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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Ultra-clean and smoky marine boundary layers frequently occur in the same season over the southeast Atlantic" *by* Sam Pennypacker et al.

Anonymous Referee #4

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Manuscript summary

The authors analyze surface observations of aerosol, gas phase composition, and cloud properties at Ascension Island over a period of 16 months, acquired during the Layered Atlantic Smoke Interactions with Clouds (LASIC) campaign. Back-trajectory calculations support the analysis. The authors distinguish three aerosol states at Ascension Island: Background conditions, polluted conditions, and ultraclean conditions. Ultraclean conditions are defined based on a daily median concentration of aerosol particles (CCN) with dry diameters between 60 nm and 1 μm < 50 cm^{-3}. The authors find 41 days with ultraclean conditions at Ascension Island. All of these occur dur-

ing the South-West African biomass burning season. A portion of the ultraclean days also exhibits carbon monoxide and refractory black carbon levels above background. Apart from ultraclean days, boundary layer CCN concentrations at Ascension Island are significantly elevated above background levels. No days with ultraclean conditions are found outside the biomass burning season, which defines background conditions. The authors conclude, based on analysis of carbon monoxide and refractory black carbon levels, statistics of precipitation and liquid water path at Ascension Island, and back-trajectory calculations that CCN concentrations are low on the ultraclean days not because originally clean air has been advected to Ascension Island, but because enhanced coalescence scavenging in low clouds has strongly reduced CCN in polluted air masses. This is an interesting result because it points to a more complex interaction between (anthropogenic) aerosol and cloud properties in the region, with causal links in both directions.

Review summary

In their analysis of the observations the authors accumulated a good amount of circumstantial evidence to render their hypothesis plausible, although the analyzed data are specific to conditions at Ascension Island only and hence do not establish a causal connection between conditions on ultraclean days and processes that may give rise to them. Although not quantitative, the back-trajectory analysis is helpful. The study is, as the authors point out in their closing statements, a good motivation and starting point for subsequent investigations.

There are a few points that I would ask the authors to look after, listed below. Otherwise, the manuscript is in good shape.

Major comments

- Could there be other explanations for the ultraclean days than enhanced coalescence scavenging in low clouds with higher liquid water content? E.g., is it possible that on the ultraclean days, the polluted air has entrained earlier into the boundary layer, hence spent a more time there compared to other days during the biomass burning season? A longer sojourn in the boundary layer would give coalescence scavenging more time to deplete the aerosol. Please comment and if applicable, discuss in the manuscript.

- Please calculate the average speed of the trajectories between 35 S and Ascension Island. Is there a difference in advection velocity between the ultraclean and non-ultraclean days during the biomass burning season? If yes, discuss what this could mean for the processes that cause ultraclean conditions.

- The criterion for what makes ultraclean conditions varies between works. Albrecht et al. (doi:10.1175/BAMS-D-17-0180.1), e.g., define ultraclean conditions as having aerosol concentrations of less than 10 cm^{-3} in the nominal range between 0.06 - 1.0 μm., while in the present work it is < 50 cm^{-3}. Please add a passage mentioning the different criteria and explain why in the present work the criterion of < 50 cm^{-3} was chosen.

- How robust is the number of ultraclean days to the UHSAS < 50 cm^{-3} criterion?

- "... with the correlation statistically indistinguishable from zero (r^2 = 0.06), ..."

To make this statement you /must/ calculate the p-value of the linear regression/correlation. Without the p-value, there is no way of telling whether a correlation coefficient/coefficient of determination is statistically indistinguishable from zero, regardless of its numerical value.

Other comments

- Please check the text for sentences that can be simplified; some are hard to understand. For example,

"The relative invariance of isobaric boundary layer back trajectories between ultraclean and the most polluted days at ASI suggests that the potential for BBA entrainment set by the vertical separation of a smoke layer and the evolution of the boundary layer cloud field plays a larger role in upwind (e.g. at ASI) aerosol variability than a systematic difference in large-scale horizontal circulation in the boundary layer."

is rather difficult to decipher.

- Please mention the meteorological input that you used to drive the HYSPLIT model.

- 500 m trajectories are not isobaric.

- Please consider if the labeling of the abscissa in the plots that show data as a function of the month is precise enough to inform the reader on the actual point in time (are the vertical lines the 1st of the month or the 15th?)

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