Replies to the editor's comments by Haochi Che et al.

We would like to thank the editor for the thoughtful and detailed comments on our paper. We feel that in responding to these comments and suggestions, we have significantly improved the quality and readability of the paper.

The editor's comments are provided in **blue in the following**, and our responses are in **black**. Changes to the manuscripts made in response to the reviewer are in **green**. In addition, some changes in the manuscript are not shown in this response but are highlighted in the revised manuscript.

Editor's Report:

Three reviewers have provided comments, with reviewer 1 suggesting minor revisions that account for the known variation in aerosol vertical structure from July-September, reviewer 2 also suggesting minor clarifications, primarily of the model and how it is used, and reviewer 3 also suggesting minor revisions, primarily related to the language.

After reviewing the comments, the authors' response, and the overall manuscript, it is my determination that the authors have not sufficiently responded to the intent of the reviewer comments. I suggest the authors give this another go-through. Reading through the manuscript, I also have several specific comments, listed below in primarily chronological order, that I would like to see addressed before the paper is finalized.

Abstract: Please read this over more carefully. Keep in mind many readers will not look past the abstract.

1. The description of 'total nucleation' in the abstract as 'tropospheric and stratospheric nucleation' is confusing. This is the first time the reader encounters this term. It's worth including an additional sentence here to define the term.

We thank the editor for the suggestion. Total nucleation includes nucleation in and above the boundary layer. Our model results find that boundary nucleation has a small effect on marine boundary layer CCN (0.2%), probably because boundary nucleation is suppressed by the sufficient condensation sinks provided by the marine boundary layer aerosols (e.g., sea salt), or limited by the available organic aerosols. In contrast, particles nucleated above the boundary layer can grow and subside into the marine boundary layer, contributing to the majority of CCN (41 % on annual average), which is consistent with the findings of Merikanto et al. (2009), who found that 45 % of the global marine boundary layer CCN (0.2%) was contributed by the free troposphere nucleation.

In order to analyze the height at which nucleation predominantly occurs, we calculated 2016-2017 mean vertical distribution of nucleation modal aerosols in the cloud box region, as shown in Figure 1. As can be seen from the figure, the nucleation mode aerosols peak at 200hPa, indicating nucleation mostly occurs in the free and upper troposphere. Therefore, we consider the nucleation in the free and upper troposphere to be the dominant source of CCN in the boundary layer.

To make it clearer, we removed the term "total nucleation" from the abstract and emphasized nucleation in the free and upper troposphere. The sentence was modified as follows.

P1L16-18

Overall, free and upper troposphere nucleated aerosols are the dominant source of boundary layer $CCN_{0.2\%}$, contributing an annual average of ~ 41 % as they subside and entrain into the marine boundary layer, which is consistent with observations highlighting the important role of nucleation for boundary layer CCN.

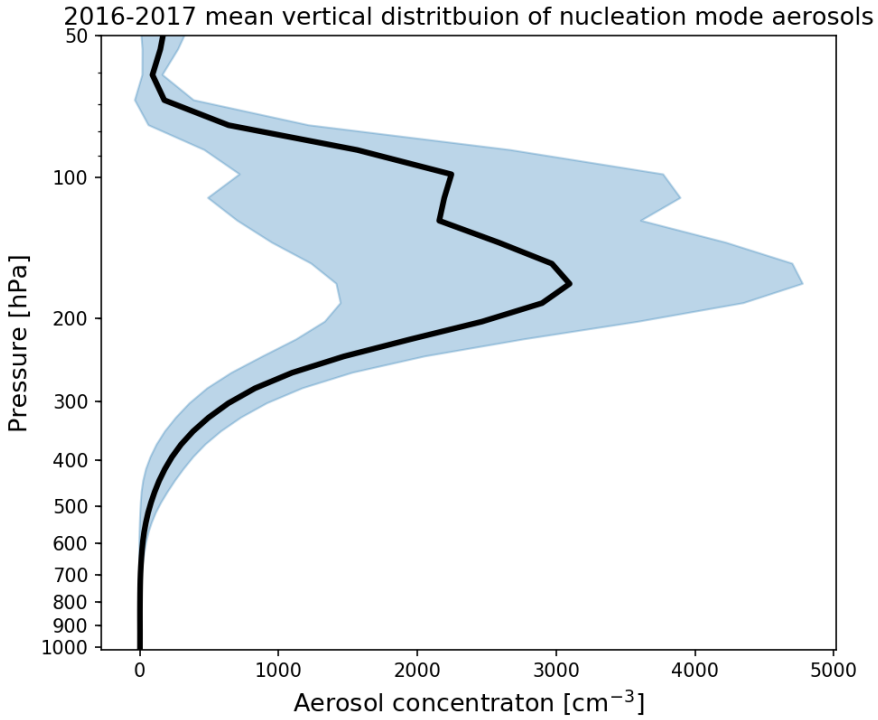


Figure 1. Mean vertical distribution of nucleation mode aerosols in the cloud box region for 2016-2017, simulated by UKESM1. The black line is the mean distribution, and the blue-shaped area represents the standard deviation.

