A unique vertical characterization of aerosol over a remote marine region: A case study over Bermuda

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Objectives

- Improved vertically resolved characterization of marine atmospheric variables is crucial for understanding atmospheric chemistry and aerosol and cloud processes.
- Difficult to obtain vertically resolved aerosol properties and trace gas data over remote marine areas, making model validation and reanalysis product improvement challenging.

Science question

- How do aerosol variables and trace gases vary temporally, vertically, and with air mass source changes at a remote marine site (Bermuda)?

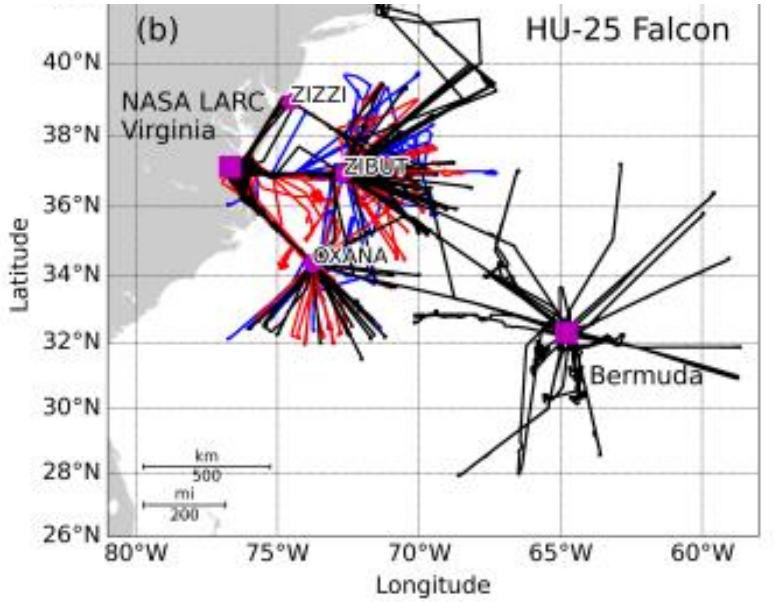
Filling a void

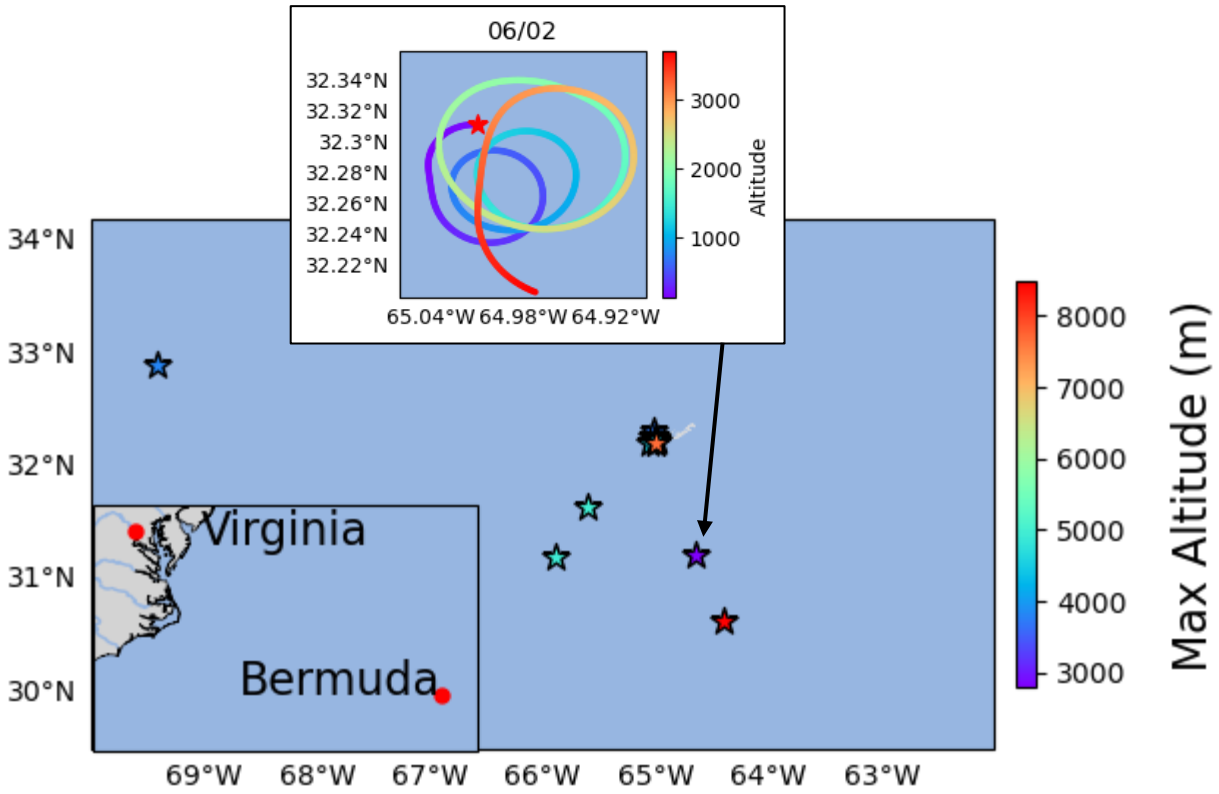
Large volume of Bermuda studies focused on surface measurements; thin inventory of reports dealing with gas/aerosol aloft

