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Title: Long-range transported continental aerosol in the Eastern North Atlantic: three multiday event regimes influence cloud condensation nuclei

Responses to Anonymous Referee #2

1.[Referee #2]: *Concerning the STP correction, I would like to mention that I was asking if the extensive parameters stated in the manuscript and presented in the figures (e.g. number concentrations) are given at standard temperature and pressure (usually 20 DegC and 1013 hPa) or at ambient conditions. The added sentence ("'Pressure for aerosol instruments is given at ambient conditions if not differently stated.") is not really addressing this.*

[Resp.]: To further clarify this point the sentence has been rewritten in the revised version of the manuscript and it reads as:

> **Page 4, Line 8**: "The pressure and temperature for the aerosol data is the same as reported in the ARM data archive which is given at ambient conditions to allow for direct comparision with the other data at ENA. If desired for future global comparion, data can be calculated at Standard Temperature and Pressure (STP) using the AOS meteorology data."

Reformating the data to Standard Temperature and Pressure (STP) would allow for comparison with other locations. However, the main focus of this manuscript is on ENA rather than global comparisons. We also did not report at STP here since different temperatures can be used for STP reporting. For example, the World Meteorological Organization and the Global Atmosphere Watch recommends reporting data at 0 ^{0}C and 1013 hPa, rather than at 20 ^{0}C as was mentioned here. For this reason, we thought this selection would be best made as is pertinent for the specific intercomparison to be made in future work.

2.[Referee #2]: *Concerning the calibration of HTDMA and CCNC: I think it is not sufficient to write "Typically, CCN counter and HTDMA instruments are calibrated once per year." and just reference instrumental hand-books. The authors should state the actual date/time of the calibrations, as well as check, and verify their data. For example, how did the calibration with ammonium sulfate or sodium chloride of HTDMA and CCNC look like? How was the sizing of the DMAs verified? This is an essential part of experimental work.*

[Resp.]: We appreciate Referee 2's concerns about instrument calibrations.We agree with Referee's 2 opinion that the calibration of the instruments is an essencial part of the experimental work. All data here is from the AOS at ENA, and all ARM AOS data is collected, ingested, and quality controlled by the U.S. DOE ARM User Facility, as indicated in Uin et al., 2019 and reported below for Referee 2's convenience:

- Uin, J., Aiken, A. C., Dubey, M. K., Kuang, C., Pekour, M., Salwen, C., Sedlacek, A. J., Senum, G., Smith, S., Wang, J., Watson, T. B., and Springston, S. R.: Atmospheric Radiation Measurement (ARM) Aerosol Observing Systems (AOS) for Surface-Based In Situ Atmospheric Aerosol and Trace Gas Measurements, J. Atmospheric Ocean. Technol., 36, 2429–2447, https://doi.org/10.1175/JTECH-D-19-0077.1, 2019

  > "Individual AOS instruments are operated under the direction of instrument mentors. Instrument mentors are experts of their respective instruments who provide the procurement specifications, perform or oversee calibrations and maintenance, work with the ARM Data Quality Office to automate (where possible) data quality assurance/quality control (QA/QC), develop documentation for and train site operators, and are the

ultimate authority for instrument data quality (Peppler et al. 2016). The AOS instrument mentors review deployment details and, if necessary, modify the written protocols for instrument operation.
Once a field site is set up, instrument mentors typically travel to the site for final integration, calibration and instrument verification. A one- to two-week period of data validation prior to the official start date allows time for data inspection and addressing unforeseen circumstances.
During the campaign, on-site operators fill in daily checklists with mentor-defined criteria for normal instrument response. Proper instrument operation is continually verified through data review by operators, mentors, the ARM Data Quality Office who have automated and manual review processes, or the principal investigator (PI) for the specific campaign who have nearly real-time access to processed data."