2. Mention the time period you are looking into.

We revised the manuscript as follows.

P1L14-16

In this paper, we use the United Kingdom Earth System Model to investigate the sources of CCN (from emission and atmospheric processes) in the SEA, and the response of cloud droplet number

concentration (CDNC), cloud liquid water path (LWP), and radiative forcing to those sources during 2016 and 2017.

3. Lines 21-23 seem to include two contradictory phrases - perhaps the authors mean to say most of the BL CCN is introduced from above through entrainment? Wouldn't BB then be the dominant BL CCN source?

We have removed this sentence. The revised text reads as follows.

Overall, free and upper troposphere nucleated aerosols are the dominant source of boundary layer $CCN_{0.2\%}$, contributing an annual average of ~ 41 % as they subside and entrain into the marine boundary layer. In terms of emission sources, anthropogenic emissions (from energy, industry, agriculture, etc.) contribute the most to the annual average $CCN_{0.2\%}$ in the marine boundary layer (~ 26 %), followed by biomass burning (BB, ~ 17 %). In the cloud layer, BB contributes about 34 % of annual $CCN_{0.2\%}$, midway between the contributions from aerosol nucleation (36%) and anthropogenic sources (31%). The contribution of aerosols from different sources to CDNC is consistent with their contribution to $CCN_{0.2\%}$ within the marine boundary layer, with free and upper troposphere aerosol nucleation being the most important source of CDNC overall. In terms of emission sources, anthropogenic sources are also the largest contributors to the annual average of CDNC, closely followed by BB.

4. Line 25: the reader doesn't yet know the model simulation places most of the aerosol above the BL. Keep reviewer 3's comments in mind here and rewrite.

We have revised the sentence as follows.

P1L25-27

The contribution of BB to CDNC is more significant than its increase to $CCN_{0.2\%}$, mainly because BB aerosols are mostly located directly above the inversion layer in the model, thus can increase CDNC by enhancing the maximum supersaturation through the dynamical feedback due to shortwave absorption.

5. State something about the model aerosol vertical structure in the abstract - results are highly dependent on the aerosol being located above the cloud.

We added the BB aerosol vertical information in response to comment 4.

We have made the following revision to L31-32 to emphasise the vertical distribution of BB aerosols.

P2 L2-4

However, as most BB aerosols are located directly above the inversion layer, their effect on clouds increase due to its absorption effect (about the same as anthropogenic sources for CDNC and more than anthropogenic sources for LWP), highlighting the crucial role of its radiative effect on clouds.

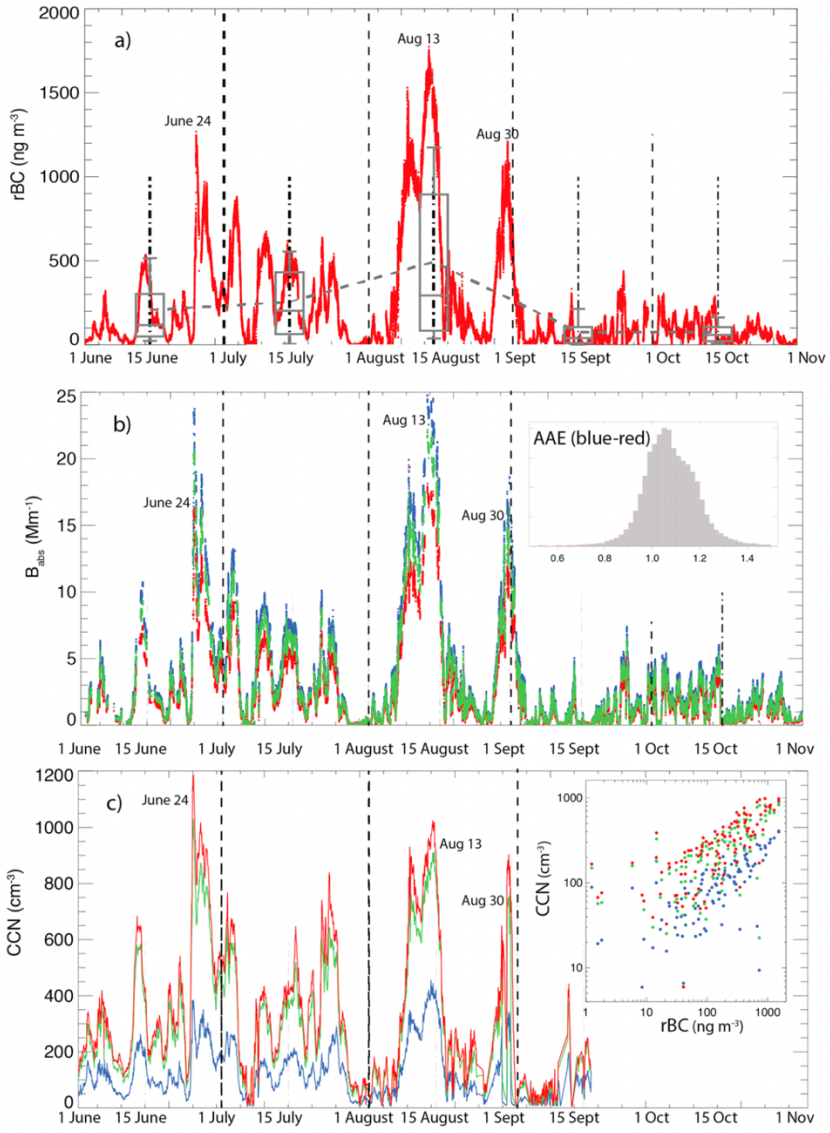
6. The reader might be reasonably surprised to read that non-BB anthro emissions are the largest contributors to MBL CCN_0.2%, above BB and sea spray. On p. 12 you clarify the CCN in the cloud layer - thus those most likely to form cloud droplets - are more likely to be BB. This would be worth mentioning in the abstract.