	Key Points
Horvath et al. (1990)	higher number/volume concentrations (and associated mean diameters of number/volume) at lower altitudes
Kim et al. (1990)	
Aryal et al. (2014)	surface optical data weakly related to column optical properties over Bermuda

ACTIVATE: aircraft campaign over Northwest Atlantic

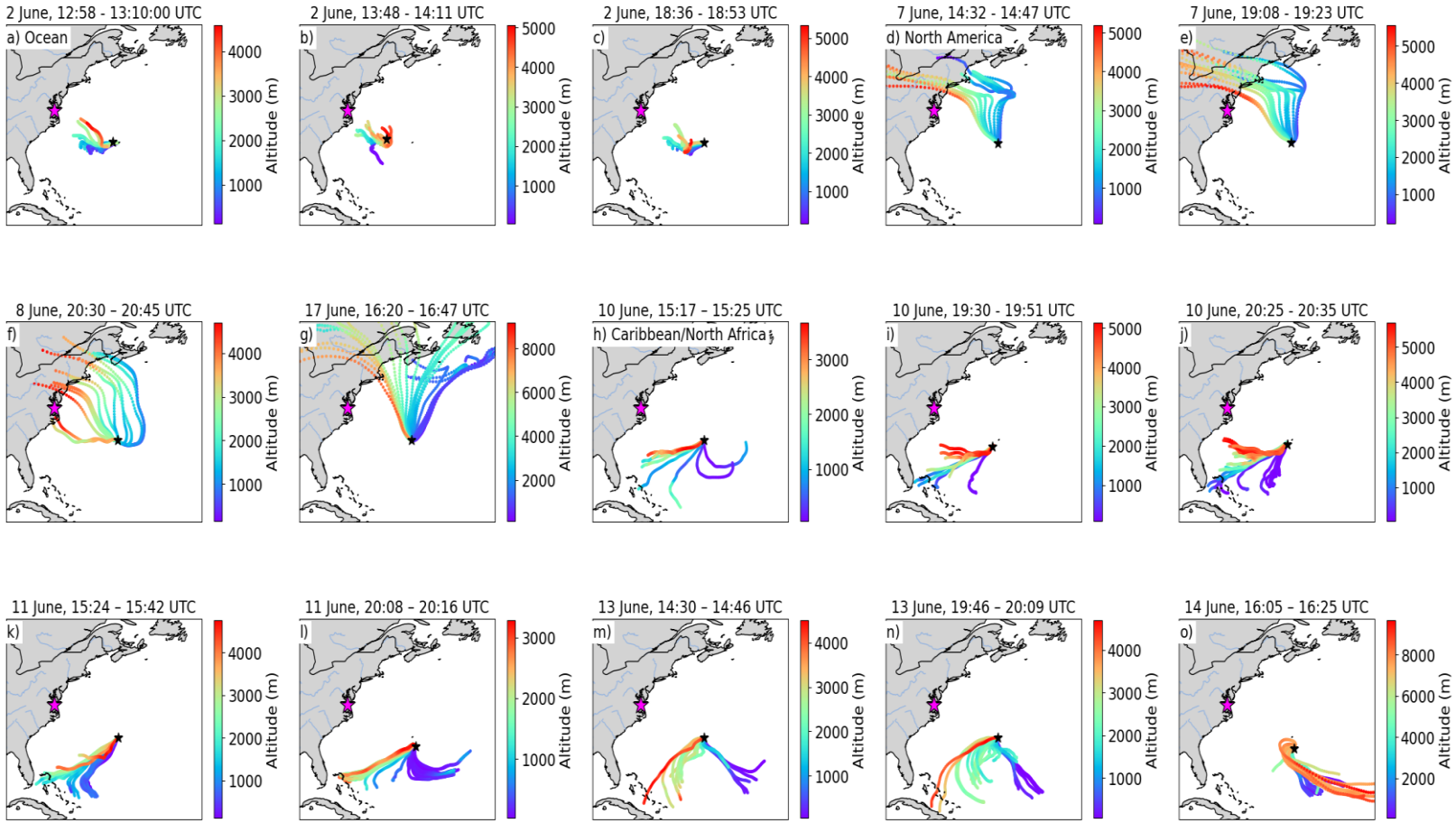
- 179 total flights, including 162 joint flights between Feb 2020 – June 2022
- 15 vertical spiral soundings conducted by Falcon between 2-17 June 2022 during Bermuda intensive
  - Coordinated in part with Tudor Hill Research Tower facility





