Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-461-RC2, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Time-dependent entrainment of smoke presents an observational challenge for assessing aerosol–cloud interactions over the southeast Atlantic Ocean" *by* Michael S. Diamond et al.

Anonymous Referee #2

Received and published: 3 August 2018

Summary of manuscript

The manuscript presents an analysis of South-East Atlantic in-situ data of boundary layer and free tropospheric biomass burning aerosol and of boundary layer cloud properties from the NASA ObseRvtions of Aerosols above CLouds and their intEractionS (ORACLES) campaign. The manuscript identifies the importance of distinguishing, in particular in satellite studies, whether or not biomass burning aerosol is merely present in the free troposphere (FT) or has entrained into the boundary layer (BL). This insight

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Discussion paper

is obtained by correlation analysis of boundary layer cloud drop number with aerosol concentration in the BL and the FT from all ORACLES flights, and by contrasting two ORACLES flights with high FT aerosol concentrations but different BL aerosol concentration and cloud drop number. The analysis is supported with satellite data, WRF-AAM simulations, and a calculation of the time scale for entrainment of FT aerosol into the boundary layer based on theoretical considerations. A consistent picture arises: Biomass burning aerosol in the free troposphere, in particular as seen by satellite, is a poor predictor of boundary layer cloud properties (specifically cloud drop number), because entrainment of aerosol from the free troposphere into the boundary layer is a comparably slow process (time scale of days). The mere presence of FT biomass burning aerosol determined in satellite observations above the boundary layer cannot be taken as an indicator of an effect on BL clouds, and accordingly, satellite studies that use "snapshots" of the state of the system may suffer from a bias in determining the effect of the biomass burning aerosol on boundary layer clouds. However, once biomass burning aerosol has entrained from the FT into the BL, it gives rise to a response of BL cloud properties that is consistent with established understanding. In conclusion, the entrainment history of a given BL air mass needs to be taken into consideration when studying biomass burning aerosol-cloud interactions, best by pursuing a Lagrangian framework that follows the air mass and accounts for entrainment of free tropospheric aerosol into the boundary layer.

The introduction gives a comprehensive but brief summary of interactions between biomass burning aerosol and boundary layer clouds in the South-East Atlantic and of pertinent work on the subject. The scope and key findings of the manuscript are summarized in the last paragraph. The data and methods section describes the ORACLES campaign and the instrumentation used, and gives a brief overview of the satellite data analysis and of the modeling approach. The results section presents analysis of aerosol and cloud data from all ORACLES flights, and contrasts two flights that highlight the key insights of the work. The discussion section elaborates on the theoretical approach to calculating the time scale for entrainment of aerosol from the FT into the

BL. The final section summarizes the elements of the previous sections, provides essential context, and lays out the conclusions.

Summary of review

The manuscript is compelling, clear, and concise, and presents relevant and new insights. It is in very good shape; some minor points require attention.

Specific comments

Page 1, line 23: "Precipitation processes are also expected to obscure the relationship between above-cloud smoke and cloud properties in parts of the southeast Atlantic, although marine boundary layer carbon monoxide concentrations for two case study flights suggest that smoke entrainment history drove the observed differences in cloud properties for those days."

This sentence could be simplified for clarity; e.g.

"Precipitation processes can obscure the relationship between above-cloud smoke and cloud properties in parts of the southeast Atlantic, but marine boundary layer carbon monoxide concentrations for two case study flights suggest that smoke entrainment history drove the observed differences in cloud properties for those days."

Page 4, line 7: "... in situ data used is available at ..."

replace with

"... in situ data used are available at ..."

Page 5, line 15: "Only data from liquid clouds in the MBL (successful liquid cloud phase retrievals with effective temperatures below 280 K) ..."

Please specify what effective temperature is and adjust wording so that it is clear what "below" means (temperature or altitude).

Page 7, line 18: "This indicates that variability in aerosol properties immediately above the MBL has little immediate impact on the microphysics of the clouds below."

This seems to follow from Table 1, Row 7, rather than from the context of this sentence.

Page 8, line 26: "Observed CO (Fig. 6) is qualitatively consistent with the WRF-AAM output, with MBL average CO values on 31 August considerably above those from 4 September and among the highest seen during the deployment (all other flights shown in thin grey lines)."

It seems a bit too much to ask the reader to compare Fig. 5 b, c (WRF-AAM data) with Fig. 6 (ORACLES data) and to come to this conclusion. Please add, e.g., a time averaged vertical profiles of CO from Fig. 5 b, c to Fig. 6 to enable the comparison.

Page 12, line 7: "For instance, because the climatological MBL flow is southerly in the SEA, an instantaneous snapshot of smoke-cloud contact in the southern reaches of the domain may underestimate the microphysical effects by not accounting for their upstream manifestation. Similarly, apparently 'clean' cases in the northern part of the domain may have been polluted downstream, complicating efforts to compare 'mixed' and 'unmixed' statistics."

This sentence is a bit confusing. It seems to make more sense if "upstream" and "downstream" are switched.

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