We added the following sentence in the abstract (L19-20)

In the cloud layer, BB contributes about 34 % of annual $CCN_{0.2\%}$, midway between the contributions from aerosol nucleation (36%) and anthropogenic sources (31%).

Overall:

The authors conclude, that because in the UKEMS1 model, the BBA is mainly in the FT for July-September, and has a poor hygroscopicity, that BBA is less important as a BL CCN, and instead, primarily serves to strengthen the inversion top. This is valuable to know about the UKESM1 model behavior. What is less clear is how well the UKESM1 simulations are capturing the observations. The reviewer comments relate to this: Reviewer 1 mentions the seasonal cycle. Rev 2 and 3 mention model characteristics.



For example: the LASIC campaign has shown that there can be significant CCN in the BL, for example, Zuidema et al 2018 Fig. 1, reproduced above, shows CCN_0.2% reaching 103 /cc. The temporal variation indicates it is primarily modulated by BC. These values I believe exceed those shown in Fig. R1, though Fig. R1 is difficult to interpret; absolute values for the CCN depicted would have helped, or at least an explanatory caption. The July-September model means shown in Fig. 2 are difficult to interpret for the BL, and a 3-month model mean doesn't communicate the range.

In addition, in several portions of the manuscript, the authors refer to Che 2021, as a model validation paper. That paper only compared aerosol extinctions along CLARIFY and ORACLES flight tracks, with the ORACLES flight tracks spending little time in the boundary layer. During September, the LASIC values also indicate a clean MBL, consistent with ORACLES-2016, but a BL lacking smoke in September does not mean the BL is also non-smoky in July and August. The comparison to the aircraft flight track data isn't a sufficient validation for the 3 months, in contrast

to the statement on p. 8, line14. Is the model genuinely capturing the boundary layer smoke in July and August? Can the authors create a figure from their model simulation that is comparable to the LASIC data? The authors also refer to several other papers as a form of validation (e.g., p. 2, line28): Deaconu 2019 and Wilcox 2010 rely on satellite datasets that have difficulty distinguishing BB within the BL from sea-spray, Gordon 2018 is a modeling study focusing on a 10-day August time period only that produced an unrealistic 8K warming in the free troposphere, and Ackerman 2004 is for a different location. These references ignore the new information we have thanks to LASIC, ORACLES, and CLARIFY. Besides the studies mentioned by Rev 1, there are also more detailed StCU-to-Cu transition papers indicating the BB can also have a radiative impact in the BL.

Similarly, Kacarab et al. 2020 is also relevant, indicating a kappa of 0.4 for smoke based on oracles observations. This study should be referenced and discussed somewhere, as it does not support the low hygroscopicity for smoke assumed here. https://acp.copernicus.org/articles/20/3029/2020/.

We thank the editor for the comment. The editor's main concern is whether our conclusion that, on the annual average (2016-2017), BB are not the dominant source of $CCN_{0.2\%}$ within the SEA boundary layer is correct? Whether the model significantly underestimates $CCN_{0.2\%}$ from BB within the boundary layer?

To answer the editor's questions. We compared $CCN_{0.2\%}$ simulated by the model (baseline run) to that measured from the LASIC campaign.

