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We express our gratitude to the reviewers for valuable feedback on the draft. We have addressed the remarks below by providing our responses in blue.

Reviewer 1

Ajayi et al report observations of trace gases, aerosol properties, and meteorological variables during 15 aircraft spirals near Bermuda in June of 2022. This study provides information on the vertical distribution of these observations at a location with a history of surface-based observations. This vertical information is unique for this area and will aid interpretation of surface and satellite-based observations. The paper mainly reports the observations, describing what they observed, without much detailed analysis or interpretation. However, this is a unique and relatively large data set that may be valuable for interpretation or reinterpretation of past and future studies in the Bermuda area, as well as for understanding satellite observations of trace gases and aerosol over the oceans. The vertical information was provided by 2 separate aircraft, one flying at higher altitudes and the other flying in spirals between 0.15 to 8 km altitude. The aircraft had multiple instruments to measure meteorological parameters, aerosol properties, and trace gas concentrations. The higher-flying aircraft deployed dropsondes to measure vertical profiles of temperature, relative humidity, and wind speed.

They categorize the different vertical soundings into three source regions using the HYSPLIT model. The three regions, North America, Ocean, and North Africa/Caribbean, identified by HYSLPIT are confirmed with the observed trace gases and aerosol chemical composition. They found considerable vertical variability in all three categories, with generally higher trace gas concentrations with increasing altitude (especially for ozone). Sub-micron particle concentrations also increased with altitude suggesting new particle formation in the free troposphere. Super-micron concentrations were highest near the surface and negligible above the boundary layer. Organics tended to dominate aerosol mass in the FT while sulfate and chloride was more important closer to the surface.

This paper is suitable for publication in ACP. This data could have comprised several papers that include a more detailed analysis of the observations. The data is high quality and will be useful for such future studies and thus warrants publication.

A few minor comments to improve clarity are below:

1. Line 224: Is it 2-3 days or 2-3 hours? I think days but I'm not sure why "hours" is there.

Response: We appreciate your observation. The appropriate time frame is "2-3 days." The necessary revisions have been made to the document:

"The range of transport times from the coastline of North America to Bermuda for this category was ~2-3 days for the MBL and ~2 days for the FT."

2. The short paragraph beginning on line 272 states differences in CO_2 concentrations between the MBL and FT but zero analysis is given. Are these differences significant? Are they expected? Why are they different?

Response: We revised the CO_2 text based on this comment as we agree maybe it is misleading to get into details of variability and differences between the MBL and FT:

"Carbon dioxide (CO_2) levels were fairly similar at the altitudes examined (Fig. 4) with median values for the altitude bins ranging from ~418 to ~422 ppm. The most variability at fixed altitude bins, as represented by whiskers in Fig. 4, was for North American air masses."

3. Section 3.6: Remind the reader the aerosol size distribution that the AMS samples at the beginning of this section. Mean numbers are given in this section. It would be good to also state the variability in some way, such as the standard deviation of the mean.

Response: We edited Section 3.6 of the manuscript to include the size range of the AMS measurements and the standard deviation associated with mean values relevant to AMS/PILS data:

"Discussion next explores AMS submicron composition (Fig. 8 and Table 6), which is relevant to the diameter range between 0.06 and 0.6 µm (Table 2). The total AMS mass concentration was highest for North America across the range of altitudes sampled, with mean values (µg m^{-3}) below 1 km being 2.66 ± 1.00 (North America), 2.03 ± 0.38 (Ocean), and 1.53 ± 0.52 (Caribbean/North Africa). Values tended to decrease with altitude with the exception of a notable enhancement between 5 – 7 km (peaking at 2.78 µg m^{-3} observed at ~ 6500 m) for North American air in line with the enhancement in aerosol volume concentration already shown (Fig. S1) that was attributed to smoke (Fig. S2).

Speciated mass fractions for North American air were distinctly different from the other two categories owing to the dominance of organics, reaching values of 0.49 ± 0.10 and 0.56 ± 0.16 for < 1 km and 1-3.5 km, respectively. Sulfate was the next most abundant constituent, followed by ammonium and with much lower mass fractions for nitrate and chloride (typically <0.03). The degree of oxygenation of the organic constituents (f_{44}) was the highest for North American air with mean values of 0.12 ± 0.06 for both altitude ranges in Table 6. These general results for North American air are consistent with past work examining the first two years of ACTIVATE flights closer to the U.S. East Coast (Dadashazar et al., 2022), where sulfate and organics dominated AMS

mass concentration but f_{44} values are lower in these Bermuda profiles than the latter study's sample set that showed values typically between 0.1-0.3.

