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Comment on acp-2022-43

Anonymous Referee #3

Referee comment on "Source attribution of cloud condensation nuclei and their impact on stratocumulus clouds and radiation in the south-eastern Atlantic" by Haochi Che et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-43-RC3, 2022

General overview

This paper describes a modelling study using UKESM to attribute the source of CCN in south east atlantic clouds and investigate the impact of the different CCN on the marine Sc and the associated radiation. Overall, the paper is OK, with clear figures and an obvious story throughout. It is general well written but some of the definitions and explanations need to be tightened up, so that it is clear to the reader where the definitions apply. As a result of this, I recommend this paper be accepted after minor revisions (see below)

Comments

Page 1, Line 15 - Define SEA (south east atlantic) in abstract

Page 1, Line 17 - 18 - Could the authors quantify “the most to the annual average CCN…”, i.e. what percentage?

Page 1, Line 17 (and abstract) - Define BB (biomass burning)

Abstract - what is the definition of total nucleation and ensure the definition is consistent throughout the manuscript (See comment below for further information

Page 2, Line 25 to 27 - This statement is true when the BB airmass sits above and close to the inversion. It is not true when the BB mass is in the boundary layer or there is a gap between the absorbing air mass and the inversion. The authors need to add some text to clarify this statement, to avoid reader confusion.

Page 4, line 25 - Add Walters et al reference for GA7.1, https://gmd.copernicus.org/articles/12/1909/2019/gmd-12-1909-2019.html

Page 4, line 29 and 30 - "The κ-Kohler activation scheme is implemented, which use a hygroscopicity parameter of each aerosol mode, κ, to calculate the activated CCN." From the present description, this differs from the standard Abdul-Razzak and Ghan scheme in the UKESM, which is fine but could the authors add some information about why they have made this change and what is the advantage/impact of this change. The authors refer to Che et al (2021) throughout the paper but Che et al uses a different activation scheme. Is this important?

Page 4, Line 29 - This work uses UKCA-mode dust, which differs from CLASSIC dust used in the UKESM. Similar comment to above, such a difference is fine but could the authors add some information about why they have made this change and whether this is important in the base simulations or when considering comparison with earlier work.

Page 5 - Source attribution is achieved by switching off the emissions of BB, sea-salt, dust and DMS, respectively. While I understand why this has been done, how does switching off these emissions impact the simulation of cloud in these sensitivity tests. At present, this paper only shows cloud from the base simulations, so it is not possible to see what the impact of the changes in emissions are. Also, in switching of the emissions, I assume that the aerosol size distribution is impacted due to the removal of mass (and number). If so, does the change in size distribution impact the results or the conclusions? Does the change in size distribution influence any competition for vapour or the altitude at which water vapour is condensed?

Page 5 - the definition of "total nucleation" is confusing. On page 5, it is defined as the sum of boundary layer and binary nucleation, while in the abstract it is defined as "total nucleation (binary nucleation)". Then on page 9 (line 15) the authors state that "Total nucleation contributes more to CCN0.2% compared to boundary layer nucleation, indicating a contribution from the free troposphere". So what is total nucleation? Could the authors clarify what they mean and ensure consistency.

Page 7 line 14 - add "layer" after boundary

Page 8 - Figure 3, why is the scale for ccn concentration from dust negative? Is this correct? If so could the authors explain what is going on?

Page 9 line 3 to 7 - Do you see differing heating rates above the cloud between the simulations with and without BB? Also, does the cloud top height differ between these 2 simulations. It would be useful to show such a difference since this will validate the authors speculation. At present Figure 3 only shows the baseline LWC, how do the clouds evolve in the sensitivity tests?

Page 11 line 15 to 17 - "This may be due to SO2 emitted from anthropogenic sources, which can increase CCN0.2% by nucleation." The authors speculate here but they do not demonstrate this. Is there anything else that could cause this? If so, the authors should state this. Ideally, it would be good to show this, if it is possible, with another sensitivity run.

Page 12 line 14 - 17 - "The increase in maximum supersaturation due to BB aerosols is caused by their shortwave radiation absorption effect. As it can warm the air due to its absorption of shortwave radiation, BB aerosol can enhance the inversion layer over clouds, preserving water vapour within the boundary layer and increasing the maximum supersaturation, consistent with the finding in Che et al. (2021)". This description is potentially misleading and confusing. In particular, "The increase in maximum supersaturation due to BB aerosols is caused by their shortwave radiation absorption effect", is not correct since the increase in boundary layer max supersaturation is caused by a dynamic feedback that results from the increased absorption. For example, Johnson et al (2004) demonstrated that an absorbing layer directly above the marine Sc deck will lead to an enhancement of the inversion strength, which will reduce the entrainment of warm and drier free troposphere air into the boundary layer. This leads to an increase in LWP compared to a simulation with no absorbing layer. In the work under review, the authors only focus on the preservation of the water vapour in the boundary layer and do not address the temperature profile. The impact of the BB layer on the inversion is referred to a lot but the authors do not present any profiles (potential temperature, vapour or RH) to demonstrate a strengthening of the temperature or moisture inversion. Could the authors add these to prove these statements about strengthening inversion?

I appreciate that the authors refer to Che et al 2021 as the reference for the impact of absorbing aerosol over marine Sc and the supplemental plots in Che et al 2021 show this impact. However, the simulations presented in this work seem to use some different parametrisations, e.g. activation, dust, so the results may not be directly comparable. Also the description in Che et al is as follows, "Near the coast, BBAs are generally above the underlying cloud deck; the absorption aerosols could strengthen the boundary layer inversion (Fig. S4) and thus decrease the dry air entrainment, resulting in increased humidity and hence supersaturation". This is a better description than "preserves water vapour in the boundary layer", since it is the RH that matters.

Page 16, line 19 to 21 - "The higher LWP caused by BB reflects the critical role of the radiative effect of BB aerosol in affecting cloud properties, and is consistent with our previous finding (Che et al., 2021)." I think it is important to state that this critical role will only occur where the absorbing layer is directly above the inversion. If there is a gap between the absorbing aerosol layer and the cloud so that the absorbing layer does not impact the inversion then this response is not seen. This is demonstrated in Haywood, J. M., S. R. Osborne, P. N. Francis, A. Keil, P. Formenti, M. O. Andreae, and P. H. Kaye, The mean physical and optical properties of regional haze dominated by biomass burning aerosol measured from the C-130 aircraft during SAFARI 2000, J. Geophys. Res., 108(D13), 8473, doi:10.1029/2002JD002226, 2003

Page 19, line 14 to 16 - " This is mainly because BB aerosol, in addition to acting as CCN like anthropogenic aerosol, also can increase the maximum supersaturation through the radiative effect of its shortwave absorption, thus additionally increasing the CDNC. " Again this comment is similar to the previous comment - I do not like this statement and I think it is misleading. BB aerosol does not increase the maximum supersaturation of the boundary layer. Instead, when BB aerosol is directly above the inversion the associated absorption will strengthen the inversion, reduce entrainment mixing of warm dry air from aloft, which will permit a higher RH and max supersaturation. If the BB aerosol is separated from the cloud layer or in the boundary layer then the associated absorption will lead to know change in the supersaturation or a decrease. Could the authors be more specific with this type of statement?