A Few Details on Methods

- NOAA Hysplit back trajectory analysis (72 hr) conducted for each minute of spiral soundings
  - 3 predominant sources: Caribbean/North Africa, North America, Ocean
- Trace gas, aerosol, and met/nav descriptive statistics by air mass source and altitude range:
  - <1 km (MBL)
  - 1-3.5 km (free troposphere, FT)
- Applied sensitivity test to AMS total mass and excluded masses < 0.4 μg m^{-3}

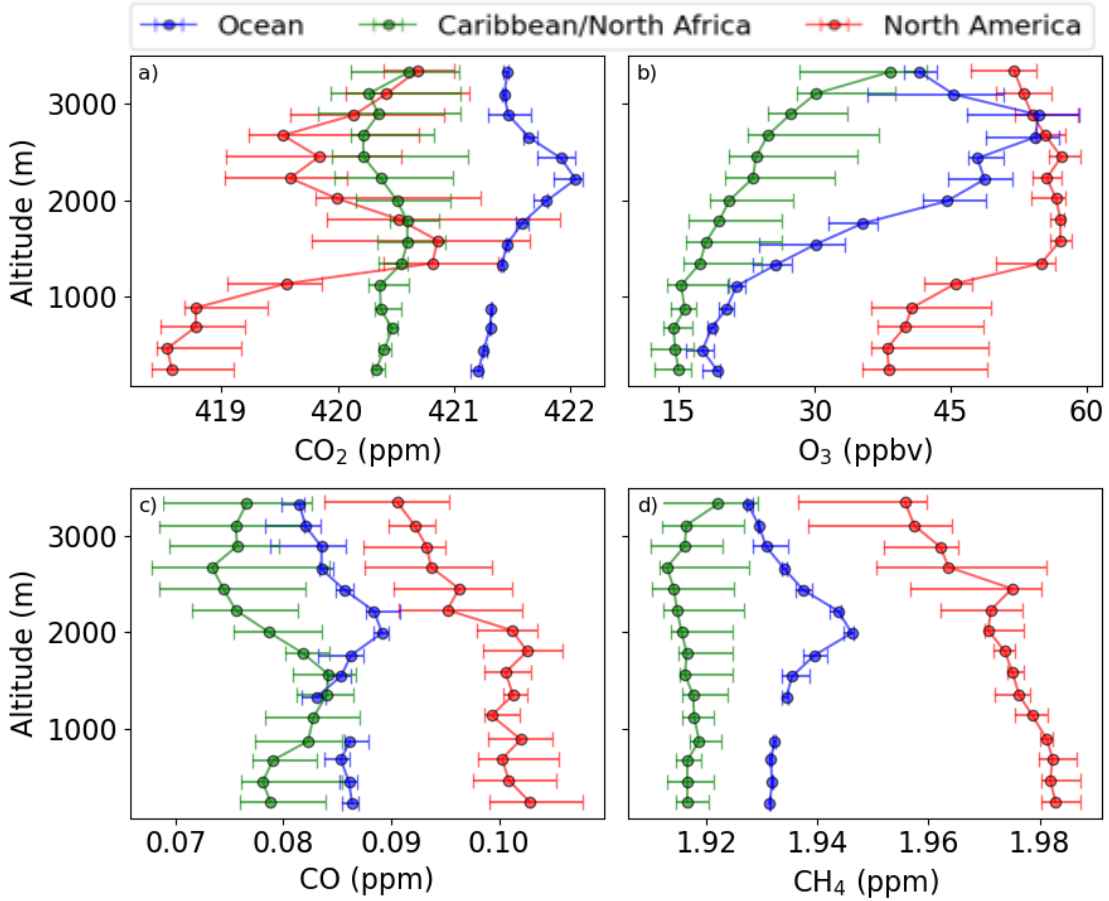


Spiral Details

- RF168 (06/08/2022): only spiral w/ cloud penetration (FCDP LWC > 0.02 g m^{-3} and N_d > 10 cm^{-3} for ≥ 2 consecutive seconds)

Research Flight & Date	Spiral Time (UTC)	Altitude Range (m)	Air Mass Source
162 06/02/2022	12:58 – 13:10	100 - 4000	Ocean
162 06/02/2022	13:48 – 14:10	100 - 3200	Ocean
163 06/02/2022	18:36 – 18:53	100 - 4000	Ocean
