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

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ABSTRACT

Sulfate and nitrate aerosols degrade air quality, modulate radiative forcing and the hydrological cycle, and affect critical biogeochemical cycles, yet their global cycles are poorly understood. To address this issue, we examined trends in 21 years of aerosol measurements made at Ragged Point, Barbados—the easternmost promontory on the island located in the eastern Caribbean Basin. Though the site has historically been used to characterize African dust transport, here we focused on changes in nitrate and non-sea salt (nss) sulfate aerosol from 1990-2011. Nitrate aerosol concentrations are stable at 0.59 ug/m^3 ± 0.04 ug/m^3. Elevated nitrate concentrations in the spring of 2010 as well as during the summer and fall of 2008 are due to transported biomass burning emissions from both northern and southern Africa to our site. In contrast, nss-sulfate decreased 30% at a rate of 0.02 ug/m^3/yr in the 1990s, which we attribute to air quality policies enacted in the U.S. and Europe. Starting in 2000, sulfate began to increase to pre-1990s levels of 0.90 ug/m^3. We used the Community Multiscale Air Quality (CMAQ) model simulations from the EPA's Air QUAlity TimE Series (EQUATES) to better understand the changes in nss-sulfate after 2000. The model simulations estimate that increases in anthropogenic emissions, likely from Northern Africa, and increased oxidation efficiency of sulfur dioxide (SO_2) explain the increase in nss-sulfate observed in Barbados. Our results serve as an incentive to better constrain emissions from developing countries and their impact on aerosol burdens in remote regions.

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1. INTRODUCTION

Sulfate and nitrate aerosol, formed from gaseous sulfur dioxide (SO_2) and nitrogen oxides (e.g., $NO_x \equiv NO+NO_2$) as well as reactive nitrogen (e.g., NO_y), contribute to aerosol direct and indirect radiative forcing, impact biogeochemical cycles (Jickells et al., 2017), and degrade air quality (Adams et al., 1999; Appel et al., 1978; Charlson et al., 1992; Lelieveld & Heintzenberg, 1992; Murphy et al., 2006). An outstanding question is, how have sulfate and nitrate aerosol burdens in remote regions changed in response to air quality policies, economic growth, and changing frequency of wildfires—all of which have affected SO_2 and NO_x emissions? Answering this question is important as remote regions are an important barometer of changing global emission inventories, contain ecosystems sensitive to changing chemical inputs (Galloway et al., 2008; Mahowald et al., 2017), and are most sensitive to fluctuations in aerosol burdens that alter aerosol-cloud interactions (Carslaw et al., 2013). Long-term measurement records in remote regions can provide insight into this question as well as further advance current chemical transport and global climate models. However, there are few long-term measurement records in remote regions. In this work, we leverage 21 years of nitrate and sulfate aerosol concentrations measured at Ragged Point, Barbados, a remote site in the eastern North Atlantic marine boundary layer, and use simulations from a hemispheric chemical transport model—the Community Multiscale Air Quality (CMAQ) model within the EPA's Air QUAlity TimE Series (EQUATES) (Foley et al., 2023)—to link our observed changes in nitrate and sulfate to changing emissions inventories and meteorological conditions. In turn, comparing the EQUATES model output to our time series provides guidance on which emissions inventories are lacking in in-situ measurements that would improve measurement-model agreement.

Ragged Point, Barbados provides a unique opportunity to understand changes in the nitrate and sulfate aerosol burden in the remote North Atlantic marine boundary layer. Aerosol sampling began in 1971 and continues to this day generating an over 50 year measurement record—the longest modern speciated aerosol record to the best of our knowledge (Prospero et al., 2021; Prospero & Mayol-Bracero, 2013). The site serves as a lynchpin for understanding the impact of long-range aerosol transport on the remote North Atlantic marine boundary layer and Caribbean. The site's primary objective has been to understand the factors affecting the long-range transport of African dust to the Caribbean and North America, which peaks in boreal summer in association with the seasonal northward shift in the intertropical convergence zone (ITCZ). Summer dust events are caused by the strong heating of North Africa, which causes hot, dry dust-laden desert air to be carried to high altitudes, e.g., 4 – 6 km. African Easterly Waves propagate dust westward within an elevated air layer known as the Saharan Air Layer (SAL) that overrides the cool, moist marine boundary layer (Adams et al., 2012; Carlson & Prospero, 1972; Goudie & Middleton, 2001; Tsamalis et al., 2013). Along with dust, anthropogenic emissions from Europe (Lelieveld et al., 2002), North America, and North Africa are also transported to Barbados (Savoie, et al., 2002).

Anthropogenic emissions of SO_2 and NO_x from the regions that affect Ragged Point have changed in recent decades due to the opposing effects of decreasing emissions mandated by national air quality policies in developed countries and increasing emissions linked to rapid economic growth in developing countries. The United States (U.S.) curbed emissions of NO_x and

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SO_2 by passing the Clean Air Act (amended in 1990), resulting in a 92% reduction in SO_2 and a 71% reduction in NO_x emissions from 1990-2022 (Aas et al., 2019; Hand et al., 2012; https://gispub.epa.gov/air/trendsreport/2023/; Smith et al., 2011; Zhao et al., 2017). European countries passed similar policies resulting in analogous reductions (Aas et al., 2019; Rafaj et al., 2015; Smith et al., 2011; Yang et al., 2020). Notably, reductions in SO_2 can reduce aerosol acidity resulting in increased nitrate aerosol. Further, reductions in pollutant gases can relieve oxidant limitations leading to more efficient oxidation and, therefore, reductions in SO_2 and NO_x may not reduce sulfate and nitrate aerosol as much as expected (Shah et al., 2018). In contrast to clean air policies enacted in the US and EU, emissions in regions such as the Middle East, India, and Africa are continuing to increase due to rapid economic growth with emissions from India predicted to overtake China as the world's largest emitter of SO_2 (Lelieveld et al., 2009; Li et al., 2017; McDuffie et al., 2020). Due to a lack of in situ measurements in many of these regions, chemical transport and emissions inventory models combined with remote sensing have been key tools to understand changing pollutants.

In addition to fossil fuel emissions, biomass burning is also a major source of SO_2 and NO_x that can impact the Atlantic (Andreae, 2019; Andreae & Merlet, 2001; Rickly et al., 2022; Roberts et al., 2009; Zuidema et al., 2018). Wildfire activity has a distinct seasonality linked to the dry seasons in Africa. Burning is most intense in the sub-Saharan regions in Northern Africa from November through May while from May through October, the savannah regions in Southern Africa are the most active fire sources (Giglio et al., 2006; Roberts et al., 2009; Van der Werf et al., 2003). African smoke can be transported to Barbados from North Africa in winter and spring (Quinn et al., 2022; Royer et al., 2023; Wex et al., 2016) and, less frequently, from Southern Africa in fall (Trapp et al., 2010). Conditions thought to be related to African climate (e.g., the North Atlantic Oscillation and the position of the Azores High) can cause large quantities of North African dust (and smoke) to be transported during the winter and spring (Chiapello et al., 2005; Doherty et al., 2008, 2012) when dust is also carried to northeastern South America (Barkley et al., 2019; Prospero et al., 1981, 2014).

Here we highlight different trends in nitrate and sulfate aerosol over the remote North Atlantic marine boundary layer and relate them to changing emissions. We then compare our observations to simulated concentrations of nitrate and sulfate aerosol using the CMAQ model from EQUATES, which was chosen due to its skill at modeling changes in nitrate and sulfate chemistry within the U.S. (Benish et al., 2022) as well as its ability to simulate constituents and sources of air pollution in remote regions, such as Dhaka, Bangladesh (Golam Sarwar et al., 2023). Our results highlight the importance of long-term atmospheric observations to understand the net outcome of changing global SO_2 and NO_x emissions on both the aerosol burden as well as air quality in distant populations.

