

## Comment on acp-2022-341

Anonymous Referee #1

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Referee comment on "African smoke particles act as cloud condensation nuclei in the wintertime tropical North Atlantic boundary layer over Barbados" by Haley M. Royer et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-341-RC1>, 2022

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### General comments

This manuscript titled "African smoke particles act as cloud condensation nuclei in the wintertime tropical North Atlantic boundary layer over Barbados" by Haley M. Royer et al. is based on analyses of size-resolved particle composition (semi-quantitatively) and cloud condensation activity (quantitatively) data and dust concentration data. It concludes that African smoke is important for the atmospheric processes and cloud formation over the Caribbean. The addressed scientific question is well within the scope of ACP; the data and ideas in this manuscript are novel; and the manuscript is good structured in general.

However, the explanation of scientific methods including details in experimental settings and data analyses and uncertainties of the size-resolved chemical composition is unconvincing. In addition, the writing and interpretation especially in the Results and Discussion section are rough and need to be revised. The authors thus need to make careful revisions and corrections to improve the overall quality of the paper for publication in the journal. I would recommend the editor reconsider the manuscript only after a major revision by the authors.

### Specific comments

- Lines 90-102: whereas the possible compositional changes of smoke particles have been well explained the explanation of morphological changes is in lack. It's better to add the latter, too.
- Lines 102-104: This sentence should be rewritten.
- Line 185: since "EDX is considered a semiquantitative method" the uncertainty of this method in determining the size-resolved chemical composition should be elaborated because it directly influences the conclusions.

- Line 188: “organic material, sea spray, dust, and anthropogenic emissions” are not parallel categories. Please revise.
- Lines 192-194: “Samples collected on Si ... generate a background signal as well.” Does this mean except for the “carbon-coated copper grids” you also used silicon filters to collect particles for CCSEM/EDX analysis? This information should be added.
- Lines 198-201: “The algorithm operates by ... and existing literature.” The details in the K-means clustering analysis and the assignment of particle types should be explained at least in the supplement. How the number concentration-size distributions in Figs. 5 and S4 were determined should also be explained.
- In the section “Size-Resolved CCN Measurements and Data Analysis”, the details in the experimental settings including flow rates of the CCNC, CPC, DMA, and SMPS should be explained.
- Lines 243-244: “Calculation of activation ... in Pöhlker et al., 2016.” These should be explained at least in the supplement.
- Line 255: Should “Sampling Conditions” be replaced by “Air Mass Characteristics”?
- Lines 260-267: please restructure this part. For example, only from Fig. 1, one cannot see the result described in lines 261-262. In addition, it is better to describe the figure before it is referenced.
- Lines 269-271: “suggesting that ... onto larger transported continental aerosols (Tomlin et al., 2021).” This part needs to be modified. For example, “suggesting that transported particles overwhelmed the background marine particle loading so that the small Aitken mode particles possibly coagulated onto the larger transported continental particles (Tomlin et al., 2021).”
- Lines 291-346: it’s better that the morphology of aged sea spray, mineral dust, and sulfate particles are also explained (and interpreted from the viewpoint of transport histories (e.g., history of atmospheric moisture conditions) if applicable).
- Lines 299-305: the explanation on aged sea spray is not enough. Based on the explanation, one cannot see if the related particle and spectra presented in Fig. 3 is of aged sea spray particle or not.
- Lines 315-317: the existence of N or ammonium cannot be seen from the related particle and spectra presented in Fig. 3.
- Lines 325-327: how about the morphologies of the smoke particles in this study, which type is dominant?
- Lines 330 and 334: please correct the elements of dust components.
- Lines 341-342: “This scarcity of ... respectively.” This sentence is incomplete.
- Lines 362-364: “These finds ... for smoke (Figure S2 and S3).” First, the origin of the data in Figs. S2 and S3 need to be explained. Next, can the CO variation during the observation period of this study indicate the arrival of smoke particles during the three CAT events?
- Lines 378-382: please rewrite this part to make it concise.
- Lines 391-393: “A large decrease ... observed in Figure 2.” This part needs more explanations. Do the authors know any existing study that reported the condensing of marine biogenic sulfur precursors onto transported particles? Do the authors mean “The large decrease in sulfate number fraction during CAT events might be caused by the condensation of marine biogenic sulfur precursors and/or Aitken mode sulfate-containing particles onto the large, transported particles as indicated in Fig. 2.”?
- Lines 399-401: please explain “Bin sizes for each decade” and “bin size”; please rewrite this sentence.
- Line 410: why these small particles are less hygroscopic should be tentatively explained.
- Lines 414-423: the difference between marine conditions and smoky conditions should be quantitatively explained and the data should be tabled for all S conditions.
- Lines 418-420: the first half and the second half of the sentence are repeating each other.
- Lines 428-432: “However, one finding of note ... African aerosols.” Please rewrite this part to make it concise.