166 06/07/2022	14:32 -14:47	100 - 3200	North America
167 06/07/2022	19:08 – 19:23	100 - 3500	North America
168 06/08/2022	20:30 – 20:45	100 - 4000	North America
169 06/10/2022	15:17 – 15:25	100 - 3500	Caribbean/North Africa
169 06/10/2022	19:30 – 19:51	100 - 5000	Caribbean/North Africa
170 06/10/2022	20:25 -20:35	100 - 5000	Caribbean/North Africa
171 06/11/2022	15:24 – 15:42	100 - 5000	Caribbean/North Africa
172 06/11/2022	20:08 – 20:30	100 - 3000	Caribbean/North Africa
173 06/13/2022	14:30 – 14:46	100 - 4500	Caribbean/North Africa
174 06/13/2022	19:46 – 20:09	100 - 4500	Caribbean/North Africa
176 06/14/2022	16:05 – 16:25	100 - 8500	Caribbean/North Africa
178 06/17/2022	16:20 – 16:47	100 - 7600	North America

Trace gas composition categorized by air mass source



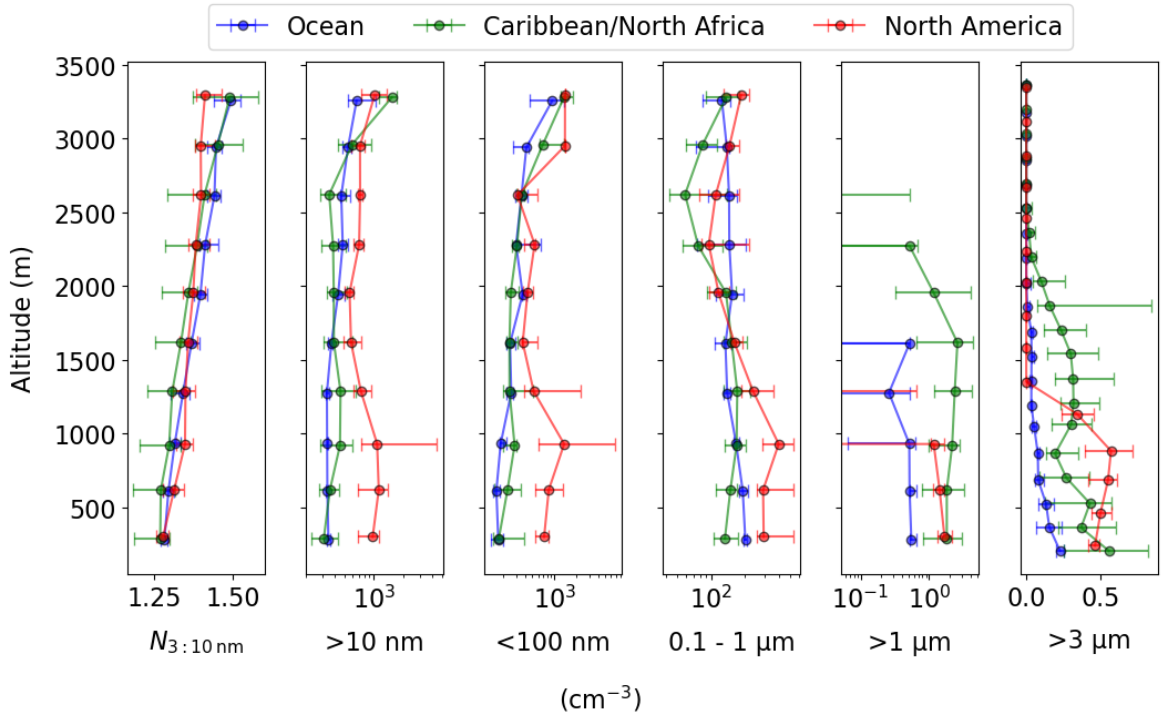
Markers are median, and whiskers are 25th/75th percentiles