2. METHODS

2.1 Aerosol Collection at the Barbados Atmospheric Chemistry Observatory: Aerosols were collected daily at the University of Miami's Barbados Atmospheric Chemistry Observatory (UM

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BACO: https://baco.rsmas.miami.edu/) located at Ragged Point, Barbados—the easternmost promontory on the east coast of the island (13.16504N, 59.43207W). The site has been operated by UM since 1971, and aerosol data has been used to document the long-range transport of African dust to the Caribbean and the Americas carried by the easterly trade winds as it is the first landmass encountered by African emissions transported across the Atlantic Ocean (Prospero et al., 2021).

Since 1989, aerosols have been collected at the top of a 17-m sampling tower that stands atop a 30-m bluff (see Fig 1).

Figure 1: Photo of the Barbados Atmospheric Chemistry Observatory (BACO) including the 17-m sampling tower and two shipping container laboratories.

A high-volume sampler pulls air at a nominal rate of 0.75 m^3/minute across a 20cm x 25cm Whatman-41 filter. Mass collection efficiencies are 90% for sulfate, 95% for nitrate, and 99% for dust (Savoie & Prospero, 1982). Filters are then folded into quarters under a laminar flow hood, placed in a clean Ziploc bag, and periodically shipped to UM for processing. To ensure that local island emissions are not sampled, the sampling pump is only operational when the wind blows directly from the ocean with speeds greater than 1 m/s. A timer is used to record the "run time", the total amount of time that the sampling pumps are on during the sampling interval between filter changes. Data with a run time less than 10% of the sampling interval are discarded—these data account for less than 10% of all data collected over the record highlighting the steady easterly winds measured at the site year-round. Procedural blanks are collected weekly by placing a Whatman-41 filter in the filter cassette with the sampling pump off for 15 minutes then placing the filter in a clean Ziploc bag; the blank is subsequently processed along with the daily filter samples.

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This work focuses on aerosol measurements conducted over 1990-2011, a period that overlaps with the implementation of more stringent air quality policies in the U.S. and Europe. Seasonal trends are also shown where winter is represented by December, January, February (DJF); spring is March, April, May (MAM); summer is June, July, August (JJA); fall is September, October, November (SON). African dust peaks annually at BACO in JJA with episodic transport in DJF and MAM in some years (Prospero & Mayol-Bracero, 2013; Quinn et al., 2022; Royer et al., 2023).

2.2 Quantification of Dust and Soluble Ion Mass Concentrations: A one-quarter filter section is extracted three times with a total volume of 20 mL of Milli-Q water to remove soluble material. The extracted filter is placed in a combustion oven (500˚C) overnight. The resulting ash is weighed ($m_{filter\ ash}$) and corrected for ash present from performing the same technique on the procedural blank ($m_{procedural\ blank}$). The gross ash weight is adjusted by a factor of 1.3 to account for the volatilization of water and minerals during the combustion process (Equation 1, Prospero, 1999; Zuidema et al., 2019). This corrected ash mass concentration is equated to mineral dust concentrations present on the filter based on previous comparisons between filter ash and concentrations of aluminum, a tracer for mineral dust (Prospero, 1999).

$$Dust = (m_{filter\ ash} - m_{procedural\ blank\ ash}) * 1.3 \quad (1)$$

The filtrate from the washing process is used to quantify soluble ion concentrations. Anions (e.g., chloride (Cl^-), nitrate (NO_3^-), and sulfate (SO_4^{2-})) were measured using ion chromatography (IC). Cations (sodium (Na^+), potassium (K^+), and calcium (Ca^{2+})) were measured with a flame photometer after 2004 while flame atomic absorption spectrophotometry was used prior to 2004 limiting cation analysis to sodium (Savoie et al., 2002). In addition to total soluble ion concentrations, we also report concentrations of non-sea salt (nss) sulfate, which is a useful tracer of sulfur from marine biogenic and pollution emissions, and nss-potassium, a tracer of biomass burning emissions (Andreae, 1983; Keene et al., 1986). Concentrations of nss-SO_4^{2-} and nss-K^+ are calculated using the following mass-based equations and assuming that Na^+ is a conservative tracer of sea spray aerosol:

$$nss - SO_4^{2-} = [SO_4^{2-}] - (0.2517 * [Na^+]) \quad (2)$$

$$nss - K^+ = [K^+] - (0.03595 * [Na^+]) \quad (3)$$

2.3 EQUATES Model Products

EPA's Air QUAlity TimE Series (EQUATES) Project uses the Community Multiscale Air Quality (CMAQ) model, a 3-D chemical transport air quality model, to simulate air quality over a continuous 2002-2019 time period (Foley et al., 2023). CMAQ accounts for gas, cloud, and aerosol chemistry, including processes such as in-cloud sulfate formation from the oxidation of SO_2. EQUATES uses CMAQv5.3.2 (Appel et al., 2021) to model the Northern Hemisphere using 108-km resolution and 44 vertical layers (Mathur et al., 2017). Meteorological data is predicted using the Weather Research and Forecasting model (WRFv.4.1.1). Emissions from outside the contiguous U.S. and China are generated using the Hemispheric Transport of Air Pollution version 2 inventory for the year 2010 (HTAPv2.2) and are scaled to other years using the Community Emissions Data System (CEDS) for the years 2002-2019. The Fire INventory from NCAR (FINN)

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is used to generate biomass burning emissions (Wiedinmyer et al., 2011), lightning NOx emissions are derived from the Global Emissions IniAtive (GEIA), biogenic volatile organic compounds (VOCs) are from MEGAN2, and soil NO_x is from CAMSv2.1.

In this study, EQUATES was used to better understand observed trends, namely in nss-sulfate, post 2000 when concentrations unexpectedly increased. The model product is not available for the years prior to 2000. The analysis focused on dust, sea spray, nitrate, sulfate, and gaseous SO_2 and NO_2. In addition to anthropogenic sources of SO_2, natural sources such as dimethyl sulfide (DMS) were included in model runs. Species predictions were extracted from the lowest CMAQ model layer (~10m in thickness) for a receptor area over the Atlantic Ocean to the east of the island from 59.5627°W to 56.5448°W longitude and 14.3989°N to 11.4566°N latitude (equivalent to 16 grid cells). Simulated concentrations of aerosol sulfate, nitrate, calcium, potassium, and sodium were obtained for fine mode aerosol (e.g., Aitken and accumulation mode) and coarse mode aerosol. Previous studies have shown that most of the aerosol mass at Ragged Point is below 10 μm diameter (Prospero et al., 2001). Because aerosol filters collected at BACO capture total suspended particulate matter, model outputs of fine and coarse mode aerosol concentrations were combined to give total aerosol mass concentrations of sulfate, nitrate, Na^+, K^+, and Ca^{2+}. Concentrations of nss-sulfate and nss-K^+ were calculated using Equations 2 and 3, respectively, and model outputs of total Na^+ mass concentrations, total sulfate mass concentrations, and total K^+ mass concentrations. We calculated concentrations of nss-Ca^{2+}, which has been shown to be a good tracer for mineral dust in Barbados (Savoie & Prospero, 1980), and dust mass concentrations were then calculated using the upper crustal abundance of Ca^{2+} in soil (4.1%) (Taylor & McLennan, 1985) as shown in Equations 4 and 5.

$$nss - Ca^{2+} = [Ca^{2+}] - (0.0376 * [Na^+]) \quad (4)$$

$$Dust = [nss - Ca^{2+}] * 24.1 \quad (5)$$

We first assessed the ability of the EQUATES CMAQ simulations to capture trends in different aerosol types observed at Ragged Point. Simulations of dust and sodium from EQUATES capture seasonal and monthly observed trends in dust (see Fig S1 and S2 of the Supporting Information (SI)). However, the model overpredicts sodium (Na^+, a proxy for sea spray) by a factor of 3-4 (Fig S2) and underpredicts dust by an average factor of ~7. This low bias for dust in CMAQv5.3.2 is consistent with CMAQ development that occurred after the EQUATES simulations were complete. A bugfix to the online dust emissions module in CMAQv5.4 increases dust emissions by a factor of 3-7 over the Sahara Dessert and parts of Asia (see the CMAQv5.4 release notes for further information; https://www.epa.gov/cmaq/cmaq-documentation#release-notes).

