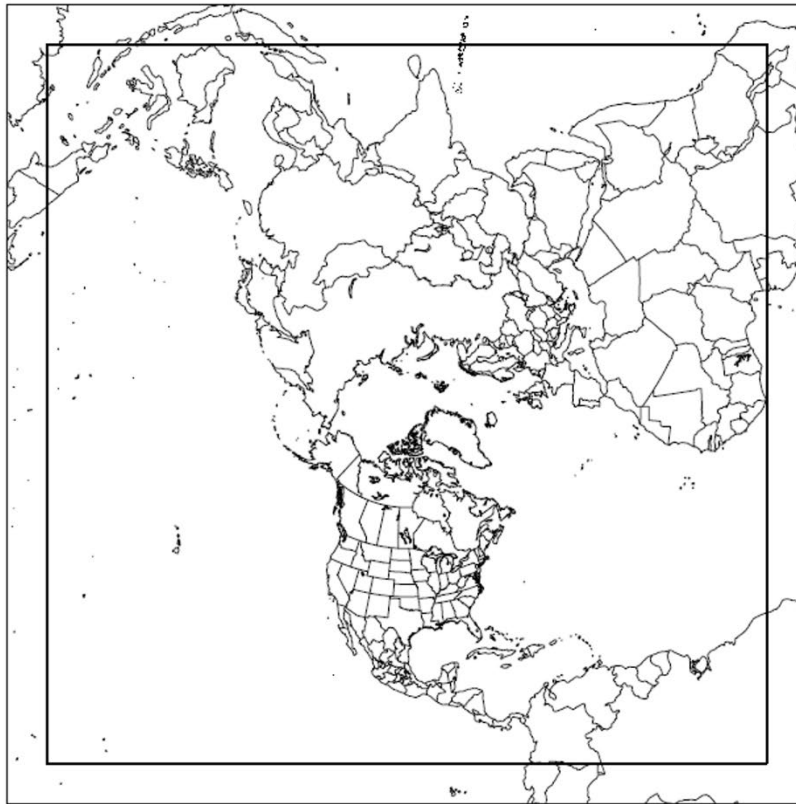
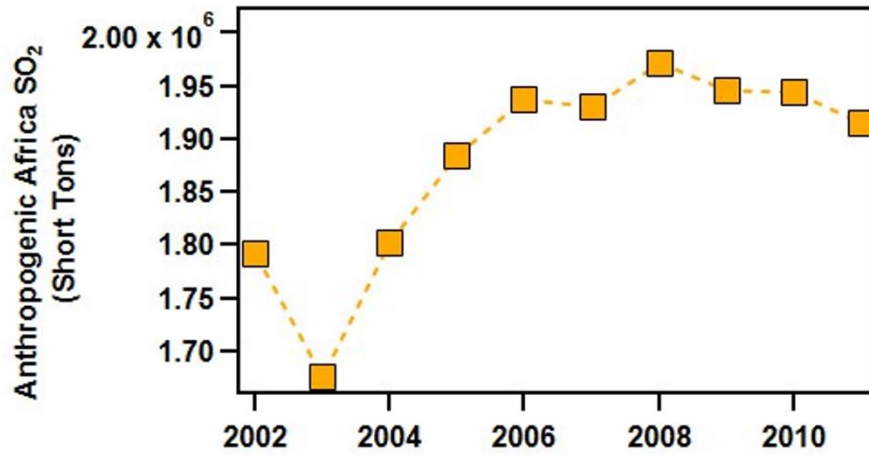


Figure S7: EQUATES simulations of SO₂ emissions from Africa from anthropogenic sources from 2002-2011 (top panel). Note that only part of Africa is included in the EQUATES Northern Hemisphere domain as shown in the bottom panel, thus, emissions do not reflect the entire continent. The bottom panel shows the EQUATES domain over the Northern Hemisphere (187 rows x 187 columns x 44 vertical layers). Horizontal grid resolution is 108 km x 108 km.

