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Vertical variability of aerosol properties and trace gases over a remote marine region: a case study over Bermuda

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Table S1. Summary of past studies relevant to aerosol particles, trace gases, and trajectory analysis for the Bermuda region.

Reference	Key Notes	Study Platform
Ahmad et al., 2010	Examined the new aerosol model for retrieval of atmospheric optical properties from SeaWiFS and MODIS sensor	Surface/Modeling
Altieri et al., 2013	Explored nitrate sources in ocean rainwater	Surface
Altieri et al., 2014	Investigated ammonium sources and N deposition in the ocean using rainwater samples with air mass history	Surface
Altieri et al., 2016	Analyzed rainwater and aerosol organic N concentrations and composition sources on Bermuda Island	Surface
Aldhaif et al., 2021	Investigated Bermuda aerosol climatology and cloud properties	Surface/Satellite
Andersson et al., 2013	Studied seawater CO_2 dynamics and sea–air CO_2 gas exchange rates during wintertime North Atlantic Ocean Eighteen Degree Water formation	Ship
Anderson et al., 1993	Assessed continental outflow's impact on western Atlantic ozone and small aerosol budgets	Aircraft
Anderson et al., 1996	Characterization of Bermuda tropospheric aerosol in fine fraction samples (<2 μm) using single particle and bulk aerosol analysis	Surface
Arimoto et al., 1992, 1995	Investigated anthropogenic and natural sources' contributions to non-sea salt sulfate budgets in aerosol particles were assessed using trace element data	Surface
Arimoto et al., 1997	Examined the link between dust mass-particle size distributions (MSDs) and atmospheric dust loadings using size-separated mineral aerosol samples	Surface
Arimoto et al., 1999	Analyzed Be and Pb activity in daily north Atlantic aerosol samples and meteorological data to determine how particle composition affects transport pathways and precipitation scavenging	Surface
Arimoto et al., 2003	Analyzed trace element recycling on atmospheric sea salt in Bermuda using aerosol particle data for Al, Fe, Na, Sb, Se V, and Zn and parameterized dry deposition velocities	Surface
Aryal et al., 2014	Compared surface and column optical properties	Surface/Column
Baker et al., 2017	Assess models and observations estimate of oceanic particulate dry nitrogen deposition	Ship/Modeling
Braun et al., 2021	Study examines atmospheric features of International Satellite Cloud Climatology Project weather states (WSs) across the Western North Atlantic Ocean	Satellite
Cornell et al., 1995	Examined atmospheric emissions of dissolved organic nitrogen to the oceans	Surface
Cornell et l., 1998	Examined urea in rainwater and atmospheric aerosol	Surface
Dadashazar et al., 2021	Evaluate aerosol mass concentrations and volume size distributions' seasonal sensitivity to accumulated precipitation along trajectories	Surface/Modeling
Harriss et al., 1984	Particulate transport via the atmosphere from North America to the North Atlantic Ocean	Aircraft
Hastie et al., 1988	Examined nitrogen and sulfur over the western Atlantic Ocean	Surface/Aircraft