In addition to simulating observed aerosol concentrations, EQUATES was also used to examine trends in gaseous indicators of anthropogenic and biomass burning emissions, oxidant concentrations and oxidant ratios important for investigating changes in the oxidation efficiency of pollutant gases and the subsequent formation of nitrate and sulfate aerosol. Further, EQUATES was used to investigate whether the oxidation efficiency of SO_2(g) changed during the 2002-2011 period. The oxidation ratio was calculated from Equation 6 (Shah et al., 2018):

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$$Oxidation\ Ratio = \frac{nss{-}SO_4^{2-}}{nss{-}SO_4^{2-}+SO_2} \quad (6)$$

For this calculation, we used EQUATES model data for SO_2(g) concentrations and filter-based observations of nss-sulfate.

2.4 HYSPLIT Back-trajectory Analysis

Air mass back-trajectory analysis was performed using NOAA's Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler & Rolph, 2011; Rolph et al., 2017; Stein et al., 2015). 315-h back-trajectories were initiated at heights of 500, 1000, and 2000-m to capture both SAL and boundary layer transport. Frequency plots were also generated to illustrate seasonal differences in air mass transport and origin. We used global National Center for Environmental Prediction (NCEP) reanalysis data that goes back in time far enough to examine trajectories from our dataset (Kramer et al., 2020). The finer resolution Global Data Assimilation System (GDAS) dataset (1° resolution) was used to examine trends in 2008, 2009, and 2010 when transport conditions strongly impacted nitrate aerosol concentrations.

3. RESULTS

3.1 Measured Trends from 1990-2011 at Ragged Point: Nitrate has remained constant while NSS-Sulfate initially declined then subsequently increased.

Figure 2 shows yearly measured average mass concentrations of non-sea salt (nss) sulfate and nitrate from 1990 through 2011. Nitrate concentrations are remarkably stable from 1990 to 2011 (R^2=0.006, p>0.05 (not significant)) with an average concentration of 0.59 ug/m^3 ± 0.04 ug/m^3. However, two anomalous peaks in nitrate are observed in 2008 and 2010 with annual average nitrate concentrations of 0.73 and 0.81 ug/m^3, respectively. In contrast, nss-sulfate decreases by 30% from an average concentration of 0.84 ug/m^3 starting in 1990 to a minimum of 0.64 ug/m^3 in 2000 at a rate of 0.022 ug/m^3/yr. Subsequently, sulfate gradually increased to 0.91 ug/m^3 in 2010 and 0.90 ug/m^3 in 2011, maximums across the entire record. The trends in the yearly average mass concentrations of nitrate and nss-sulfate are significantly different (p-value < 0.005), which can be explained by either different sources or different rates of change for precursor NO_x and SO_2 emissions or different responses in aerosol production to changing emissions (Shah et al., 2018; Vasilakos et al., 2018).

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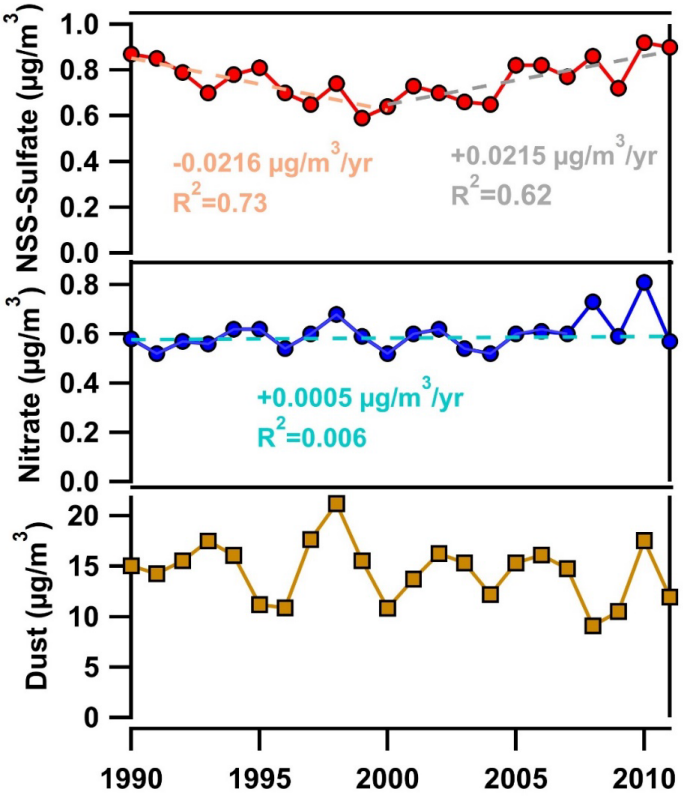

Figure 2: Yearly averages of non-sea salt sulfate (NSS Sulfate, red line, top panel), nitrate (middle panel, blue line), and dust (bottom panel, brown line with squares) measured at Ragged Point, Barbados from 1990-2011. Dashed lines show changes in NSS Sulfate pre-2000 (orange), post-2000 (gray line) and for nitrate (teal line) without the spikes in 2008 and 2010 considered.

The Barbados yearly trends differ from long-term observations of aerosol and precipitation chemistry measured at Tudor Hill, a site on the west coast of Bermuda, from 1989-1997 and from 2006-2009 as part of the same program as that at Barbados and using the same protocols including sampling only when winds are easterly and over the ocean (Keene et al., 2014; Savoie, et al., 2002). Aerosol nitrate is constant at both sites; however, the nitrate annual mean at Bermuda is ~1.05 ug/m^3, roughly double the nitrate observed at Ragged Point (Keene et al., 2014; Savoie, et al., 2002) while the decline in nss-sulfate observed in Bermuda from ~2.59 ug/m^3 in 1989 to ~1.63 ug/m^3 in 2009 is also greater than our observations at Ragged Point (Keene et al., 2014). Furthermore, sulfate aerosol in Bermuda declines over the entire record and does not exhibit the same reversal in the 2000s that we observe at Barbados. The difference in concentrations and trend in nss-sulfate observed at Ragged Point compared to that at Bermuda is not entirely surprising given that the Bermuda site is more directly influenced by anthropogenic emissions from the U.S. compared to Barbados, which is a more remote site influenced by a multitude of emission sources (Savoie, et al., 2002).

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To assess whether annual trends in nss-sulfate and nitrate observed in Barbados are associated with African aerosol transport conditions, annual average measured dust mass concentrations are also shown in Fig 2. Annually averaged dust concentrations show no appreciable increase or decrease from 1990-2011. Therefore, nss-sulfate aerosol shows no correlation with African dust mass concentrations. However, nitrate aerosol is modestly correlated with dust (R^2=0.3). Comparing seasonal nitrate and dust mass concentrations by year reveals tighter correlations between nitrate and dust for DJF and MAM (0.46 and 0.4, respectively, see Fig S3). DJF and MAM are not the peak dust transport seasons to the Caribbean, but they are the seasons that favor co-transport of dust and biomass burning emissions from Northern Africa (Royer et al., 2023).

3.1.2 Seasonal Patterns in Air Mass Trajectories and Nitrate Concentrations: Transport of African biomass burning likely explains elevated nitrate concentrations

HYSPLIT air mass back-trajectories reveal similar seasonal patterns each year with predominantly easterly transport to the site year-round (see Fig S4, which shows representative 5-day air mass back-trajectory frequency plots). DJF has some trajectories that intercept North America and some from the northern part of the Atlantic Ocean toward Europe, MAM trajectories are easterly, JJA trajectories are from the northeast along the coast of Africa, and SON trajectories are from the east with some from the southeast close to the coast of South America.