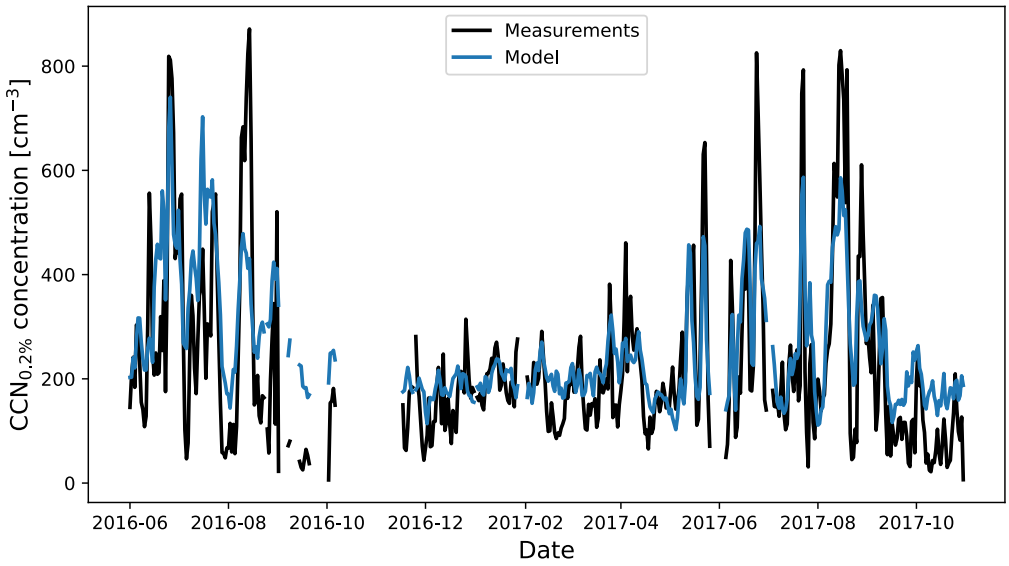


Figure 2. Comparison of modelled and observed daily mean $CCN_{0.2\%}$ concentrations. The measured $CCN_{0.2\%}$ is from the LASIC campaign. The modelled $CCN_{0.2\%}$ is from the baseline simulation and interpolated to LASIC coordinates.

The modelled $CCN_{0.2\%}$ concentrations are collocated with observations from LASIC at 340 m on Ascension Island, representing the marine boundary layer CCN in the SEA. Due to the temporal

resolution of the model output, we compared the daily average values. From the figure, the modelled $CCN_{0.2\%}$ is in good agreement with the observation, especially can capture the daily variation of $CCN_{0.2\%}$ during the BB season. The campaign averaged $CCN_{0.2\%}$ is 225 cm^{-3}, and the modelled corresponding mean is 239 cm^{-3}, with the mean relative error of the modelled $CCN_{0.2\%}$ 6.3%. However, the observed CCN peaks during the BB season are higher than simulations admittedly, indicating that the model is still inadequate for capturing those peak values. Given that we mainly investigate the annual mean CCN in this paper, the small error and the well-matched temporal variability with observation suggest that the model is reasonably in reproducing the CCN in the marine boundary layer in SEA.

We have now included this evaluation in the manuscript. There are several revisions in the manuscript, as shown below.

In the introduction (P4L13-16):

In addition, a ground-based in-situ field measurement campaign (LASIC, Layered Atlantic Smoke Interactions with Clouds) was carried out on Ascension Island, which provided 18 months of observations for aerosols and clouds within the marine boundary layer from June 2016 to October 2017 (Zuidema et al., 2018b).

In the method section (P9-P10)

2.3 Model evaluation

The model has been evaluated with the ORACLES (2016, 2017) and CLARIFY measurements by examining the collocated aerosol extinction in our previous paper. The result shows the model can generally capture the spatial and vertical distributions of BB plume (Che et al., 2021). However, as these aircraft observations are mainly located in the free troposphere, we further evaluated modelled CCN within the marine boundary layer using LASIC observations.

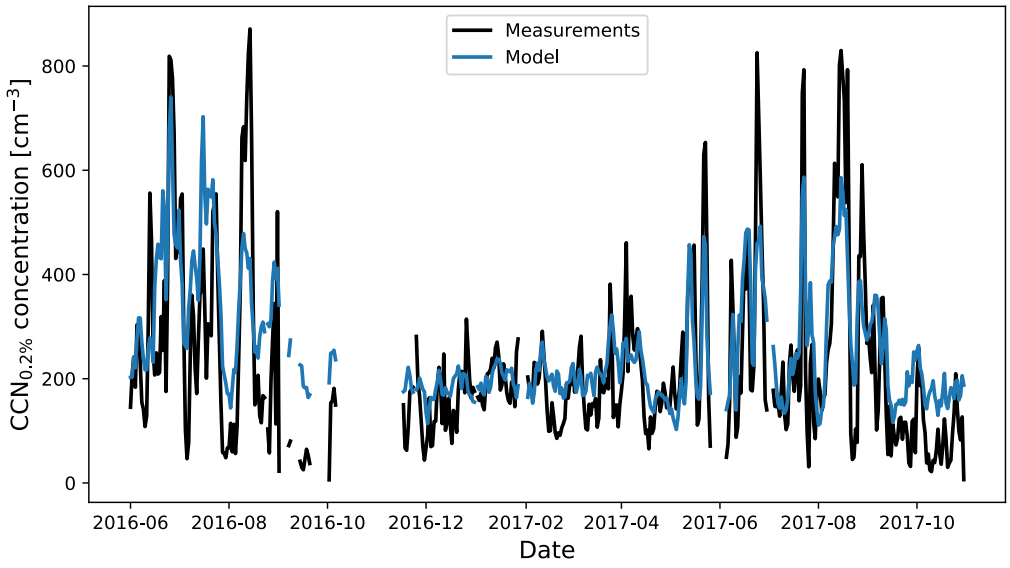


Figure 3. Comparison of modelled and observed daily mean $CCN_{0.2\%}$ (CCN at 0.2 % supersaturation) concentrations. The measured $CCN_{0.2\%}$ is from the LASIC campaign. The modelled $CCN_{0.2\%}$ is from the baseline simulation and interpolated to the LASIC coordinates.