Hegarty et al., 2010	Studied winter and summer continental influences on tropospheric O_3 and CO in the WNAO observed by TES	Satellite
Horvath et al., 1990	Determined the atmospheric aerosol's coarse mode using forward-scattering spectrometer probe data	Aircraft
Jickells et al., 1998	Investigated air-borne dust fluxes to deep-water sediment trap in the Sargasso Sea	Aircraft
Kawamura et al., 2017	Examined OC/EC, low molecular weight dicarboxylic acids, lipid class chemicals, sugars, and secondary organic aerosol (SOA) tracers in aerosol samples	Ship
Keene et al., 1986	Investigated sea-salt corrections and interpretation of constituent ratios in marine precipitation	Surface
Keene et al., 2007	Examined the physical and chemical properties of newly created aerosols from bubble bursts at a modeled air-sea interface	Surface/Laboratory
Keene et al., 2014	Investigated long-term trends in aerosol and precipitation composition across the WNAO at Bermuda	Surface
Kim et al., 1990	Investigated volume/surface area size distribution of marine aerosols during the GCE/CASE/WATOX.	Aircraft
Lin et al., 2012	Sulphur isotope data was used to evaluate the importance of sulfate sources in the Atlantic Ocean	Ship
Mackey et al., 2012	Studied how atmospheric metal deposition affects open-ocean and coastal phytoplankton ecosystems	Surface
Merrill et al., 1996	Meteorological analysis of tropospheric ozone profiles in Bermuda	Ozonesondes
Miller & Harris, 1985	Examined Bermuda's flow climatology and its impact on long-range transport	Aircraft
Milne et al., 2000	Export of ozone precursors to Bermuda	Aircraft
Moody et al., 1995	Assessed Bermuda transport climatology of tropospheric ozone	Surface
Moody & Galloway, 1988	Examined the impact of various atmospheric flow patterns on precipitation composition in Bermuda	Surface
Moody et al., 1996	Investigated the large-scale view of O_3 transport over WNAO	Surface/Ozonesonde
Moody et al., 2014	Studied flow climatology for physicochemical parameters of nominal super- and sub-μm aerosol in Bermuda	Surface
Muhs et al., 2012	Studied the importance of African dust transport and deposition in Bermuda	Surface
Oltmans & Levy, 1992	Examined the seasonal cycle of surface ozone over the western North Atlantic	Surface
Oltmans et al., 2006	Investigated the long-term changes in tropospheric ozone	Surface
Oltmans et al., 2013	Investigated the recent tropospheric ozone changes	Satellite/Surface
Parrish et al., 2016	Examined seasonal cycles of O_3 in the marine boundary layer	Surface/Modeling
PéTron et al., 2002	Modeled carbon monoxide surface emissions inversely using Climate Monitoring and Diagnostics Laboratory network observation	Modeling
Piotrowicz et al., 1990	Observation of ozone and carbon monoxide over the north Atlantic during a boreal summer	Ship
Prados et al., 1999	Investigated ozone and pollutant transport from North America to the North Atlantic Ocean during the 1996 AEROCE experiment	Aircraft

Ray et al., 1990	Investigated marine troposphere H_2O_2 and O_3 over the western Atlantic Ocean	Aircraft
Savoie et al., 2002	Examined marine biogenic and anthropogenic contribution to non-sea-salt sulfate in the North Atlantic Ocean's MBL	Surface
Saikawa et al., 2014	Examined the global and regional emissions estimates for N_2O	Surface/Modeling
Sholkovitz & Sedwick, 2006	Analyzed aerosol samples for iron and soluble sodium (as a proxy for sea salt)	Bouy/Surface
Sholkovitz et al., 2009	Created an empirical approach to estimate aerosol iron solubility using bulk Fe, V, and Al concentrations	Ship
Sievering et al., 1990	Examined size distributions and statistical analysis of nitrate, excess sulfate, and chloride deficiency in the marine boundary layer during GCE/CASE/WATOX	Ship
Sievering et al., 1991	Investigated the role of aerosol water content and size distribution on heterogeneous sulfur conversion in sea-salt aerosol particles	Ship
Smirnov et al., 1998	Examined optical properties of Saharan dust during ACE 2	Surface
Smirnov et al., 2000	Measured and analyzed atmospheric optical parameters on U.S. Atlantic coast sites, ships, and Bermuda during TARFOX	Surface
Smirnov et al., 2003	Developed a model for maritime aerosols using AERONET	Surface/Modeling
Tanré et al., 1999	Retrieved aerosol optical thickness and size distribution over ocean using MODIS airborne simulator during TARFOX	Satellite
Thompson et al., 2014	Time-dependent Bayesian inversion technique was used to estimate N_2O surface fluxes from 1999 to 2009	Surface/ship
Todd et al., 2003	Examined the impact of wet deposition on atmospheric optical properties in Bermuda and Barbados	Surface
Tomza et al., 2001	Examined color-related differences in the chemical composition	Surface
Turekian et al., 2001	Used stable sulfur isotopes to differentiate sources of size-resolved sulfate in polluted marine air in Bermuda during spring	Surface
Turekian et al., 2003	Analyzed Bermuda's size-segregated aerosols for organic carbon, oxalate, and stable isotope compositions to understand chemical processes and their sources	Surface
Volpe & Spivack, 1994	Researched the stable chlorine isotopic composition of western Atlantic Ocean marine aerosol particles	Surface
Witek et al., 2013	Conducted measurements of ocean aerosol optical depths using MISR retrievals and collocated MAN and AERONET measurements	Surface
Wolff et al., 1986	Investigated SO_x, NO_x, and aerosol species measurement in Bermuda	Surface

Table S2. Summary of abbreviations and acronyms used in the paper.