Figure 3 shows trends in nss-sulfate and nitrate during different seasons (e.g., DJF, MAM, JJA, and SON). Increases in nitrate in 2008 are driven by high nitrate concentrations in JJA and SON in 2008, with most of the increase in September, while high nitrate levels in 2010 are primarily observed during MAM. In 2010, a transition from an El Niño to the strongest La Niña event on record occurred (Wolter & Timlin, 2011; Zhang et al., 2019), and long-range transport from Africa was anomalously high during the springtime as evidenced by high mass concentrations of dust in the spring of 2010 (Zuidema et al., 2019). Daily air mass back-trajectories passed more frequently over the African continent in MAM of 2010 compared to MAM of 2009 (26 vs 6 days) (Fig 4a and b), and nitrate levels exceeded 1 ug/m^3 on over half of those days when trajectories traversed the N African continent. Figure 4 focuses on trajectories initiated at 500-m height, and if trajectories initiated at 1000-m are also included, then transport over the N African continent occurs on 36 days in MAM of 2010 or 82% of days when nitrate levels exceeded 1 ug/m^3. However, during the summertime peak (JJA) in African dust transport in 2010, nitrate does not show the same increase as dust despite frequent transport from N Africa (see Fig 4d). Further, Fig 4c and d compares daily and monthly mean concentrations of nitrate, dust, and non-sea salt potassium (e.g., nss-K^+), which is a marker for biomass burning emissions (Andreae & Merlet, 2001). Nitrate and nss-K^+ clearly track each other and are both elevated in the spring of 2010. Although biomass burning peaks in December and January while MAM is the tail end of the burn season in North Africa (Giglio et al., 2013; Roberts et al., 2009), the high nitrate loadings observed in 2010 in spring are likely due to strongly favorable transport conditions that transport both dust and smoke to Barbados during this year.

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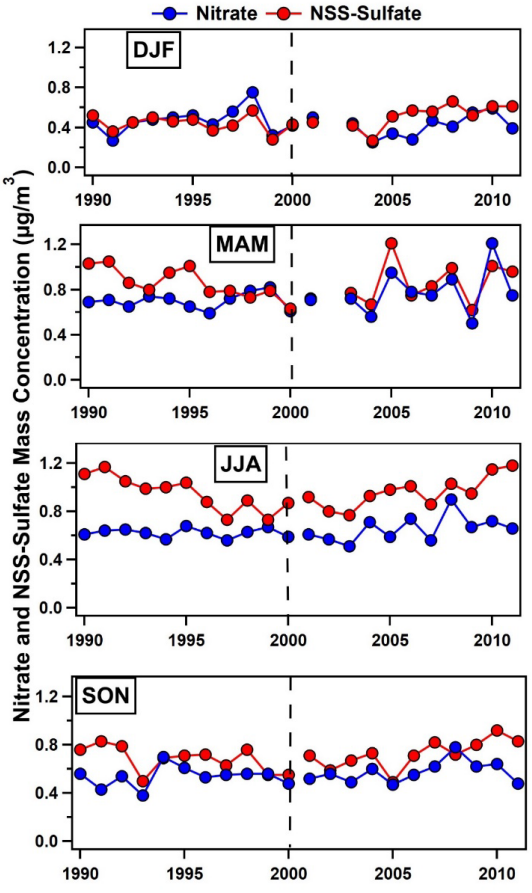

Figure 3: Average yearly concentrations of nitrate (blue markers), and nss-sulfate (red markers) for winter (DJF), spring (MAM), summer (JJA), and fall (SON). The black dashed line denotes the year 2000 when nss-sulfate trends shifted from decreasing to increasing.

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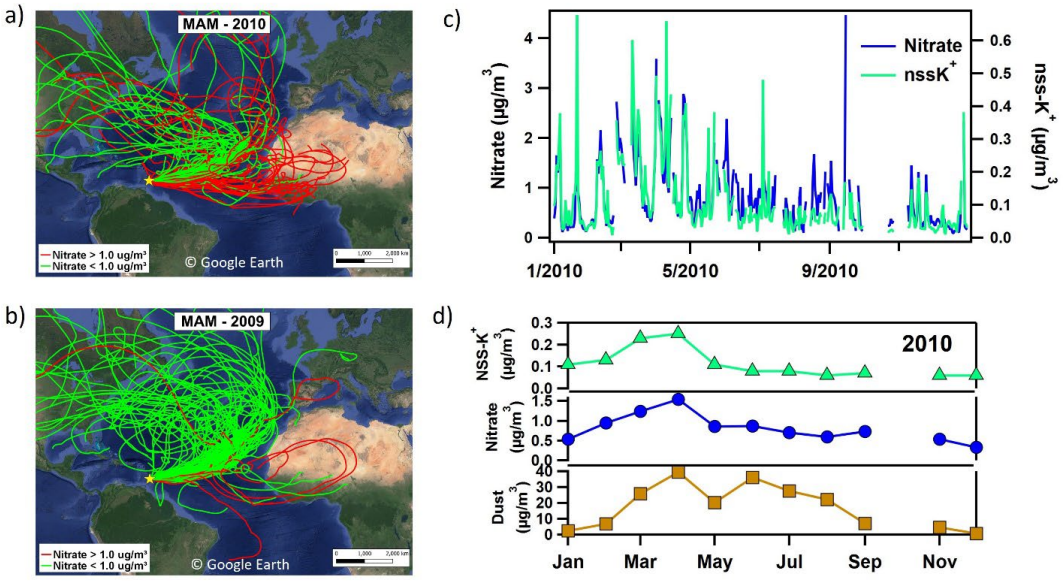

Figure 4: 315-h air mass back-trajectories initiated at 500-m for MAM a) 2010 and b) 2009. Back-trajectories labeled in green are for days where nitrate concentrations measured at Ragged Point were < 1 ug/m^3 while trajectories labeled in red had nitrate > 1 ug/m^3 (from Google Earth). c) Daily comparison of nitrate and non-sea salt potassium (nss-K$^+$), a biomass burning marker, show a tight correlation (R^2=0.62). d) Monthly average concentrations of dust (brown), nitrate (blue), and nss-K$^+$ (green) for 2010.

Similar to 2010, air mass back-trajectories traversed the African continent on just 40% of the days in JJA but 82% of days in September of 2008 with nitrate concentrations > 1 ug/m^3 (Fig S5 and Fig 5a). If trajectories initiated at the 1000-m level are included, then African transport occurs on 64% of days in JJA with nitrate concentrations > 1 ug/m^3. In JJA, most of the air mass back-trajectories pass through the N African continent. However, in September, trajectories take a more southerly route traversing near the South American coast, sub-Saharan Africa, and southern Africa (Fig 5a). Figure 5b shows trajectories for September but in 2009, when fewer trajectories take a southerly route and nitrate concentrations rarely exceed 1 ug/m^3. September is during the peak of the burn season in sub-Saharan and southern Africa and also dovetails with strong smoke transport over the Atlantic Ocean due to the increased intensity of the African Easterly Jet (Adams et al., 2012; Adebiyi & Zuidema, 2016; Zuidema et al., 2018). Air mass back-trajectories in September from southern and sub-Saharan Africa to our measurement sites in Barbados and Cayenne have been linked with African smoke transport (Barkley et al., 2019; Trapp et al., 2010) further suggesting a link between elevated concentrations of nitrate measured at Barbados with transported African smoke. Further, biomass burning in the Amazon also peaks in SON (Adams et al., 2012) and could also explain the increase in nitrate in this season in 2008.