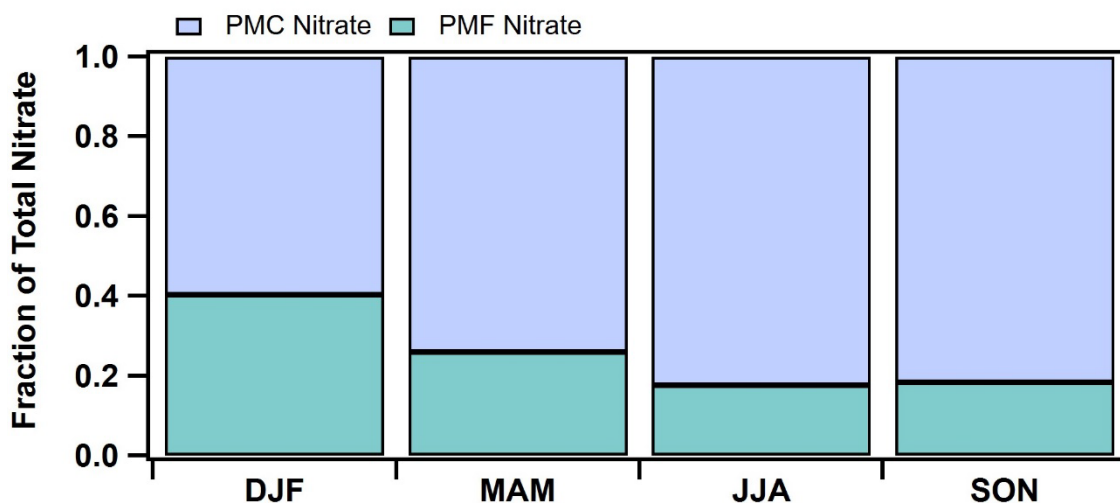


Figure S8: Seasonal averages of coarse mode (PMC, light blue) and fine mode (PMF, dark teal) contributions to aerosol nitrate predicted by EQUATES.

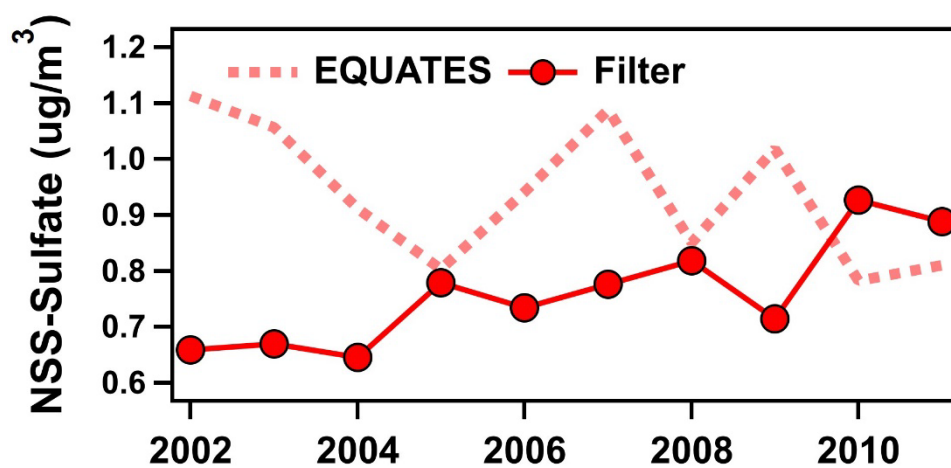


Figure S9: Annual averages of NSS-Sulfate measured from filters collected at Ragged Point (solid red line) and predicted by EQUATES (dashed pink line).