ACE-ENA	Aerosol and Cloud Experiment in the Eastern North Atlantic
ACTIVATE	Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment
AMS	Aerosol Mass Spectrometer
APT	Accumulated Precipitation along Trajectories
AVAPS	Airborne Vertical Atmospheric Profiling System
CALIOP	Cloud-Aerosol Lidar with Orthogonal Polarization
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations
CCN	Cloud Condensation Nuclei
DLH	Diode Laser Hygrometer
DMA	Differential Mobility Analyzer
DMS	Dimethylsulfide
FCDP	Fast Cloud Droplet Probe
f(RH)	Hygroscopicity Parameter
FT	Free Troposphere
GDAS	Global Data Assimilation System
HSRL-2	High Spectral Resolution Lidar – Generation 2
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory model
LaRC	NASA Langley Research Center
LAS	Laser Aerosol Spectrometer
LWC	Liquid Water Content
MBLH	Marine Boundary Layer Height
MF_{org}	Mass Fraction of Organics From AMS
MLH	Mixed Layer Height
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NOAA	National Oceanic and Atmospheric Administration
NSS	Non-Sea salt
PILS	Particle into Liquid Sampler
RF	Research Flight
RH	Relative Humidity
SMPS	Scanning Mobility Particle Sizer
UTC	Coordinated Universal Time
VOC	Volatile Organic Compounds

Table S3. Summary of median aerosol volume concentration statistics for different diameter size ranges for the three major air mass types.

	V ($\mu m^3 cm^{-3}$)			
<1 km	<100 nm	0.1 – 1 µm	1 – 5 µm	>3 µm
North America	0.08	1.36	3.33	5.17
Ocean	0.02	1.20	0.71	4.91
Caribbean/North Africa	0.02	1.41	6.15	5.42
1 – 3.5 km				
North America	0.05	0.42	0.00	0.59
Ocean	0.02	0.60	0.00	1.60
Caribbean/North Africa	0.03	0.68	0.54	2.99