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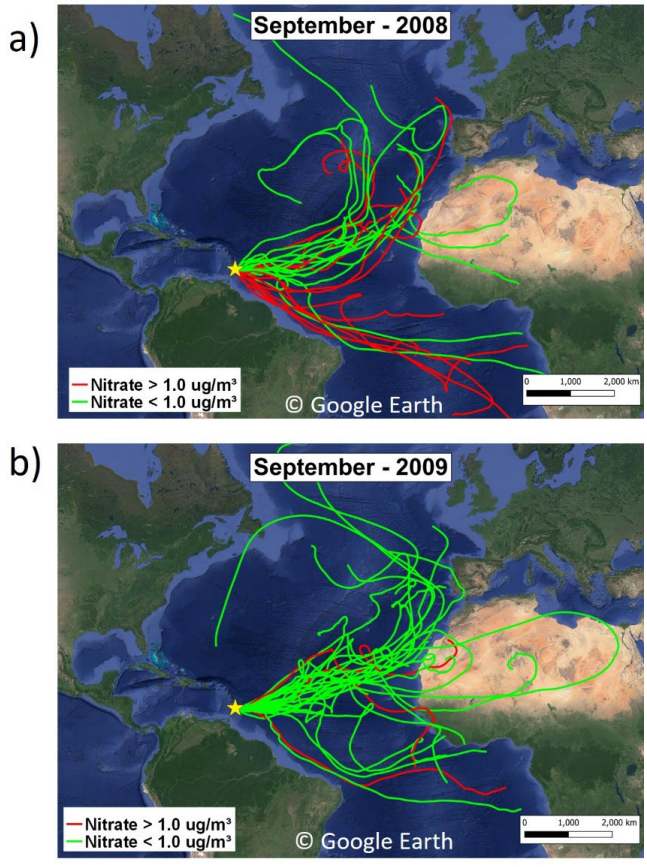

Figure 5: 315-h air mass back-trajectories initiated at 500-m for September a) 2008 and b) 2009. Back-trajectories labeled in green are for days where nitrate concentrations measured at Ragged Point were < 1 ug/m^3 while trajectories labeled in red had nitrate > 1 ug/m^3 (from Google Earth).

3.1.3 Seasonal Patterns in Non-Sea Salt-Sulfate, Impacts of SO_2 Reductions in the US and EU, and Possible Impacts from SO_2 Increases in Africa

Figure 3 reveals that the decrease in sulfate and subsequent increase are measured year-round during all seasons. Table 1 further provides correlation coefficients and the rate of change in either nitrate or sulfate in μg m^{-3}yr^{-1} for data collected pre- and post-2000 when the trend in nss-sulfate changed. Pre-2000, nss-sulfate shows a consistent decline in each season except for winter with the strongest reduction measured in JJA at -0.037 μg m^{-3}yr^{-1} (R^2=0.72, Table 1). Post-2000, nss-sulfate increased during every season except MAM with the highest increases again in JJA at a similar rate to the pre-2000 decline at +0.030 μg m^{-3}yr^{-1} (R^2=0.61, Table 1). In contrast, nitrate shows no trend (e.g., R^2<0.2) in any season except a slight increase of 0.015 μg m^{-3}yr^{-1} post-2000 in JJA. Unlike nitrate which showed intermittent spikes associated with increased biomass burning

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aerosol transport periods, the persistent increases in nss-sulfate observed during all seasons more likely reflect either increased emissions or more efficient oxidation of SO_2.

Table 1: Seasonal trends in nitrate and nss-sulfate shown pre- and post-2000. The rate of change in ug/m^3/yr is shown and the correlation coefficient is also included. Values of R^2<0.2 were denoted as having "No Trend". DJF represents winter, MAM represents spring, JJA represents summer, and SON represents fall.

Season	Compound	Pre-2000 Rate of Change (ug/m3/yr)	Post-2000 Rate of Change (ug/m3/yr)
DJF	Nitrate	+0.0088 (R2=0.05, No Trend)	+0.0094 (R2=0.07, No Trend)
	NSS-Sulfate	-0.0055 (R2=0.05, No Trend)	+0.025 (R2=0.48)
MAM	Nitrate	+0.0023 (R2=0.01, No Trend)	+0.018 (R2=0.08, No Trend)
	NSS-Sulfate	-0.033 (R2=0.64)	+0.017 (R2=0.09, No Trend)
JJA	Nitrate	-0.0015 (R2=0.02, No Trend)	+0.015 (R2=0.21)
	NSS-Sulfate	-0.037 (R2=0.72)	+0.030 (R2=0.61)
SON	Nitrate	+0.0031 (R2=0.02, No Trend)	+0.01 (R2=0.13, No Trend)
	NSS-Sulfate	-0.018 (R2=0.29)	+0.024 (R2=0.44)

Figure 6 compares yearly trends in Ragged Point nss-sulfate along with SO_2 emissions reported from the Community Emissions Data System (CEDS) (McDuffie et al., 2020). We focus on the most likely sources to impact Ragged Point including emissions from the US, EU, and Africa. A similar figure comparing nitrate concentrations measured at Ragged Point and nitrogen dioxide (NO_2) emissions from CEDS can be found in the SI (Fig S6). Our near constant nitrate mass concentrations do not match the decline in NO_2 observed in the EU and US and increase in NO_2 in Africa. In contrast, decreases in nss-sulfate observed from 1990-2000 at Ragged Point closely follow the 32% and 58% reductions of SO_2 emissions in the U.S. and Europe, respectively (Fig 6) (Aas et al., 2019; Hand et al., 2012; McDuffie et al., 2020; Rafaj et al., 2015; Yang et al., 2020). Our finding that changing SO_2 emissions in the EU and US is reflected in our record at Ragged Point is in agreement with previous work examining both anthropogenic and biogenic sulfate (Savoie, et al., 2002). Figure 6 also compares the recovery of nss-sulfate observed at Ragged Point after 2000 to increasing emissions of SO_2 from Africa where emissions of SO_2 increased by 37% (McDuffie et al., 2020). We next utilized the CMAQ model results from EQUATES to gain further insight into the observed recovery of nss-sulfate (post-2000) and determine if other factors such as changes in the oxidation efficiency of SO_2 and meteorological changes and emissions from smoke or anthropogenic emissions affected our observations.

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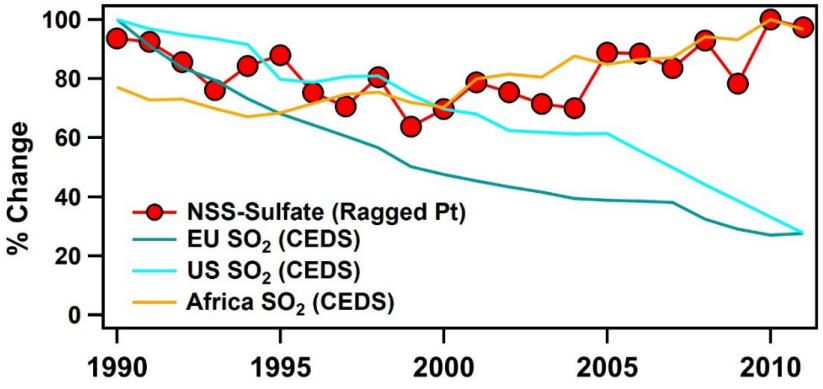

Figure 6: NSS-Sulfate mass concentrations measured at Ragged Point from 1990-2011 are shown in red. NSS-Sulfate mass decreases from 0.87 ug/m^3 in 1990 to 0.64 ug/m^3 in 2000 and increases again to 0.90 ug/m^3. Emissions of sulfur dioxide (SO_2) from the CEDS emissions inventory from McDuffie et al., 2020 are included for comparison. Decreasing emissions of SO_2 from the US and EU are shown in blue lines. U.S. SO_2 reduces from 21.12 to 5.85 Tg S/yr and EU SO_2 reduces from 28.06 to 7.74 Tg S/yr. SO_2 emissions from Africa increase from 4.13 to 5.95 Tg S/yr.

