Manuscript No.: ACP-2021-300

Title: Vertical profiles of trace gas and aerosol properties over the Eastern North Atlantic: Variations with season and synoptic condition

The paper presents results of vertical profiles of gas and aerosol properties during ACE-ENA that were measured in summer 2017 and winter 2018. The combination of instruments onboard the aircraft (FIMS, AMS, SP2, thermal denuder, etc.) allowed for a thorough analysis of the impacts of seasonal differences, transport pathways, and synoptic situations on atmospheric composition (gas and aerosol) and aerosol microphysical and cloud-nucleating properties. It is well-written and an important contribution towards the understanding of factors controlling variability in aerosol properties, including CCN, in the eastern North Atlantic region. I only have a few very minor comments.

Response: We thank anonymous referee #1 for the positive feedback. We have revised the manuscript accordingly, and please find our point-to-point responses below.

Line 127: Should be a lower cut-off size of 10 nm.

Response: Thank you. This information has been corrected.

Figure 3 caption: Please describe what the 3 colors correspond to.

Response: The three colors (red, green, and blue) are used to represent the three different clusters of back trajectories. This information has been added to the caption of Fig. 3.

Line 157: Change to "North America"

Response: Thank you. The typo has been corrected.

Line 475: Should be 35 nm.

Response: The value has been corrected.

Lines 480 – 482: *What does "mixing" refer to here?*

Response: The sentence has been reworded as:

"The gradual change of CO mixing ratio and $m_{\rm BC}$ at altitudes above 1500 m also suggests the mixing between the layers of high Aitken-mode concentration due to NPF and long-range transported continental emissions."

It would be helpful to provide a brief description of what is meant by a decoupled MBL since it Is an important topic in the paper.

Response: Thank you. Additional discussion on the formation of the decoupled MBL is now added in Sect. 3.5.1:

"As the MBL deepens, the turbulence produced from surface-heating and cloud-top radiative cooling becomes insufficient to maintain a well-mixed layer. Consequently, the MBL begins to "decouple" into a surface mixed layer and an upper decoupled layer (Wood and Bretherton, 2004; Bretherton et al., 2010)."

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The manuscript presents a nice summary of vertical profiles of size-resolved aerosol concentrations and trace gas mixing ratios observed during the two seasons of the recent DOE ACE-ENA campaign. Clear differences in both aerosol and gas concentrations are observed between the winter and the summer with higher concentrations generally observed in the summer. The only exceptions are carbon monoxide and boundary layer ozone concentrations, which were elevated in the winter relative to the summer. Typical bimodal marine aerosol size distributions are presented for the boundary layer, which the authors use to infer CCN-activated vs. -unactivated (i.e., "pre-CCN") fractions. Elevated pre-CCN concentrations in the freetroposphere support the well-established idea that new particle formation does not tend to occur in the marine boundary layer due to the existing aerosol condensational sink. Back of the envelope calculations of the entrainment rate support the idea that these pre-CCN are entrained into the boundary layer, where they grow to CCN-relevant sizes. Overall, the manuscript is well written and the data are well analyzed and presented. Nice job! The paper is relevant for Atmospheric Chemistry and Physics, and I recommend that it be accepted for publication. While I have no major comments or corrections, I did note the following minor issues while reading the manuscript:

Response: We thank anonymous referee #2 for the positive feedback. We have revised the manuscript accordingly, and please find our point-to-point responses below.

Line 475: Change "~350 nm" to "~35 nm".

Response: The value has been corrected.

Please provide a link (preferably https://doi.org/...) directly to the ENA dataset under the Data Availability section.

Response: Thanks. The website linking to the ACE-ENA campaign is now added in the Data availability section.

Figure 2a: Is the map of the Azores from geographic guide net subject to copyright and reproduced here with permission?

Response: The original maps are now replaced by maps obtained from Google Maps, which can be reproduced openly.

Figures 6-10: Please note in both the captions and in the axis labels if the concentration units are standard (scm-3 or cm-3 STP) or volumetric.

Response: The concentration units are standard (STP), as noted in the figure captions. Due to the limited spaces for the axis labels (e.g., Fig. 8-10), and for consistency of the manuscript, we introduce this STP information in the captions only.

Tables 2-3: Same comment as above regarding standard or volumetric units and consistency between the captions and the table headings.

Response: Thank you. This information is now included in the captions of Tables 2 and 3.