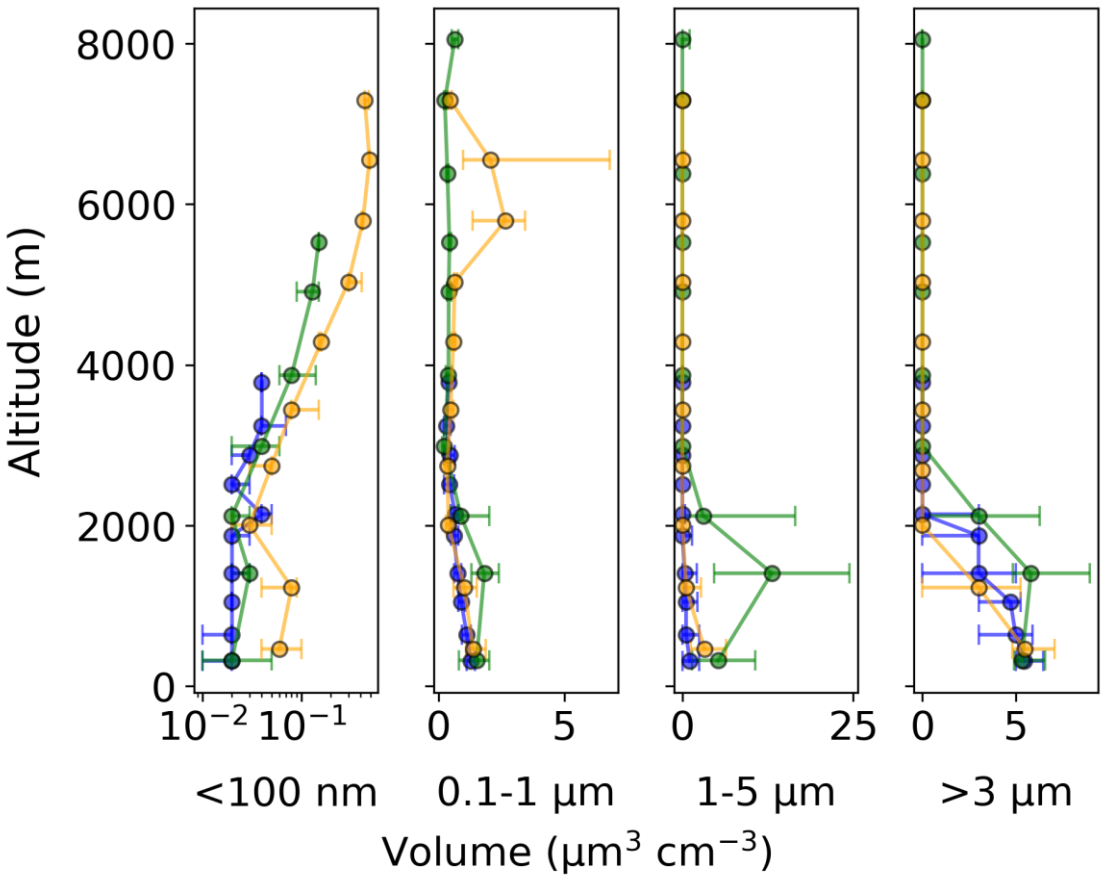

Figure S1: Vertical distribution of aerosol volume concentration for flight data grouped into similar air mass source categories (Orange = North America; blue = Ocean; green = Caribbean/North Africa). Markers are median values and whiskers are 25th/75th percentiles.