3.2 Comparison of Measured and Modeled Trends of Nitrate and Sulfate Aerosol

Monthly concentrations of simulated nss-sulfate and nitrate (for both the fine and coarse mode combined) were compared with mass concentrations measured on filters collected at Ragged Point (see Fig 7 and 8, respectively). While EQUATES predicts that nss-sulfate is almost exclusively in the fine mode, nitrate concentrations are split between the fine and coarse aerosol sizes with increased fine nitrate in DJF (40%) and MAM (26%) compared to JJA and SON (18% for both seasons, see Fig S7) likely due to increased contributions of nitrate from fine biomass burning aerosol from northern Africa in winter and spring.

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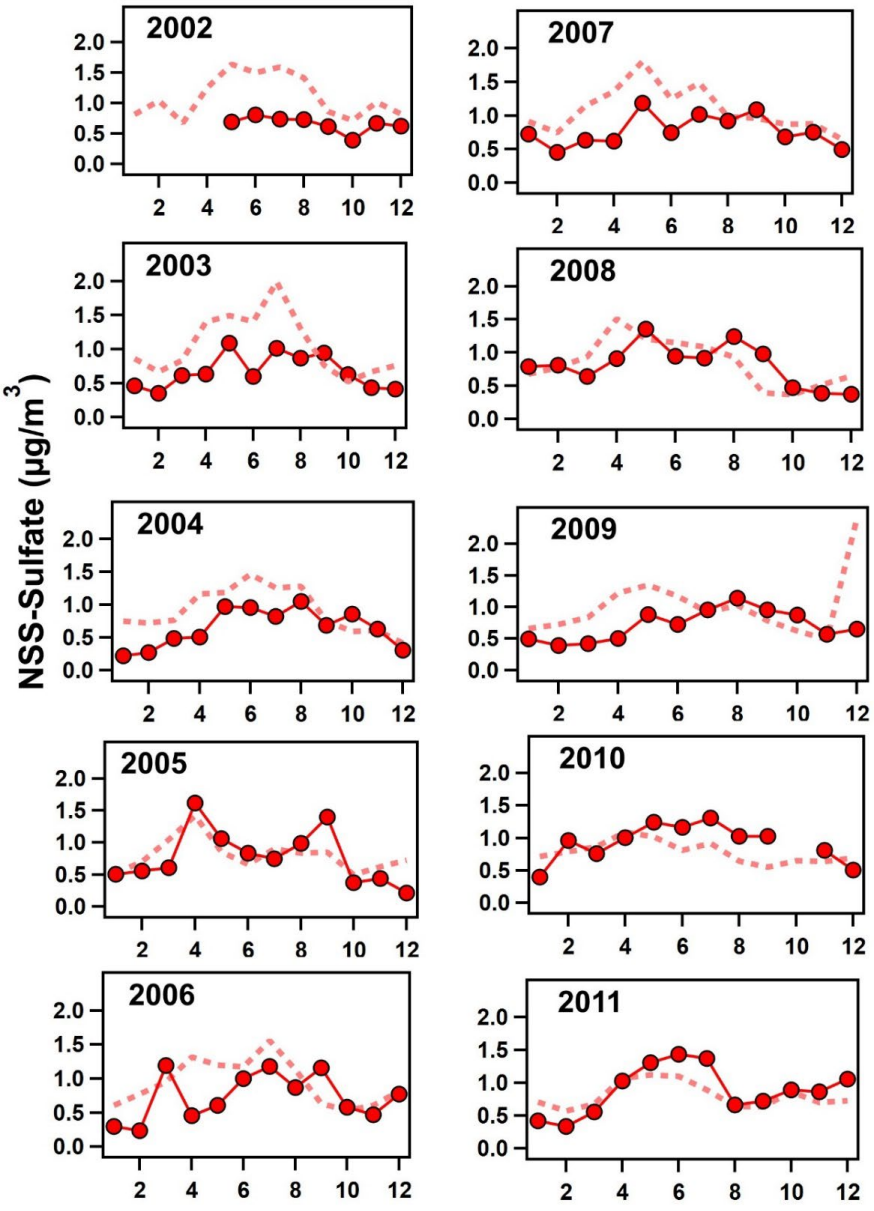

Figure 7: Monthly averages of non-sea salt sulfate (NSS-sulfate) mass concentrations measured at Barbados on filters (solid red line with red circles) compared to monthly averages of NSS-sulfate calculated from EQUATES model simulations of combined fine and coarse mode sodium (Na) and sulfate using equation 2 (dashed red line) for 2002-2011.

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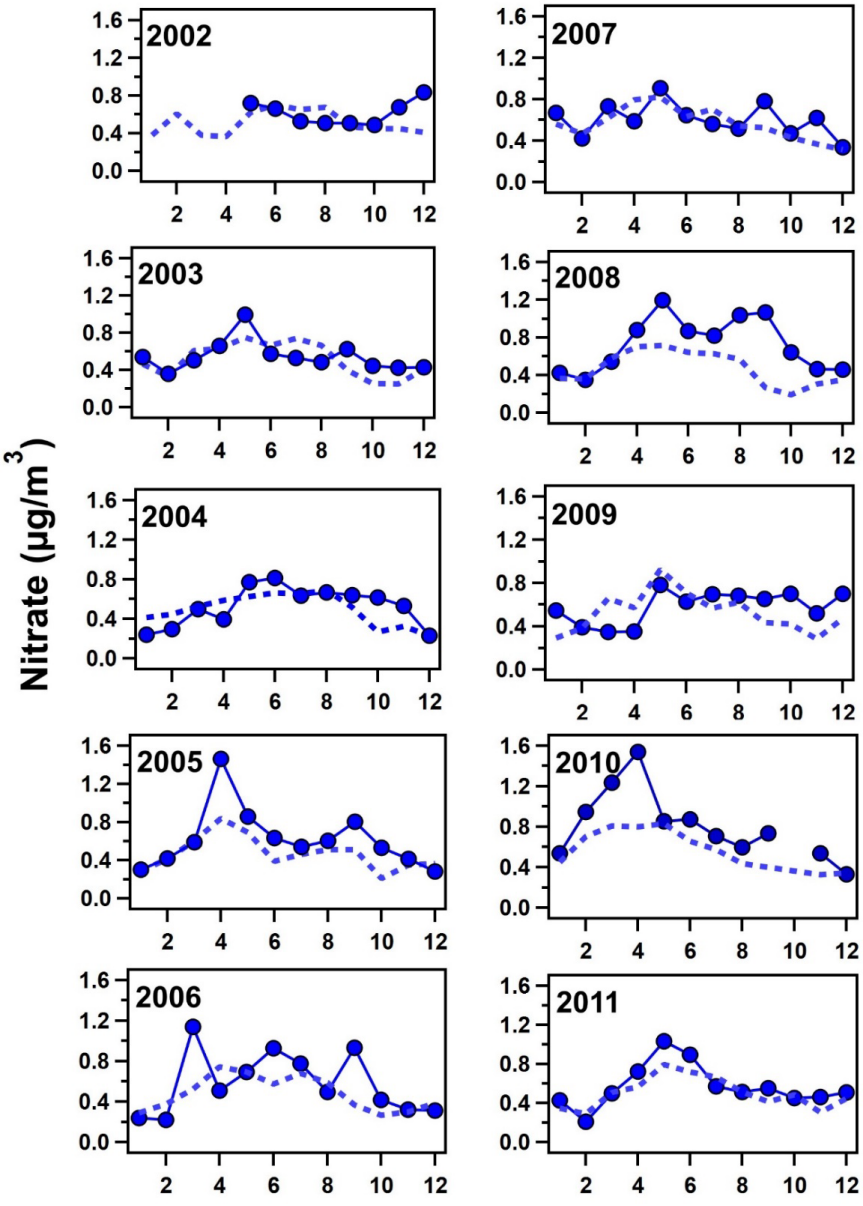

Figure 8: Monthly averages of nitrate mass concentrations measured in Barbados on filters (solid blue line with blue circles) compared to monthly averages of combined fine and coarse mode nitrate calculated from EQUATES model simulations (dashed blue line) for 2002-2011.