Median values

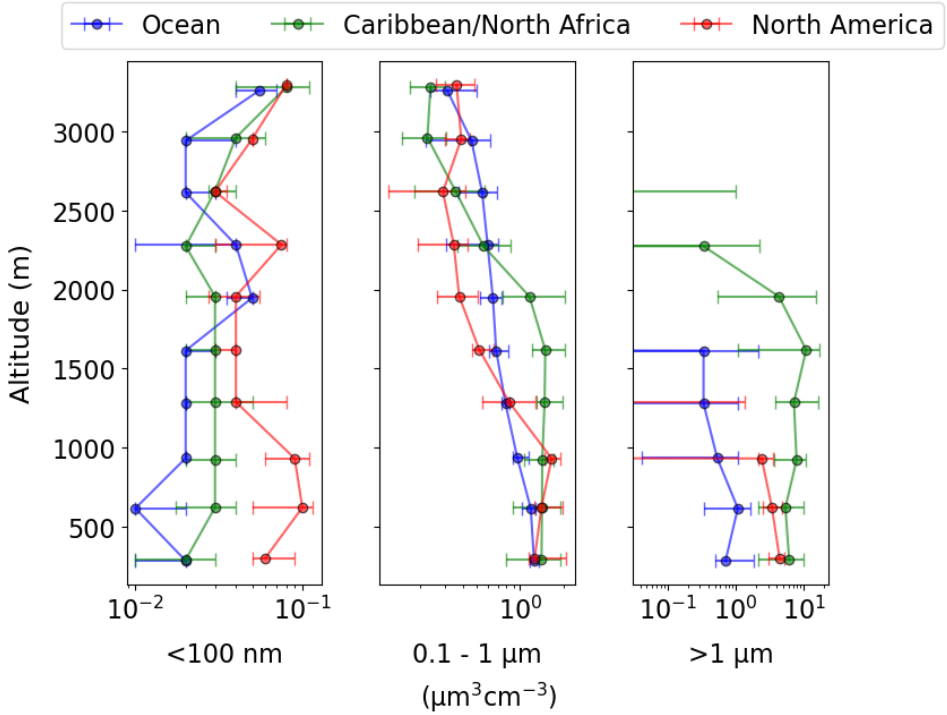
< 1 km	T (°C)	RH (%)	CH_4 (ppm)	CO_2 (ppm)	CO (ppm)	O_3 (ppbv)
North America	17.82	72.97	1.98	419.20	0.10	49.40
Ocean	20.86	82.13	1.93	421.29	0.09	18.93
Caribbean/North Africa	22.00	77.87	1.92	420.32	0.08	14.95
1 – 3.5 km						
North America	12.36	22.28	1.97	420.03	0.10	55.65
Ocean	12.14	50.61	1.93	421.56	0.08	43.50
Caribbean/North Africa	13.14	69.10	1.92	420.49	0.08	25.35

- North America CO/CH_4/O_3 systematically higher
- O_3 enhanced in FT
- Much drier in FT of North America air masses