The LASIC campaign was carried out on the Atmospheric Radiation Measurement (ARM) Mobile Facility 1 site at Ascension Island, located at a latitude of -7.97°, longitude of-14.35° and altitude of 340.7664 m. The LASIC CCN was measured by a cloud condensation nuclei counter (CCNC-200), which provides the CCN concentration at fixed supersaturations (Roberts and Nenes, 2005; Atmospheric Radiation Measurement (ARM) user facility, 2016). A more detailed description of the sampling location and instruments can be found in the campaign report (Zuidema et al., 2018a). The modelled CCN concentration at 0.2% supersaturation ($CCN_{0.2\%}$) from the baseline simulation is collocated with observations. Due to the temporal resolution of the model output, we compared the daily averages as illustrated in Fig. 3.

As evident in Fig. 3, the modelled $CCN_{0.2\%}$ is in good agreement with the observation, and can capture the daily variation of $CCN_{0.2\%}$ during the BB season. The campaign averaged $CCN_{0.2\%}$ is 225 cm^{-3}, and the modelled corresponding means of 239 cm^{-3}, with the mean relative error of the modelled $CCN_{0.2\%}$ around 6.3%. However, the observed CCN peaks during the BB season are higher than simulations, indicating that the model is still inadequate for capturing those peak values. One possible reason is that when BC particles have a thick coating, the calculated overall κ may be underestimated by the volume mixing rule, which may further underestimate the CCN concentration associated with BB (Kacarab et al., 2020). In addition, uncertainties in the BB emissions, including the magnitude, size and, height of fires, can lead to incorrect estimates of BB aerosol peak concentrations, which can lead to such underestimations of CCN. Given that we mainly investigate the annual mean CCN in this study, the relatively small error and the well-matched temporal variability with observation suggest that the model is fairly reasonably in reproducing the CCN in the marine boundary layer in the SEA. Therefore, this result provides confidence in this study.

In Discussion and conclusion (P20 L3-5)

The model has been evaluated with aircraft measurements from CLARIFY and ORACLES for the aerosol distribution, and is further evaluated in this study with LASIC in-situ observations for the marine boundary layer CCN.

P21 L20-24

By comparing the modelled $CCN_{0.2\%}$ with observations, we find that although the model is generally in good agreement with the measurements, it still underestimates the peak $CCN_{0.2\%}$ during the BB season, suggesting that BB associated $CCN_{0.2\%}$ may be underestimated during the BB season. Also, when BC particles have a thick coating, the calculated κ may be underestimated by the volume mixing rule, which may further underestimate CCN concentrations associated with BB.

The modelled annual mean $CCN_{0.2\%}$ in the SEA marine boundary layer during 2016-2017 is 290 cm^{-3}, while the mean $CCN_{0.2\%}$ during BB season (July-September) is 331 cm^{-3}, 14 % higher than

the annual mean, confirming the influence of BB on $CCN_{0.2\%}$. This is also consistent with the LASIC observation data. For the annual average, BB contributes 17% of the CCN within the marine boundary layer in the SEA, which is lower than anthropogenic sources (26%). Therefore, in terms of emission sources, anthropogenic sources are considered to be the largest source of CCN in this area. However, during the BB season, BB contributed 19% of $CCN_{0.2\%}$, slightly less than anthropogenic sources (21%), indicating the equally important roles of BB and anthropogenic sources to $CCN_{0.2\%}$ during the BB season in this region. Figure 5 in the manuscript and figure S4 in the supplement show the contribution of different sources to the annual mean and BB seasonal mean $CCN_{0.2\%}$, respectively, with the absolute concentrations and relative fractions of $CCN_{0.2\%}$ from each source labelled in the figure. We therefore added the total concentration of annual and BB seasonal averaged $CCN_{0.2\%}$ in their figure caption, respectively.

P12L0-11 (manuscript):

Using the simulation of the present day as the baseline (annual mean $CCN_{0.2\%}$ around 290 cm^{-3}), the contribution of each source to $CCN_{0.2\%}$ is marked at the top of the corresponding bar in percentage.

P5L6-8 (supplement)

Using the simulation of the present day as the baseline (BB seasonal mean $CCN_{0.2\%}$ around 331 cm^{-3}), the contribution of each source to $CCN_{0.2\%}$ is marked at the top of the corresponding bar in percentage.

We also updated the reference as suggested by the editor. The introduction is revised as follows.

P2 L28- P3 L3

Previous studies suggest that as the BB aerosols are mainly located above and near the inversion layer, when above the inversion layer, the main role of their radiative effect in the SEA is to strengthen the capping inversion and reduce dry air entrainment from cloud tops, thereby increasing the LWP and low-level cloud fraction, resulting in a significant impact on the radiation balance (Wilcox, 2010; Gordon et al., 2018; Deaconu et al., 2019; Mallet et al., 2020; Herbert et al., 2020; Chaboureau et al., 2022). When BB aerosols are located in the marine boundary layer, their radiative effect can enhance the decoupled boundary layer and result in a reduction in cloud cover and LWP, shifting the stratocumulus-to-cumulus transition to the upwind area (Zhang and Zuidema, 2019; Ajoku et al., 2021).