- Line 431: "such as dust that activate as CCN". Dust might haven't acted as CCN but it still could have influenced the activation diameter and thus the overall estimated kappa. I suggest deleting "that activate as CCN".
- Lines 441-443: the existence of organics may have lowered the observed kappa. Please rethink this conclusion.
- Lines 444-447: please explain the box plots in detail and the meaning of the whiskers of  $d_{50}$ .
- Lines 458-459: "In Fig 7b, there is a clear and direct relationship" should be rewritten. For example, "Figure 7b shows a positive correlation". In addition, this positive correlation should be further elaborated with the data for example in Table 1.
- Line 460: "may act as CCN" should be "may have acted as CCN".
- Lines 464-466: "The large number of smoke particles ... on sulfate or organic particles." Do you want to say that the number of sulfate and organic particles were not enough for the measured CCN counts?
- Lines 469-472: what are the possible surfactants? Could you explain?
- Lines 472-479: First, can any evidence be given from the single particle composition data that aging of particles occurred. Second, what you want to conclude from this part is not clear.
- Lines 485-486: "It is likely that ... of factors." seems redundant. It's better to delete it.
- Lines 450-488: the writing and organization of this paragraph are quite rough and should be revised.
- Table 1 should be modified according to <https://www.atmospheric-chemistry-and-physics.net/submission.html#figurestables>
- Line 508: the activation diameter should be explained with the measured S condition.
- Line 513: "Overall, we find that smoke has a larger effect on CCN number concentrations than dust." Does this conclusion apply for all time or for the studied period or season?
- Line 522: what's the difference between AOD and AOT? And where are the data of Figs. S2 and S3 from?

## Technical corrections

- Line 73: Should "though" be replaced by "however" or other similar phrases?
- Line 143: "composition" should be "compositions".
- Line 182: "affects" should be "affect".
- Line 218: "Rose et al., 2008" should be "Rose et al. (2008)". Please modify similar expressions in other places.
- Line 227: "0.71" should be "0.71 %".
- Lines 251-252: "single particle data (e.g., CCSEM-EDX), and air mass history (e.g., NOAA's HYSPLIT model)", the "e.g., " in the parentheses should be deleted.
- The color shadings in Fig. 2 should have been explained in the caption.
- Line 350: "represent" should be "present"?
- Line 362: "observe" should be "observed".
- Lines 369-370: "wildfire smoke appears to overwhelm the number fraction of the submicron aerosol loading" should be "wildfire smoke particles appeared to dominate the number fraction of submicron aerosol." Please also correct the wrong use of overwhelm in other places throughout the manuscript, e.g., line 510.
- Line 388: "Figure 4" should be "Figure 5".
- Line 452: "N Atlantic MBL" should be "North Atlantic MBL".
- Line 455: "Fig 7a suggests" should be "Figure 7 suggests". Please refer to <https://www.atmospheric-chemistry-and-physics.net/submission.html#figurestables>

- Line 476: “to oxidants” should be “to oxidize”.