Aerosol number (N) profiles



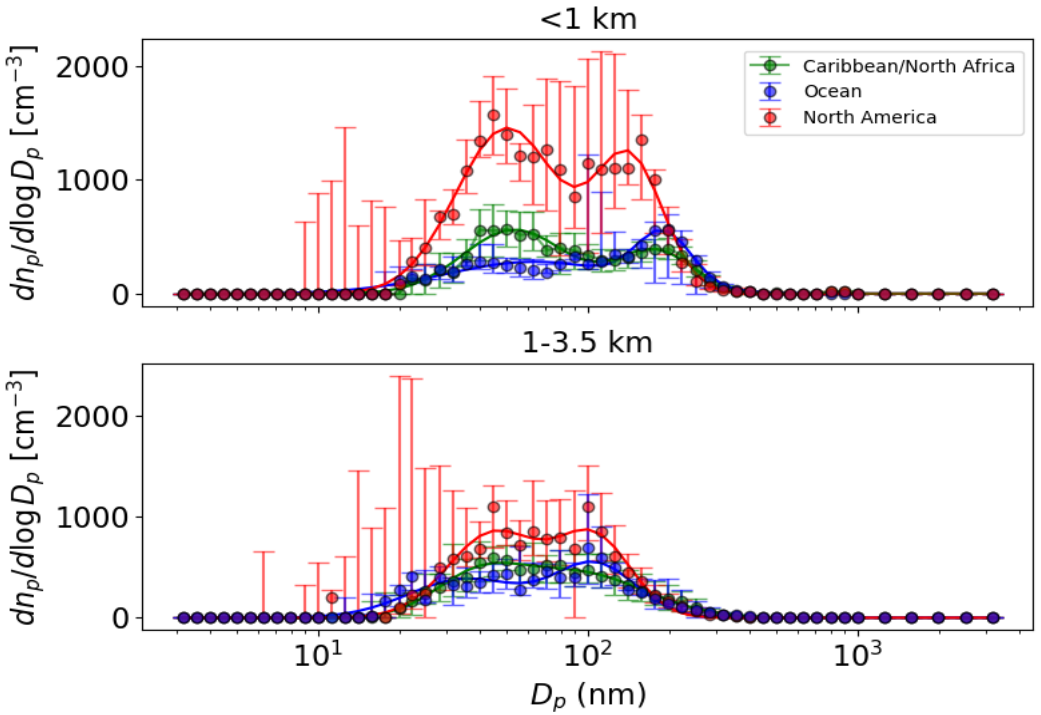
Aerosol volume (V) profiles



- Caribbean/North Africa: highest N/V concentrations of particles >1 µm
- North America: highest N for submicron range and >3 µm in MBL
- Ocean: generally lowest N/V for all size categories
- $N_{3:10\,nm}$ increases with altitude for all air source regions

Aerosol Size Distributions (SMPS + LAS)