Similarly, Kacarab et al. 2020 is also relevant, indicating a kappa of 0.4 for smoke based on oracles observations. This study should be referenced and discussed somewhere, as it does not support the low hygroscopicity for smoke assumed here.

In the UKESM1 model, κ is calculated with the simple volume mixing rule (Petters and Kreidenweis, 2007), which assumes for certain soluble mode aerosols, all particles are homogeneously mixed, so that the overall κ is determined based on the volume fraction of the different species and the hygroscopicity of each component. However, since κ is set to 0 for BC,

this volumetric mixing rule may underestimate the overall κ of when the BC has a thicker coating. This was illustrated by Kacarab et al. (2020), who found a high overall κ of about 0.4 from eight ORACLES 2017 aircraft observations. However, Zhang et al. (2022) found the overall κ around 0.24 in the marine boundary layer from ORACLES 2018 observations, which may be consistent with our assumption that BB reduces the overall κ.

Therefore, we revised the manuscript as follows.

P5 L13-18 (Method)

The internal volume mixing rule (Petters and Kreidenweis, 2007) is used to calculate the mean hygroscopicity of each mode. Therefore, a higher fraction of less hygroscopic components (e.g. organic and black carbon) can reduce the overall *κ*. However, the overall κ may be underestimated when BC has a thicker coating. This was illustrated by Kacarab et al. (2020), who found a high averaged κ of ~ 0.4 from eight ORACLES 2017 aircraft observations. However, Zhang et al. (2022) found an averaged κ of ~ 0.24 in the marine boundary layer from ORACLES 2018 observations, which is consistent with our assumption that BB reduces the overall κ.

P10L1-4 (Method)

However, the observed CCN peaks during the BB season are higher than simulations, indicating that the model is still inadequate for capturing those peak values. One possible reason is that when BC particles have a thick coating, the calculated overall κ may be underestimated by the volume mixing rule, which may further underestimate the CCN concentration associated with BB (Kacarab et al., 2020).

P21L23-24 (Discussion and conclusion)

Also, when BC particles have a thick coating, the calculated κ may be underestimated by the volume mixing rule, which may further underestimate CCN concentrations associated with BB

The manuscript should also be read over again by a native English speaker, to clarify some of the language. I mention a few specific comments below:

Thanks for the comment. We have carefully proofread the manuscript and have extensively revised it. All revisions are highlighted in the manuscript.

Line 12 p 3: of ->from

Thanks to the editor, we have corrected this.

Line 13 p 3: to the -> to that of Line 16: after -> by

Thanks to the editor, we have corrected these.

p. 3 line 20-21: Kalahari dust doesn't advect far according to https://acp.copernicus.org/articles/21/8169/2021/acp-21-8169-2021.pdf, and it certainly wasn't one of the most observed aerosol at Ascension Island during the LASIC/CLARIFY campaigns. The Begue result for the Netherlands isn't relevant here. The first author could draw on their own work assessing kappa using LASIC measurements. Overall this paragraph and its emphasis on dust lacks support and is misleading.

Thanks for the comment. We have deleted those descriptions.

p. 5: are the BC aerosols internally mixed? Results don't acknowledge that a kappa of 0 for BC doesn't reflect that all of it is likely internally mixed (e.g., Dang et al., 2021), with the BC particle size helping cloud nucleation. How well does the Petters and Kreidenweis internal mixing rule work for this region based on what we know so far from the observations?

Yes. BC aerosols are internally mixed in soluble modes in GLOMAP. This assumes all particles are homogeneously mixed, and their overall κ is based on the volume fraction of different species. However, as the κ of BC is set to 0, this volume mixing calculation can underestimate the overall κ of the aerosol when the BC has a thicker coating, which may further underestimate the CCN concentration. Therefore, we acknowledged the CCN from biomass burning might be underestimated by the model in the discussion, as follows.

P5 L14-19 (Method)

The internal volume mixing rule (Petters and Kreidenweis, 2007) is used to calculate the mean hygroscopicity of each mode. Therefore, a higher fraction of less hygroscopic components (e.g. organic and black carbon) can reduce the overall κ. However, the overall κ may be underestimated when BC has a thicker coating. This was illustrated by Kacarab et al. (2020), who found a high averaged κ of ~ 0.4 from eight ORACLES 2017 aircraft observations. However, Zhang et al. (2022) found an averaged κ of ~ 0.24 in the marine boundary layer from ORACLES 2018 observations, which is consistent with our assumption that BB reduces the overall κ.

P10L1-4 (Method)

However, the observed CCN peaks during the BB season are higher than simulations, indicating that the model is still inadequate for capturing those peak values. One possible reason is that when BC particles have a thick coating, the calculated overall κ may be underestimated by the volume mixing rule, which may further underestimate the CCN concentration associated with BB (Kacarab et al., 2020).

P21L23-24 (Discussion and conclusion)

Also, when BC particles have a thick coating, the calculated κ may be underestimated by the volume mixing rule, which may further underestimate CCN concentrations associated with BB.