For the Ocean category, sulfate was the most abundant constituent in the lowest 1 km with a mean mass fraction of 0.64 ± 0.07, followed by ammonium (0.24 ± 0.07), and organics (0.11 ± 0.09). The composition of this category shifted between 1-3.5 km to have comparable levels of sulfate and organics with their mean mass fractions being 0.38 ± 0.16 and 0.40 ± 0.19, respectively, while ammonium was 0.20 ± 0.16.

Lastly for the Caribbean/North Africa category, the mean mass fractions were similar to the Ocean category with a sulfate-rich profile in the lowest km (mass fraction of 0.65 ± 0.12), that became somewhat more comparable between sulfate and organics between 1-3.5 km with mean mass fractions of 0.49 ± 0.54 and 0.37 ± 0.41, respectively. Extending up higher to 8 km, the organic mass fraction for this category kept increasing with values between 0.5-1.0 for various altitude bins between 2 and 6 km.

Table 7 summarizes the water-soluble composition results from the PILS, which offers insight into species not measured by the AMS and also a look at a broader size range up to 5 µm. PILS data were combined for entire spiral soundings without separation into altitude layers owing to the lengthier time resolution to obtain samples as compared to the other instrument datasets. When viewing total mass as the sum of concentrations (µg m^{-3}) of the species shown (using total sulfate rather than nss sulfate for calculation of total mass), North America exhibited the highest mean value (13.26 ± 6.47), followed by Caribbean/North Africa (5.18 ± 3.26) and Ocean (3.65 ± 2.27). Noteworthy was the stretch of three flights between 7-8 June for North American air where the total masses were significantly higher than other flights (up to 20.03 µg m^{-3} for RF166), which we attribute to the weather conditions associated with the passage of tropical storm Alex where high winds could have helped promote more sea salt emissions. This is confirmed by the high Na^+ and Cl^- mass fractions for those particular flights (0.29-0.55 for Na^+; 0.32-0.47 for Cl^-). The high APT values associated with RF178 do not translate to significantly lower PILS mass concentrations as compared to other flights; RF178 had the second highest total PILS mass (12.91 µg m^{-3}) due to high Na^+ and Cl^- contributions from sea salt. As it relates to APT, the mean value for each spiral showed no significant relationship with either AMS/PILS total mass or the total volume concentration between 0.01-5 µm from the LAS (Fig. S3). Past work for surface aerosol over Bermuda showed that the winter season is characterized by most wet scavenging as compared to

other seasons in terms of air masses arriving at Bermuda from distant continents (Dadashazar et al., 2021). Aside from limitations of relying on APT as a metric for wet scavenging of aerosol, a lack of any relationship with APT (Fig. S3) also builds on findings that precipitation may actually be a source of particles rather than just a sink (e.g., Khadir et al., 2023).

The unique feature from the Caribbean/North Africa spirals was the higher mass fraction for Ca^{2+}, especially on RF 174 (0.31), which is supportive of the presence of dust and consistent with results shown already for how supermicron particles were enhanced for this air mass type (e.g., Figs. 5-6, and S1) for particles larger than 1 μm.

We assessed the potential for sea salt reactivity in the form of chloride depletion by comparing the mass ratio of Cl^-:Na^+ between different spiral soundings, with a value of 1.8 being attributed to pure sea salt. The mean Cl^-:Na^+ ratio for the three air mass types was 0.70 ± 0.40 (Ocean), 1.39 ± 0.37 (North America), and 1.40 ± 0.69 (Caribbean/North Africa). While the Ocean spirals exhibited the lowest overall mass concentrations, they did have the highest relative combined amount of nitrate, nss sulfate, and oxalate, which are key acidic species participating in chloride depletion. The results suggest that the sea salt sampled, regardless of air mass origin, had potentially experienced a loss of chloride due to reactions with acidic species. We caution though that the use of the Cl^-:Na^+ ratio to assess chloride depletion has limitations owing to the assumption that sea salt is the only source of the two species used in the ratio, which at least becomes a challenge for the Caribbean/North Africa category where there is evidence supporting the presence of dust. The reader is referred elsewhere for a more detailed discussion of sea salt reactivity during ACTIVATE and over Bermuda (Edwards et al., 2023)."