Fitted two lognormal model curve categorized by air mass source

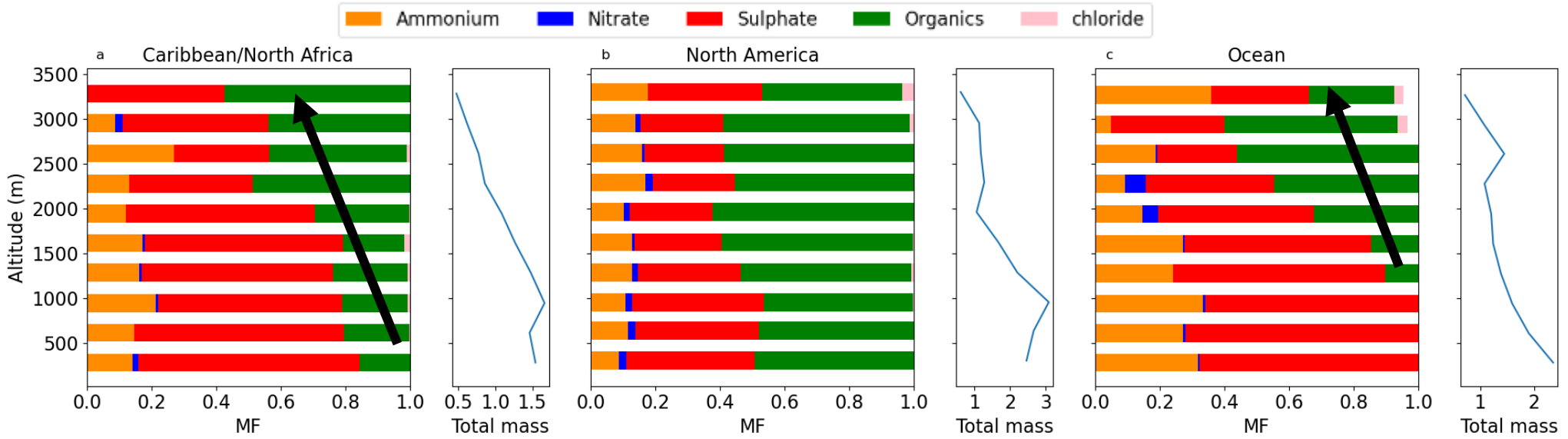


Two mode lognormal model parameter categorized by source region

< 1 km	$N_1(cm^{-3})$	$D_{g,1}(nm)$	$\sigma_{g,1}(nm)$	$N_2(cm^{-3})$	$D_{g,2}(nm)$	$\sigma_{g,2}\ (nm)$	$V(um^3cm^{-3})$
North America	1594	50.32	1.54	855.12	142.68	1.34	3.99
Ocean	537.80	63.35	2.15	273.82	191.93	1.27	1.72
Caribbean/ North Africa	619.64	53.47	1.55	311.92	176.83	1.39	3.34
1 – 3.5 km							
North America	828.97	45.03	1.48	612.68	105.97	1.37	0.94
Ocean	472.57	35.45	1.63	454.78	106.82	1.41	0.90
Caribbean/ North Africa	344.83	38.97	1.45	627.41	92.13	1.76	0.81

- Significant variability in North America air mass concentrations
- Aitken mode more pronounced in MBL except for Ocean
  - Ocean $\sigma_{g,1}$ highest and $\sigma_{g,2}$ lowest among all source regions in MBL
- Mode heights more similar for Aitken/Accumulation modes in FT versus MBL
- N generally reduced in FT for Aitken mode but enhanced for accumulation mode for Ocean and Caribbean/North Africa

Submicrometer aerosol (AMS) composition



Mass Fractions (MF)

<1 km	Organics	SO_4	NO_3	NH_4	Cl	f_{44}	*AMS total mass* (μg m^{-3})
North America	0.49	0.39	0.023	0.10	0.00	0.12	2.66
Ocean	0.19	0.95	0.002	0.31	0.01	-0.39	2.03
Caribbean/ North Africa	0.19	0.65	0.01	0.16	0.00	-0.40	1.50
1 – 3.5 km							
North America	0.56	0.28	0.01	0.15	0.01	0.12	1.41
Ocean	0.46	0.48	0.01	0.19	0.01	0.03	1.20
Caribbean/ North Africa	0.37	0.49	-0.01	0.15	<0	0.05	0.99

- North America: highest total mass concentration, organic MF, and f_{44} with altitude
- Ocean and Caribbean/North Africa: Highest sulfate MF and increasing organic MF in FT
- Ocean: Highest ammonium MF

PILS water-soluble composition (< 5 µm)

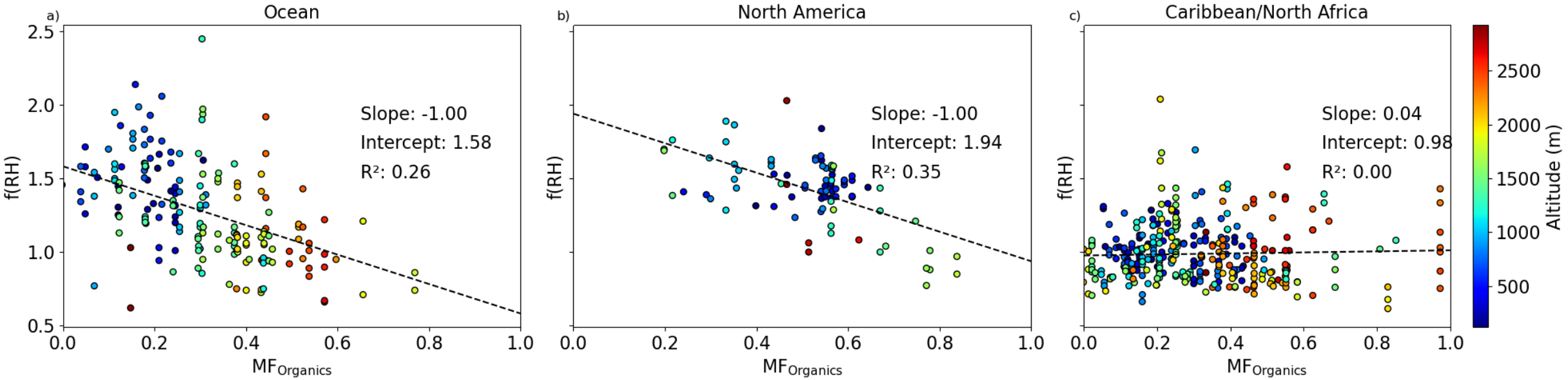
Mean statistics for the fifteen spirals categorized based on the source region