P. 5: clarify that anthropogenic does not include BB. You could consider calling it 'non-BB anthropogenic'.

We clarified this in the manuscript as follows.

P5L30-P6L2

Note that although black carbon (BC) and organic carbon (OC) are the main components of BB emissions, these two types of aerosols are also present in anthropogenic emissions. However, the 'anthropogenic' emissions defined here do not include BB aerosols, although BB in southern Africa is associated with human activities (Roberts et al., 2009). In our model setup, BC and OC from our 'anthropogenic' emissions are mainly from fossil fuels and biofuels, and their emission sectors are energy, industrial, shipping, transportation, solvents, waste, agriculture, and residential. In comparison, BC and OC from BB are mainly emitted from the burning of agricultural land, peat, savanna, forest, and deforestation.

Language on nucleation confusing - authors use the same term for gas to particle production of aerosols, and for cloud activation. Here it might be worth adding additional detail to the naming, meaning, to use the longer term of 'aerosol nucleation'. Mention how boundary nucleation differs from total nucleation in the boundary layer for caption of fig. 3. Also, the comment from Rev 2 that the Hadley center models lend more emphasis on nucleation driven CDNC than other models should be mentioned, including the citation to Bellouin et al 2013.

Thanks for the suggestion. We changed the term to 'aerosol nucleation' in the manuscript. The difference between boundary layer nucleation and total nucleation is briefly described in the caption of Fig. 4. (We added a new figure before, thus Fig. 3 in the last version becomes Fig. 4)

P10 L12 – P11 L3

Figure 4. UKESM1 simulated annual mean vertical profiles of CCN concentration at 0.2% supersaturation ($CCN_{0.2\%}$) from different sources (at the standard temperature and pressure STP). Profiles are averaged along the latitudes of the cloud box. The contributions of different sources to $CCN_{0.2\%}$ are listed in (a) to (h), where the contribution of emissions is shown in the yellow frame, and the contribution of atmospheric processes is shown in the light blue frame. Note boundary layer aerosol nucleation is based on organic-mediated aerosol nucleation and is limited to the boundary layer. Total aerosol nucleation includes boundary layer nucleation and homogeneous binary aerosol nucleation in the free troposphere and stratospheric. The contour lines in each subplot are the cloud specific water content from the baseline simulation at the same temporal and spatial average. The same colourmap scale is used in each subplot to facilitate comparison, but the range differs for each plot, corresponding to the maximum and minimum of $CCN_{0.2\%}$.

We also revised the manuscript to address the comment by reviwer#2 about the nucleation.

P16 L4-9

Previous studies have found that more than half of the CCN in the global marine boundary layer is contributed by aerosol nucleation (Clarke et al., 2013; Merikanto et al., 2009b; Williamson et

al., 2019; Clarke and Kapustin, 2002), consistent with our result. However, source attribution in multiple models is recommended to confirm the importance of aerosol nucleation to the CDNC, as the nucleation and Aitken mode aerosol concentrations are significantly overpredicted by HadGEM models (Ranjithkumar et al., 2021; Gordon et al., 2020; Hardacre et al., 2021; Bellouin et al., 2013), suggesting the nucleation contributed CDNC may also be overestimated in our model.

p. 7 line 11: Kacarab et al. 2019 is not consistent with the low model hygroscopicity. p. 8: Note Redemann 2021 shows satellite-derived Nd that are clearly elevated

We have revised the manuscript as follows.

P7L14-19

BB aerosols contributes around 76 % of total AOD in the cloud box during BB season, and can result in a clearly elevated CDNC in the SEA from satellite observations (Redemann et al., 2021), implying the potentially dominant role of BB aerosol in affecting CCN and cloud that motivated the ORACLES, CLARIFY and LASIC campaigns. However, as most of the BB aerosol is above the stratocumulus cloud deck (Fig. 2), combined with a large fraction of low hygroscopic particles such as BC and OC, the fraction of BB aerosol to activate as cloud droplets is uncertain.

We also revised the manuscript about CDNC source attribution as follows.

P16 L20-24

BB aerosols not only can provide CCN to increase CDNC, but also increase CDNC by influencing the vertical distribution of temperature through shortwave absorption, which in turn increases the maximum supersaturation in clouds (Che et al., 2021). This is also evidenced by Fig. 6. As a result, BB becomes the most important emission source of CDNC during the BB season. This result is also supported by a satellite study that found an elevated CDNC with the presence of BB aerosols in this region (Redemann et al., 2021).

p. 16, line 7: to say that 'nucleation is important to CDNC' is very unclear - if you're talking about cloud nucleation, it's overstating the obvious. You don't mean cloud nucleation I recognize, but this is simply not clear writing. Explain what nucleation means in these sentences.

We have revised this as the following.

P16 L9-11

Even during the BB season, the concentration of CDNC contributed by total aerosol nucleation is similar to that of BB (Fig. S5), indicating that total aerosol nucleation remains the most significant source of CDNC throughout the years.