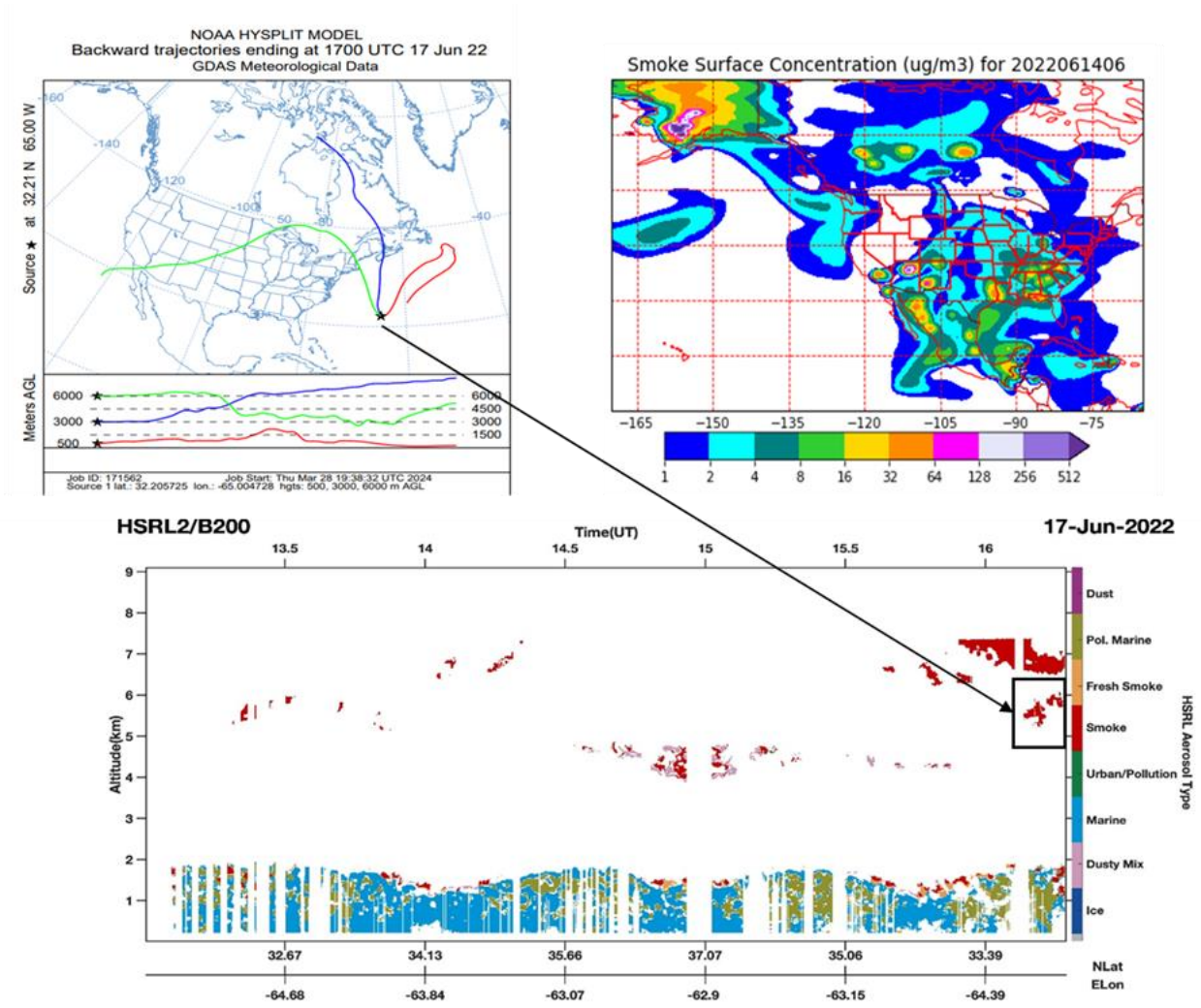

Figure S2: A visual summary of (a) 5-day HYSPLIT back-trajectory results ending at the point of the Falcon spiral on 17 June 2022 (RF 178), (b) a spatial distribution of smoke surface concentrations across North America from NAAPS (13 June chosen for display to account for transport time to Bermuda on 17 June), and (c) HSRL-2 aerosol type vertical distribution pertaining to 17 June 2022 where elevated aerosol extinction was observed around 5-6 km altitude (Fig. 6a). Most relevant in panel (a) is the green trajectory ending at 6 km over Bermuda where data suggest there was a smoke layer.

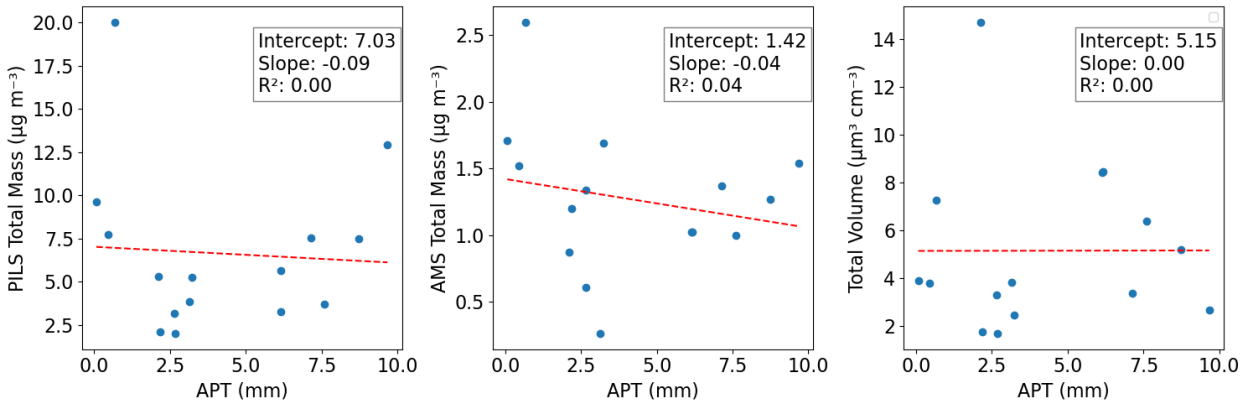

Figure S3: Scatterplots of PILS total mass, AMS total mass, and total volume concentration (D_p: 0.01-5 µm) versus APT for all spirals, with each marker corresponding to the mean value for a given spiral. The linear best-fit line is in red with relevant fit metrics in respective legends.

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