Mass Fractions (MF)

Research Flight & Date	Total Mass (µg m^{-3})	Na	K	Mg	Ca	Cl	NO_3	SO_4	NSS SO_4	Oxalate	Cl:Na	Air mass category
162 06/02/2022	2.00	0.17	0.01	0.03	0.04	0.15	0.26	0.35	0.31	0.00	0.89	Ocean
162 06/02/2022	5.24	0.21	0.01	0.02	0.02	0.19	0.22	0.32	0.27	0.00	0.93	Ocean
163 06/02/2022	2.09	0.40	0.02	0.04	0.03	0.06	0.16	0.28	0.18	0.01	0.27	Ocean
166 06/07/2022	20.03	0.29	0.01	0.02	0.01	0.45	0.10	0.11	0.04	0.00	1.55	North America
167 06/07/2022	7.72	0.55	0.01	0.04	0.00	0.32	0.03	0.05	0	0.00	0.59	North America
168 06/08/2022	9.63	0.32	0.01	0.02	0.02	0.47	0.04	0.12	0.04	0.00	1.47	North America
169 06/10/2022	7.53	0.26	0.00	0.01	0.01	0.42	0.09	0.21	0.14	0.00	1.63	Caribbean/North Africa
169 06/10/2022	3.70	0.36	0.01	0.03	0.03	0.15	0.17	0.23	0.14	0.00	0.61	Caribbean/North Africa
170 06/10/2022	7.47	0.20	0.02	0.03	0.01	0.30	0.18	0.27	0.22	0.01	1.52	Caribbean/North Africa
171 06/11/2022	3.26	0.22	0.02	0.03	0.07	0.19	0.23	0.24	0.19	0.00	0.85	Caribbean/North Africa
172 06/11/2022	5.64	0.10	0.02	0.06	0.07	0.13	0.29	0.28	0.25	0.05	2.18	Caribbean/North Africa
173 06/13/2022	5.31	0.20	0.01	0.03	0.02	0.29	0.18	0.27	0.22	0.01	1.45	Caribbean/North Africa
174 06/13/2022	3.15	0.08	0.05	0.04	0.31	0.13	0.16	0.23	0.21	0.01	1.50	Caribbean/North Africa
176 06/14/2022	3.86	0.15	0.01	0.02	0.03	0.20	0.27	0.31	0.27	0.00	1.29	Caribbean/North Africa
178 06/17/2022	12.91	0.28	0.01	0.03	0.00	0.43	0.12	0.13	0.06	0.00	1.54	North America
Air mass type												
North America	6.33	0.33	0.01	0.03	0.01	0.43	0.08	0.11	0.05	0.00	1.39	
Ocean	3.65	0.27	0.02	0.03	0.03	0.14	0.20	0.32	0.25	0.00	0.70	
Caribbean/North Africa	5.18	0.20	0.02	0.03	0.06	0.22	0.20	0.26	0.21	0.01	1.40	

- Similar to AMS, total mass highest for North America but due to high sea salt levels in some flights (e.g., RF 166-168, 178)
- Low Cl:Na ratio for Ocean source; consistent with Mosello et al. (2005) and Anne et al. (2008)
  - Chloride depletion yields excess chloride via HCl (Edwards et al., in review)
- Low NSS SO_4 mass fraction for North America similar to AMS
- Enhanced Ca for some Caribbean/North Africa flights presumed to be due to dust (e.g., RF 173)

Submicrometer f(RH)



- Enhanced organic mass fraction usually coincides with reduced aerosol hygroscopicity
  - Anticipated anticorrelation observed only for Ocean and North America
  - Possible influence of refractory species in yielding the lack of anticorrelation for Caribbean/North Africa

Conclusions and Future Work

- Differences observed in trace gases, aerosol properties (size distribution, composition/hygroscopity), and meteorology due to varying air mass source regions and altitude range
- North American air masses in particular yield enhanced aerosol number/mass concentrations, particulate organics, and trace gas levels (e.g., CO, CH_4)
- Caribbean/North Africa shows signatures of some dust influence, consistent with other reports of dust during the Bermuda intensive

- Any feedback appreciated…