4. Table 6 caption: "total mass threshold > 0.4…" Threshold for what? Is this the detection limit?

Response: We updated and fixed the text as such in Section 2.2 and do not make mention of this mass threshold later in the paper as it is unnecessary after the first mention in Section 2.2:

"To allow for better data quality, m/z 44 data are used when organic mass concentration exceeded 0.4 μg m^{-3}."

5. Paragraph beginning on line 509: Please provide information on the variability about the mean for the Cl/Na ratio.

Response: We edited the text to include information about variability

"We assessed the potential for sea salt reactivity in the form of chloride depletion by comparing the mass ratio of Cl^-:Na^+ between different spiral soundings, with a value of 1.8 being attributed to pure sea salt. The mean Cl^-:Na^+ ratio for the three air mass types was 0.70 ± 0.40 (Ocean), 1.39 ± 0.37 (North America), and 1.40 ± 0.69 (Caribbean/North Africa)."

Reviewer 2

The manuscript "Vertical variability of aerosol properties and trace gases over a remote marine region: A case study over Bermuda" presents an analysis of airborne data collected during 15 vertical spiral soundings over Bermuda as part of the NASA ACTIVATE field campaign. The study focuses on understanding the vertical distribution of trace gases and aerosol properties from different air mass source regions (North America, Ocean, Caribbean/North Africa). The data from this paper is valuable to the community, as such vertical measurements of the marine atmosphere are rare.

I do not have any major concerns about this paper. There are some minor suggestions and comments:

1. Abstract: When describing the major findings, the structure of the five bullet points is inconsistent. Some are complete sentences, while others are not. I suggest using the same sentence structure throughout.

Response: We edited the structure of the major findings in the abstract of the manuscript to ensure completeness and consistency:

"(i) the strongest pollution signature is from North American air masses, while the weakest is from the Ocean category; (ii) North American air has the highest levels of CO, CH_4, submicron particle number concentration, AMS mass, and highest organic mass fraction along with smoke layers in free troposphere (FT); (iii) Ocean air has the highest relative amount of nitrate, non-sea-salt sulfate, and oxalate, which are key acidic species participating in chloride depletion; (iv) air masses from Caribbean/North Africa showed a pronounced coarse aerosol signature in the FT and reduced aerosol hygroscopicity, which is associated with dust transport; and (v) there is considerable vertical heterogeneity for almost all variables examined, including higher O_3 and submicron particle concentrations with altitude, suggestive that the FT is a potential contributor of both constituents in the marine boundary layer".

2. There are many abbreviations in this paper. I suggest summarizing them in a table for clarity.

Response: We summarized the abbreviations in the supplemental file in the form of the following table below and we introduced the table at end of introduction:

"To aid with the paper, Table S2 provides definitions of acronyms and abbreviations used subsequently."

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

Based on Figure 3, the wind speed at ~8000 m is only 5-10 m/s. Why is the wind speed so low? Additionally, I recommend not using red and green to distinguish different markers as they can be difficult to differentiate for colorblind readers.

Response: The low wind speeds of 5-10 m/s at approximately 8000 meters were checked and confirmed to be correct. They can be attributed to a host of conditions that are outside the scope of this work as we aren't focused on wind speed variations with altitude.

Also, the colors have been adjusted for colorblind accessibility (orange, blue, and green). An example is shown here for Figure 3:

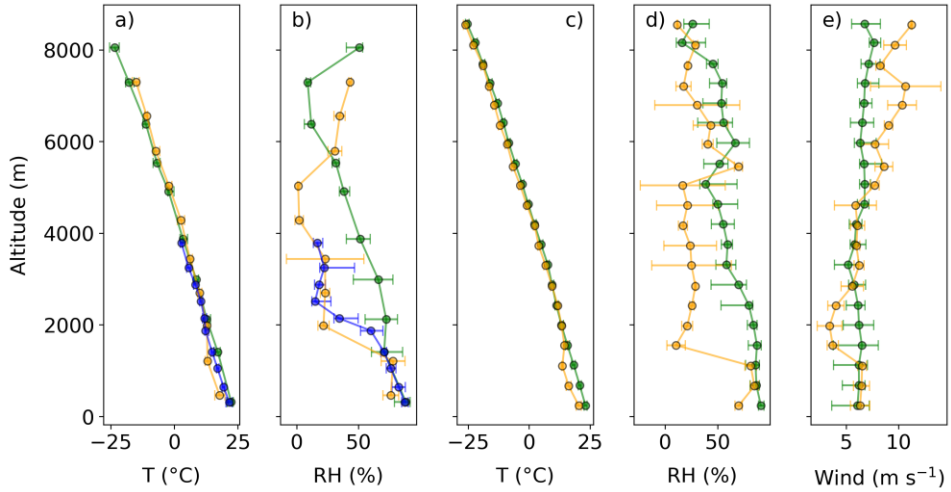

Figure 3: Vertical distribution of meteorological variables as measured (a-b) in-situ by the Falcon and (c-e) with dropsondes launched from the King Air. Shown are temperature (T), relative humidity (RH), and wind speed (only for King Air) grouped into similar air mass source categories (orange = North America; green = Caribbean/North Africa; blue = Ocean). Markers are median values and whiskers are 25th/75th percentiles. Data were unavailable for the Ocean category for dropsondes.

3. Figure 5: What is the type of aerosol diameter used in Figure 5? Is it electrical mobility diameter or aerodynamic diameter?

Response: The aerosol diameters used in Figure 5 are based on optical techniques (e.g., LAS and FCDP) and not those mentioned in the comment. We feel our explanation of diameters in Section 2.2 was sufficient and no extra text is warranted in figure captions about this.

4. Figure 8: I suggest adding a legend to the figure to explain the color representations.

Response: We updated Figure 8 to include the legend in the manuscript as shown below.

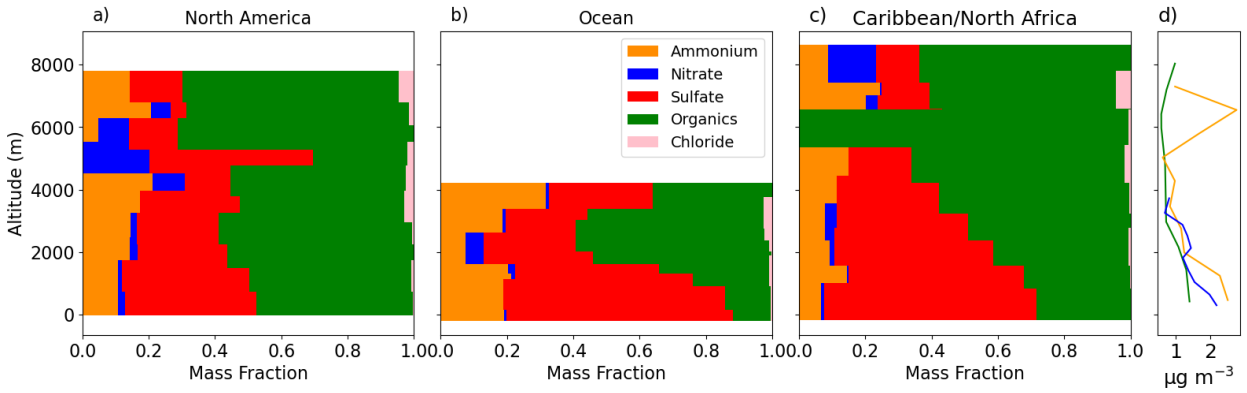

Figure 8: Vertical distribution of AMS (a-c) speciated mass fractions (red = sulfate; green = organics; blue = nitrate; orange = ammonium; pink = chloride) and (d) total mass concentrations for each air mass type (orange = North America; blue = Ocean; green = Caribbean/North Africa).

5. Why does the sulfate concentration decrease with increasing altitude?

Response: The primary source of sulfate is likely the marine boundary layer and the free troposphere did not have a strong influence on sulfate levels. We did not feel the need to make changes to the manuscript in response to this comment.

6. Figure 9: How do you calculate MForangic? Seaspray contains a lot of NaCl, which is a refractory material to the AMS. I suggest using AMS organic mass/PM1 to get MForangic.

Response: The mass fraction (e.g., $MF_{organic}$) was determined by dividing the mass of each non-refractory species found by the AMS by the total mass of non-refractory species detected. Since AMS instruments generally detect particles within the size range of 0.06 – 0.6 micrometers, they cannot adequately characterize sea spray particles, especially also because they are comprised of refractory substances that the AMS struggles with. So in a way, we did quantify already AMS organic mass divided by PM_1 since the AMS is measuring close to what can be considered PM_1. As a result, we don't think changes are needed in the paper to address this comment.