Peppler, RA, Kehoe, KE, Sonntag, KL, Bahrmann, CP, Richardson, SJ, Christensen, SW, McCord, RA, Doty, DJ, Wagener, Richard, Eagan, RC, Lijegren, JC, Orr, BW, Sisterson, DL, Halter, TD, Keck, NN, Long, CN, Macduff, MC, Mather, JH, Perez, RC, Voyles, JW, Ivey, MD, Moore, ST, Nitschke, DL, Perkins, BD, and Turner, DD. Quality Assurance of ARM Program Climate Research Facility Data. United States, doi:10.2172/948030, 2008

To address Referee #2's concerns, we added the following sentence and the reference to Peppler et al., 2008 and Uin et al. 2019 to the revised version of the manuscript.

Page 4, Line 11: "All of the data is collected, ingested, quality controlled by the U.S. DOE ARM User Facility (Peppler et al., 2008, Uin et al., 2019)."

To further address instrument calibrations directly, calibration methodology is standardized for all ARM datasets and publicly reported in detail in the ARM instrument handbooks that can be found online. We cite the official ARM handbooks written by the individual instrument mentors for ARM in the manuscript in both Section 2 and Table 1. We also provide direct links to the documents in the reference section of the manuscript. For Referee 2's convenience, we report below the ARM handbook sections concerning CCN and HTMA calibration methodologies here.

- Uin, J.: Cloud Condensation Nuclei Counter (CCN) Instrument Handbook, 2016, https://www.arm.gov/publications/tech_reports/handbooks/ccn_handbook.pdf

"**6.5 Calibration Database**: During deployment the CCN is periodically calibrated by instrument mentors. CCN calibration involves generating and size-selecting ammonium sulfate particles and recording their total number concentration before and after activation in the CCN as a function of particle size. This is done for several humidifier column temperature gradients. Next, the 50% activation diameter (particle diameter where 50% of the generated ammonium sulfate particles are activated) is calculated for each humidifier column temperature gradient. As the activation characteristics of ammonium sulfate are known [2], supersaturation % can be calculated from the 50% activation diameter to establish/verify the relation between the temperature gradient of the humidifier column and the supersaturation %. Calibration coefficients are entered into a CCN configuration file and applied automatically by the instrument software. Coefficients are also recorded in the ARM Operation Status System (OSS, https://oss.arm.gov/oss.php) and archived by the instrument mentors."

"**12.0 Calibration**: The CCN is calibrated by the manufacturer before delivery to the user and during instrument maintenance at the manufacturer's facilities. The instrument mentors also typically perform calibration before and after each deployment at conditions (altitude) similar to the measurement site and during deployment if it has been more than a year from the last calibration and the deployment is not yet

ending. The calibration schedule is flexible because it depends on the availability of the calibration equipment (calibration scanning mobility particle sizer spectrometer [SMPS]). Calibration coefficients are entered into a CCN configuration file and applied automatically by the instrument software. Calibration results are also used by the mentors to assess the overall condition of the instrument, especially the humidifier column. Manufacturer's calibration includes:

- Supersaturation calibration with ammonium sulfate aerosol particles (see below).
- Size calibration of the OPC with National Institute of Standards and Technology (NIST)-traceable polystyrene latex (PSL) particles.
- Flow calibrations with a precision flow meter.

Mentor calibration of the CCN involves generating and size-selecting ammonium sulfate particles and recording their total number concentration before and after activation in the CCN as a function of particle size. This is done for several humidifier column temperature gradients. Next, the 50% activation diameter (particle diameter where 50% of the generated ammonium sulfate particles are activated) is calculated for each humidifier column temperature gradient. As the activation characteristics of ammonium sulfate are known [2], supersaturation % can be calculated from the 50% activation diameter to establish/verify."

[2] Rose, D, GP Frank, U Dusek, SS Gunthe, MO Andreae, and U Pöschl. 2007. "Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment." Atmospheric Chemistry and Physics Discussions 7(3): 8193−8260, http://doi.org/10.5194/acpd-7-8193-2007

- Uin, J.: 3002 Humidified Tandem Differential Mobility Analyzer Instrument Handbook, 2016, https://www.arm.gov/publications/tech_reports/handbooks/htdma_handbook.pdf

"**6.5 Calibration Data Base**: During deployment, the HT-DMA system is periodically (every six or twelve months) calibrated by the instrument mentors. This includes recording the growth factor of generated aerosol particles with a known chemical composition (ammonium nitrate or ammonium sulfate), as a function of RH. The growth factor value is compared to the Köhler model's theoretical values to ensure the HT-DMA is in a proper operational state. No correction factors are usually derived. Calibration data are collected and maintained by the instrument mentors."