More detail on the non-BB anthro emissions would also be useful. I recognize these are difficult to validate - do the sulfate contributions from the non-BB anthro+sea spray match what was measured at Ascension Island?

We didn't output aerosol chemical compositions from the model, so we cannot directly compare the modelled sulphate with the LASIC campaign.

To validate whether the anthropogenic sources in this region are overestimated, we compared the AOD from anthropogenic sources with the AOD level 1 data observed from AERONET on Ascension Island. This is shown in figure 3.

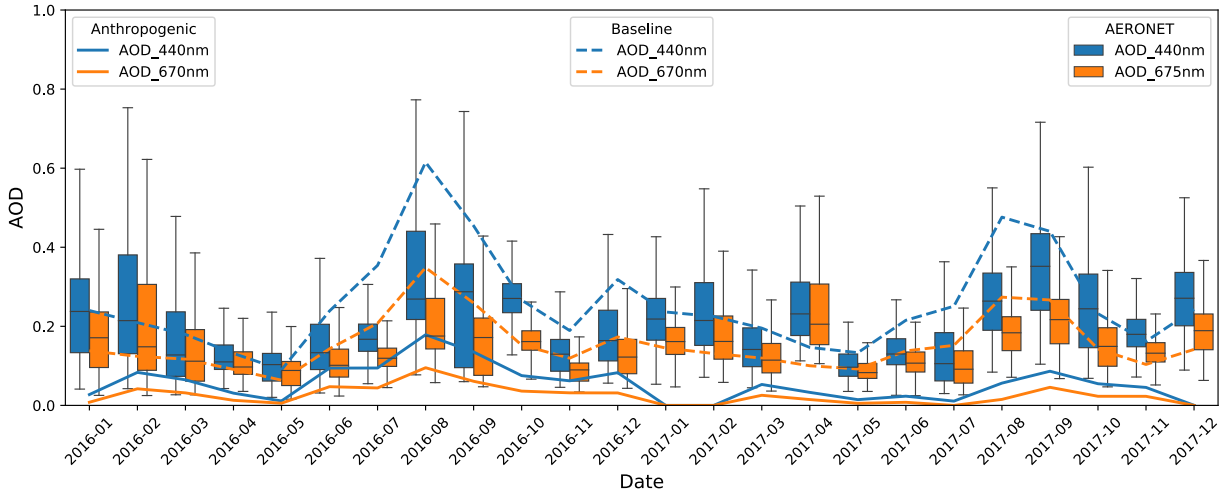


Figure 3. Monthly mean modelled AOD and box-whisker plots of monthly AOD percentiles (10%, 25%, 50%, 75%, and 90%) measured from AERONET (level 1 data) on Ascension Island. The dashed lines are the AOD from the baseline simulation, and the solid lines are the AOD from the anthropogenic source. Blue and orange colours indicate the AOD at 440 and 670 nm, respectively.

Due to the time setting of model inputs, we compared the monthly average AOD. Note that modelled AOD are averaged over the whole month, while the observations are not available for the whole time during each month, which will result in some uncertainties in the comparison.

The baseline simulated AOD correlates well with the AERONET and has a consistent trend with observations, both being higher in the winter months (also the BB season) for the Southern Hemisphere. In contrast, the modelled values are higher than the AERONET observations during biomass burning season, which may be due to the high proportion of clouds resulting in missing values during these months. Except for January and February of 2017, AOD from anthropogenic sources had a consistent trend with observations. Comparing the values, it can be found that the AOD contributed by anthropogenic sources are smaller than those measured from AERONET, and this difference becomes larger during BB season. Therefore, anthropogenic emissions are likely not overestimated in the model, suggesting the finding in the manuscript that anthropogenic emissions are the largest source of $CCN_{0.2\%}$ in the SEA marine boundary layer is not a result of the model's overestimation of anthropogenic sources. However, as the editor suggests, a comparison of aerosol chemical composition would be more appropriate. However, this is beyond the scope of this work and we hope to discuss this issue in the future.

A bit more effort could be made in the last section mentioning how the new observations can be used to assess and/or improve the model, including using the enhanced resolution of the seasonal cycle.

Thanks for the comment. We revised the manuscript as follows.

The discussion of different sources of CCN and their effects on clouds and radiation in this work is based on the averages during the BB season. However, from July to September, BB aerosol emissions vary with the burning conditions and areas, the marine boundary layer also evolves as the sea surface temperature decrease, and the stratocumulus cloud fraction also varies in different months. Therefore, the impacts of aerosol sources on CCN, clouds and radiation can be different for each month during the BB season, and require future studies. In addition, the influence of aerosols at different heights (boundary layer, cloud layer, free troposphere) on clouds and radiation is also an interesting issue that needs future investigation. The LASIC observational campaign can provide valuable continuous measurement data during the BB season in 2016 and 2017, which can be used to validate the model's performance in the SEA marine boundary layer at a higher output resolution. ORACLES and CLARIFY aircraft observations can provide cloud and aerosol measurements at different altitudes, contributing to future studies of the effects of aerosols at different heights on clouds and radiation. The long-term LASIC observations also can provide sufficient data for the study of seasonal variation, benefiting future studies.

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