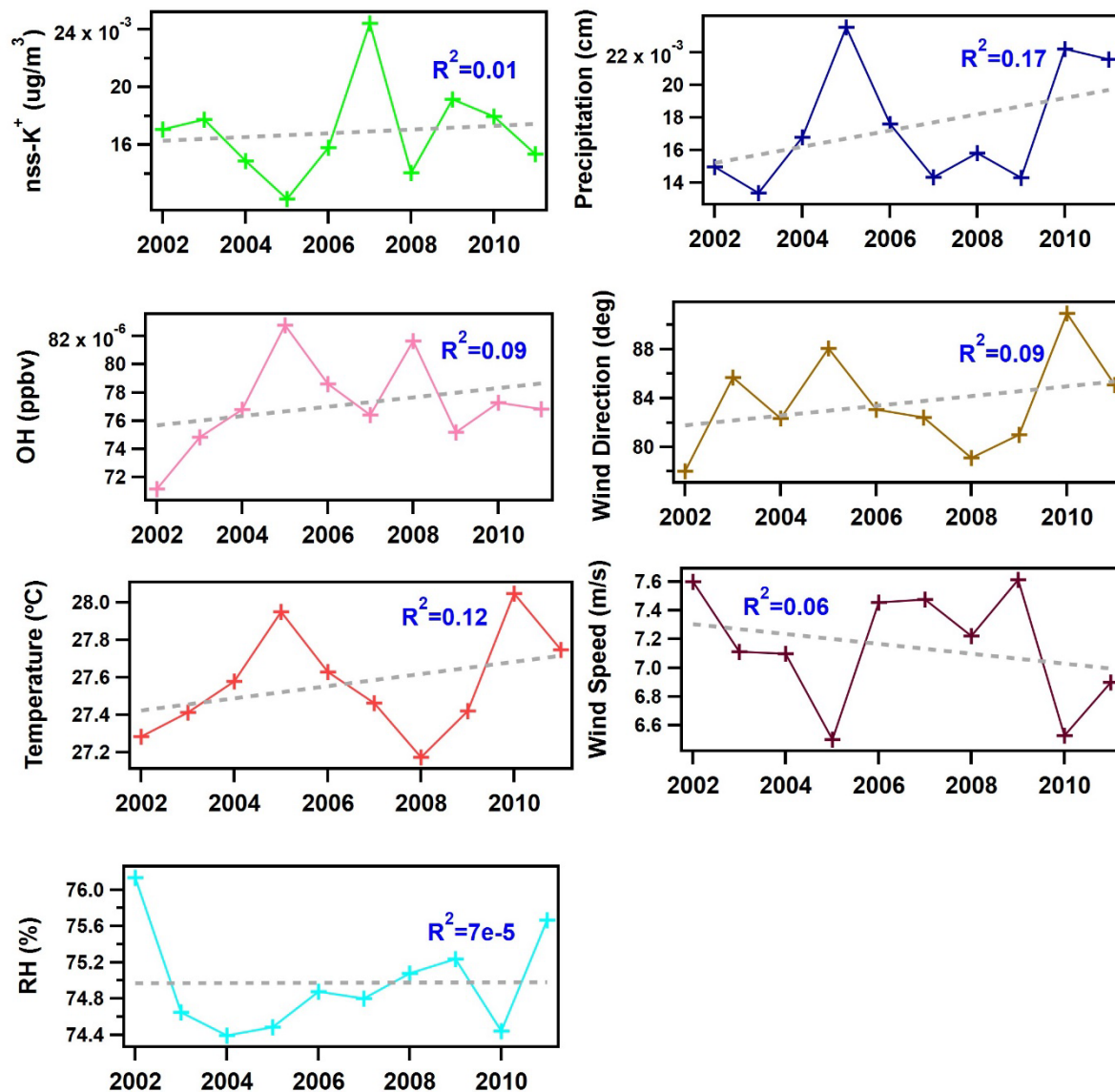


Figure S10: Annual averages of non-sea salt-potassium (nss-K⁺), hydroxyl radical concentrations (OH), temperature, relative humidity (RH), wind speed, and wind direction calculated from EQUATES model predictions. Linear fits and correlation coefficients are also shown.