"**12.0 Calibration**: The HT-DMA system is calibrated and validated by the manufacturer prior to delivery and during routine instrument maintenance tasks, performed at the manufacturing facilities. During deployment, subsets of the manufacturer's calibration are performed every six or twelve months by the instrument mentors. Manufacturer's calibration includes:

- Measuring the size distribution of a NIST traceable monodisperse calibration aerosol (PSL) to validate the proper operation of the DMAs.
- Measuring the growth factor of generated aerosol particles with a known chemical composition (ammonium sulfate or ammonium nitrate) as a function of RH, and comparing the resulting curve to a theoretical one from the Köhler model in order to validate the proper operation of the HT-DMA.
- Calibrating the high-voltage power supply, absolute pressure sensor, and laminar flow element against precision instruments and recording-derived correction factors in the software configuration files, which is used automatically.

Calibration and validation completed by the instrument mentors, similar to the manufacturer's calibration activities, includes:

- Recording the growth factor of generated aerosol particles with a known chemical composition (ammonium sulfate or ammonium nitrate), as a function of RH and comparing the resulting curve to a theoretical one from the Köhler model to validate the proper operation of the HT-DMA. If the recorded growth factor curve differs from the theoretical curve by more than the accuracy value specified by the manufacturer, it should be investigated on a case-by-case basis and corrected.
- Correction factors (if any) are derived according to the issue in question. Please consult the manufacturer's manual for more details."

Specifically, during the data period reported here, two calibrations (2016-09-29, and 2017-05-28) were performed by Janek Uin, a co-author on this manuscript and the instrument mentor for both instruments at ENA. He reported that there were no abnormal findings in the calibrations. We have added the following sentence to the text:

> **Page 4, Line 13**: " "Calibrations were completed in accordance with the ARM instrument handbooks. No abnormalities were found during the periods that would affect the data reported here."

In summary, we have added clarifying sentences about how the data is handled in terms of data quality and instrument calibrations. We also report details in the references cited in the manuscript as shown above in case the reader would like further information and detail. Explicit description of instrument calibration methodologies and specific reports were not added to the manuscript since they are standardized by the ARM Facility as described and referenced above. Adding this information for all of the datasets reported from the AOS at ENA would add significant length to this manuscript, and we find this information to be beyond the scope of this manuscript since instrument calibrations are provided by the ARM Facility and references to those details can be found online.

References:

Peppler, RA, Kehoe, KE, Sonntag, KL, Bahrmann, CP, Richardson, SJ, Christensen, SW, McCord, RA, Doty, DJ, Wagener, Richard, Eagan, RC, Lijegren, JC, Orr, BW, Sisterson, DL, Halter, TD, Keck, NN, Long, CN, Macduff, MC, Mather, JH, Perez, RC, Voyles, JW, Ivey, MD, Moore, ST, Nitschke, DL, Perkins, BD, and Turner, DD. Quality Assurance of ARM Program Climate Research Facility Data. United States, doi:10.2172/948030, 2008

Uin, J., Aiken, A. C., Dubey, M. K., Kuang, C., Pekour, M., Salwen, C., Sedlacek, A. J., Senum, G., Smith, S., Wang, J., Watson, T. B., and Springston, S. R.: Atmospheric Radiation Measurement (ARM) Aerosol Observing Systems (AOS) for Surface-Based In Situ Atmospheric Aerosol and Trace Gas Measurements, J. Atmospheric Ocean. Technol., 36, 2429–2447, https://doi.org/10.1175/JTECH-D-19-0077.1, 2019

Uin, J.: 3002 Humidified Tandem Differential Mobility Analyzer Instrument Handbook, 2016, https://www.arm.gov/publications/tech_reports/handbooks/htdma_handbook.pdf

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WMO/GAW Aerosol Measurement Procedures, Guidelines and Recommendations, 2nd Edition. https://library.wmo.int/doc_num.php?explnum_id=3073, 2016.