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To assess the performance of the model compared to our observations, we calculated the normalized mean bias (NMB) and Pearson correlation coefficient (r) for nss-sulfate and nitrate (see Table S1 (Boylan & Russell, 2006)). Additional calculations of the mean bias (MB) and root-mean-square error (RMSE) can also be found in Table S1 of the SI. NMB for nitrate was generally within ±20%, better than predictions of nitrate within the U.S. (Kelly et al., 2019), with the exception of 2008 (-35.06%) and 2010 (-28.72%) when the model underpredicted measurements likely due to the elevated African smoke transport events. The model overpredicts nss-sulfate in the earlier years (2002-2007) then converges closer to our measurements at Ragged Point after 2008 as shown in Fig 7. The NMB for nss-sulfate also reflects this trend as high values are observed in 2002 (+81.45%) then the model beings to fall within ±20% starting in 2008 (see Table S1). As such, trends in nss-sulfate simulated by EQUATES show a decrease over time rather than an increase post-2000 (see Fig S8). The decrease in EQUATES simulations of nss-sulfate post-2000 compared to the increase observed in Barbados could be related to a changing bias over time. For example, recent predictions for 2019 indicate CMAQ underpredicts sulfate by about 50% in the eastern U.S. (45% underestimate across entire U.S.) (Vannucci et al., 2024) while previous simulations for 2002 and 2016 indicated more modest normalized mean biases of 20% or less (Appel et al., 2021; Sarwar et al., 2011). As a result, EQUATES may simulate a stronger decline in transported U.S. sulfate over 2002-2019 than observations indicate, potentially masking increasing signatures from other regions. Pearson correlation coefficients range from -0.22 to 0.88 by year for nitrate with a mean of 0.54. The poorest correlation (-0.22) occurs for 2002 when filter observations were limited. For nss-sulfate, r ranges from 0.23 to 0.82 by year with a mean of 0.59. The greatest variation between the model and measurements is for 2009 (see Fig 7), which was also the year that had an erroneously high concentration of sea spray aerosol in winter. Overall, the model is capturing both the magnitude and seasonal and interannual variation of nss-sulfate and nitrate at our remote location in the tropical North Atlantic Basin.

3.2.1 Using EQUATES to Understand the Recovery in Aerosol Sulfate

Figure 9 shows a decrease in $SO_2(g)$ in the region near Barbados and an increase in the oxidation ratio of $SO_2(g)$, consistent with a predicted decrease in $SO_2(g)$ and the observed increase in nss-sulfate. Hydrogen peroxide (H_2O_2) concentrations also increased over this period (see Fig 10) and is likely linked to decreases in $SO_2(g)$, which is a major sink of H_2O_2 during the aqueous phase formation of sulfate (Manktelow et al., 2007). We note that the predicted oxidation ratios calculated are likely an overestimate because EQUATES overpredicts nss-sulfate compared to observations and the overprediction reduces with time.

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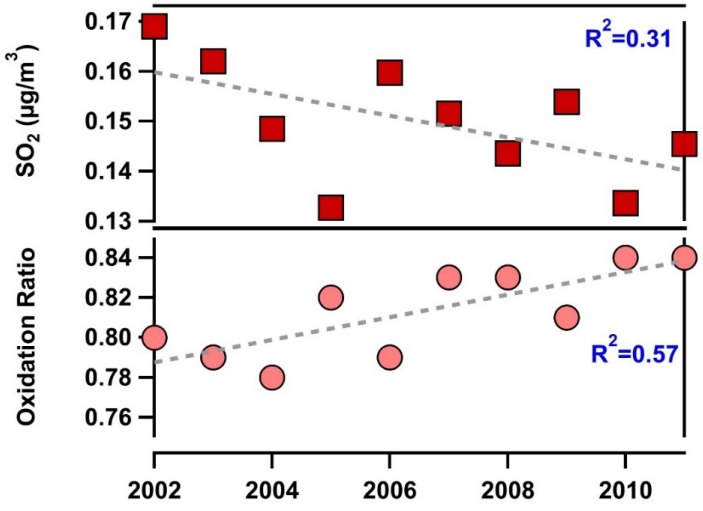

Figure 9: Annual trends in sulfur dioxide (SO_2) simulated by the EQUATES model and the calculated oxidation ratio for Ragged Point. Linear fits and corresponding correlation coefficients are also shown.

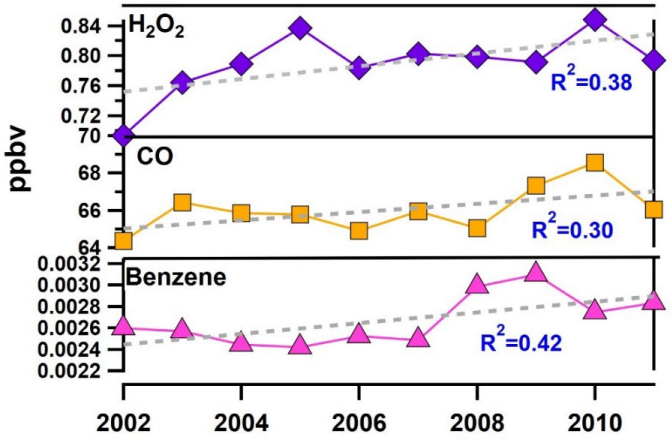

Figure 10: Annual trends in select gas phase species including benzene (pink trace, bottom panel), carbon monoxide (CO) (orange trace, middle panel), and hydrogen peroxide (H_2O_2) (purple trace, top panel) simulated by the EQUATES model at Ragged Point. Linear fits and corresponding correlation coefficients are also shown.

Changes in biomass burning, anthropogenic emissions, oxidant concentrations, and meteorological parameters were also investigated using EQUATES. Fine mode non-sea salt potassium (nss-K^+) was used as our tracer for smoke emissions. We used EQUATES data rather than our nss-K^+ observations, which were non-existent for most years from 2002-2011. No

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significant increase in nss-K^+ was observed from our analysis (Fig S9). Meteorological parameters (temperature, RH, wind speed, wind direction) were constant over time showing no shift in rainfall, winds, temperature, or RH (Fig S9). Concentrations of the hydroxyl radical ($^{\bullet}OH$) did not show a significant change from 2002-2011 (Fig S9). EQUATES concentrations of benzene, a tracer for fossil fuel combustion, and carbon monoxide (CO), a tracer for combustion from both fossil fuels and biomass burning, both increase from 2002-2011 (Fig 10). Our results suggest that anthropogenic emissions in addition to an increase in the oxidation efficiency of SO_2(g) to sulfate are responsible for the increase of nss-sulfate post-2000.

4. DISCUSSION & CONCLUSIONS

Our over 20-year record (1990-2011) of nitrate and nss-sulfate aerosol shows two distinct trends over the tropical N Atlantic Basin. Nitrate shows no significant change other than two spikes in JJA and September of 2008 and MAM of 2010. Dust transport, particularly in DJF and MAM, explained interannual oscillations in nitrate concentrations while increased transport of northern African smoke in MAM 2010 and smoke from southern African and South America in September 2008 caused the increased levels of nitrate observed in those years. Nitrate has been shown to be enhanced in smoke by up to 5 fold over background due to high emissions of NO_x that are rapidly converted to nitrate (Adon et al., 2010; Hickman et al., 2021; Perron et al., 2022; Schlosser et al., 2017). Notably, nitrate was not enhanced in JJA of 2010 even though high quantities of dust were transported to Barbados. We speculate that the lack of enhanced nitrate during summer of 2010 is because smoke is not co-transported with the dust during the summer season. This finding in addition to our observations of enhanced nitrate associated with dust in DJF and MAM suggests that the nitrate associated with African aerosol transport primarily originates from NO_x emissions from African wildfires that are rapidly converted to nitrate prior to transport.

In contrast to the relatively flat trend in aerosol nitrate, nss-sulfate decreases by 30% from 1990-2000 then increases from 2000-2011, recovering to concentrations measured in the early 1990s. Reductions in nss-sulfate observed in Barbados are most likely due to decreased emissions of SO_2 in the US and EU due to clean air policies implemented via technologies such as flue-gas desulfurization devices installed in power plants (Aas et al., 2019; Kharol et al., 2017; Smith et al., 2011). Thus, our results highlight that regulations aimed to improve national and regional air quality also impact more distant locations such as the remote North Atlantic marine boundary layer and Caribbean.

As shown in Fig 6, increases in SO_2 in Africa, namely from anthropogenic sources, are the most probable cause for the increase in nss-sulfate levels from 2000-2011 in Barbados. Anthropogenic sources of SO_2 in Africa include emissions from electricity generation, diesel combustion and transportation (Assamoi & Liousse, 2010; Keita et al., 2021; Liousse et al., 2014), refineries, gas flaring, and smelting (Doumbia et al., 2019; Osuji & Avwiri, 2005). Levels of pollution in African cities are on par with Asian megacities with the largest rate of increases in SO_2 observed in western Africa (Adon et al., 2016; Hopkins et al., 2009; Liousse et al., 2014; Val et al., 2013). The industrial sector in the Highveld region of South Africa also shows some of the highest increases in SO_2 due to an increase in the number of coal-fired power plants arising from an increased demand for electricity (Keita et al., 2021; Liousse et al., 2014; Shikwambana et al.,

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2020). The emissions inventory for Africa is also likely underestimated due to a lack of measurements (McDuffie et al., 2020). Therefore, we speculate that anthropogenic SO_2 emissions are likely higher than shown from the CEDS model and are driving the observed increases in nss-sulfate in Barbados.

In addition to sources of SO_2 within Africa, SO_2 emissions from other nearby countries and regions have been shown to be exported to Africa (Koch et al., 2007). In particular, SO_2 emissions in India have rapidly risen and overtaken China as the largest emitter of SO_2 (Li et al., 2017) while remote sensing observations have highlighted that SO_2 emitted from oil and gas operations in the Persian Gulf have been greatly underestimated in emissions inventories (McLinden et al., 2016). It is possible that these two regions may also contribute to the increase in nss-sulfate observed at our measurement site.

While some of the increases in nss-sulfate after 2000 that occur in all seasons could be due, in part, to marine biogenic, shipping, biomass burning, and volcanic emissions, their contributions are likely not the dominant cause of the observed trends. Marine biogenic sulfate is estimated to contribute up to 50% of nss-sulfate at Ragged Point during non-dust transport conditions (Li-Jones & Prospero, 1998; Royer et al., 2023; Savoie, et al., 2002). However, our predictions of nss-sulfate concentrations with EQUATES includes DMS chemistry that is not explaining our observed trends. Further, a recent modeling study factoring in changes in phytoplankton dynamics found that DMS emissions have not appreciably changed from the preindustrial to the present-day (Wang et al., 2018). Shipping emissions likely do not explain our trends in nss-sulfate because Barbados is somewhat isolated from proximal shipping impacts—heavy shipping is concentrated in the Caribbean west of Barbados and along the north coast of South America (Czermański et al., 2021). Biomass burning has declined in northern Africa starting in the early 2000s (Andela et al., 2017; Niels Andela & Van Der Werf, 2014; Zubkova et al., 2019). Further, increased nss-sulfate has been observed year-round rather than just during the main burn seasons, suggesting that this source alone is likely not responsible for the major increases in nss-sulfate observed at our site. The largest natural source of SO_2 in Africa is volcanic emissions from Mount Nyiragongo in the Goma region of the Democratic Republic of Congo, which has been shown to impact sulfate aerosol at the Amazon Tall Tower Observatory (ATTO) in Brazil (Opio et al., 2021; Saturno et al., 2018). Volcanic emissions likely do impact nss-sulfate measured in Barbados but there is no evidence that emissions from this source are increasing. These lines of evidence further support an anthropogenic source of SO_2 as the cause of the increase in nss-sulfate observed in Barbados.

In addition to increased SO_2 emissions from North Africa, the oxidation ratio also increased from 2002-2011, which would more efficiently convert transported SO_2 emissions from Africa to sulfate. Further, concentrations of H_2O_2 increased in the post-2000 period indicating a potential increase in the efficiency of aqueous phase oxidation. We would expect that even if the total burden of SO_2 has been reduced globally (McDuffie et al., 2020; Smith et al., 2011), SO_2 emitted from North Africa is more efficiently converted to sulfate due to greater availability of oxidants at lower latitudes (Manktelow et al., 2007) and therefore have a greater impact on aerosol burdens over the Caribbean than SO_2 emitted from the US and EU. It is important to note, however, that the oxidation ratio calculated is most appropriate for accounting for changing oxidation efficiencies

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of SO_2 and sulfate formation near the site, and the oxidation ratio does not account for changes in the oxidation efficiency of already formed sulfate aerosol that has been transported to the site. For example, SO_2 emitted from Africa might be oxidized to form sulfate prior to being transported to our site.

One question that persists is why nitrate did not increase from 2000-2011 alongside the increase in nss-sulfate? One possible explanation is a combination of reduced NO_x from smoke concurrent with increased dust and smoke transport that offset any changes in nitrate other than the observed spikes in 2008 and 2010. In the 2000s, biomass burning emissions declined in northern equatorial Africa due to a combination of increased precipitation in DJF associated with a shift from more frequent El Niño events in the 1990s to more frequent La Niña events in the 2000s and land use practices converting tropical savanna to cropland (Andela et al., 2017; Andela & Van Der Werf, 2014; Hickman et al., 2021; Zubkova et al., 2019). The recently updated Barbados dust record highlights that dust is being transported to the Caribbean earlier in the year and arriving more frequently in spring (Zuidema et al., 2019), which would increase the transport of biomass burning emissions and associated nitrate to the Caribbean and remote North Atlantic, which may effectively cancel out the impact of reduced smoke emissions.

The Ragged Point site in Barbados has historically been associated with research on African dust transport (Prospero et al., 2021). However, this work highlights that the site is also an excellent indicator of long-term and large-scale changes in emissions and the impact of air quality policies or the lack of them or poor compliance to them. Looking forward, building upon the existing time series of nitrate and sulfate aerosol while also expanding the measurement capabilities at Ragged Point to incorporate measurements of metals, which will increase our ability to apportion aerosol sources, and carbonaceous aerosol will provide needed insight into the impact of anthropogenic and biomass burning on sulfate and nitrate burdens over the remote N. Atlantic that complement recent work performed in the S Atlantic (Zuidema et al., 2016, 2018).

Data Availability Statement: Measured nitrate, nss-sulfate, and sea salt concentrations will be put on the University of Miami's repository in addition to EQUATES simulations of nitrate, nss-sulfate, sea salt, gaseous tracers (SO_2, benzene, CO, H_2O_2) and meteorological parameters. Dust mass concentrations from Barbados can be found in the data repository for Zuidema et al., 2019. EQUATES data is available via the Remote Sensing Information Gateway (RSIG): https://www.epa.gov/hesc/remote-sensing-information-gateway.

Author Contribution: CJG analyzed measurement and model data and wrote the manuscript. JMP, LC, EB, PS collected data, operated the site, and analyzed filters for dust, sea salt, sulfate and nitrate mass concentrations. KF and HOTP provided EQUATES model simulations and helped with their interpretation. JAC performed HYSPLIT analysis. All authors read and edited the manuscript.

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Competing Interests: The authors declare that they have no conflict of interest.

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