Response to reviews and comments on "*An aerosol activation metamodel of v1.2.0 of the pyrcel cloud parcel model: Development and offline assessment for use in an aerosol-climate model*"

We would like to think the reviewers for their thoughtful, thorough, and helpful suggestions and feedback on our manuscript. We appreciate the reviewers' recognition of the potential for emulator approaches, such as the one we discuss in our manuscript, to improve parameterizations of aerosol-cloud interactions in global models, and have made efforts (as documented below) to address the additional questions and issues they identified pertaining to the manuscript.

Below, we respond to each of the "General" and "Specific" comments from the three reviewers, and recorded the actions taken in response to the "Minor" comments provided. We first respond generally to three additional, common "General" comments raised by all three reviewers here:

1. All three reviewers requested additional details on the computational expense of the metamodels, and more information on how they could be used in global models. Up to and during the review of this manuscript, we completed the implementation of a set of our activation metamodels into a global climate mode. Although we defer the discussion of the full impacts of these schemes on the simulated climate to a future manuscript currently *in preparation*, we are able to include a discussion of the impact the emulators have on the computational expense of the global model, compared with the other activation schemes we consider in this manuscript. This discussion has been added to the concluding section of the manuscript. In short, 3 of the 4 activation models yield negligible impact on the performance of MARC versus the reference ARG implementation. The MBN scheme increases the computational expense by about 7%; the 4th-order gCCN scheme incurs an increase of just 3%.

2. All three reviewers expressed concern that the manuscript was much too long, did not explicitly clarify its differences with Rothenberg and Wang (2016), and did not focus narrowly enough on the develop and evaluation of the metamodels. To address these points, we have re-written the Introduction nearly in its entirety to exclude historical details of activation parameterization development better recounted elsewhere (e.g. in Ghan et al. (2011)), and consolidated the discussion in Sections 2 and 3 into a single section entitled "Emulation of aerosol activation". Furthermore, we have worked to eliminate the reproduction of material available in Rothenberg and Wang (2016), opting to refer the reader there for details on the parcel model used here and on the polynomial chaos expansion technique.

3. All three reviewers included comments on redundancies in Section 2 and the iterative activation calculations. As mentioned previously, we have consolidated this Sections 2 and 3 and streamlined the discussion in both. Where possible, we have tried to include additional references to justify our decisions on how to exclude particular aerosol modes. Overall, we still feel it is important to include this discussion in the manuscript; a key difficulty in employing emulators and other statistical approaches to study physical processes and to develop parameterizations is the "curse of dimensionality," where increasing the number of important parameters tends to greatly increase the expense of building and evaluating a given emulator or statistical model. A critical component of such an effort, then, is rigorously searching for reduced sets of parameters to use in building the statistical models, often referred to as "feature extraction" in the machine learning community or otherwise "dimensionality reduction". Although there is a large literature on algorithms and automatic ways to accomplish this, these techniques often use a brute-force or exhaustive search approach. Our iterative calcu-

lations are the result of attempting to build a physically-informed algorithm for approaching feature extraction, and we hope that such an approach will be informative for researchers who pursue similar lines of study in the future. Therefore, we have tried to retain at least a limited discussion of the iterative calculations and results in the revised manuscript.

The remaining reviewers' comments are addressed below. We have made our best effort to address each individual comment in the sections labeled by the reviewers (General/Specific/Major/Minor/Technical comments, where denoted), and in the same order/numbering as they were presented. For clarity, we have included a summary of the reviewer's comment **in bold** before each response. Although this resulting document is somewhat long, we hope it fully documents our attempts to incorporate all of the thoughtful critiques provided by the reviewers and outlines our planned revisions to the manuscript.

Reviewer #1

General Comments

- **Lengthy historical introduction**

  We have opted to extensively re-write the introduction to streamline the introduction, referring the reader to other literature for more information. The new introduction is focused on the critical literature most pertinent to the development of our new method and parameterization.

- **Failure to focus on description of emulator development and related work**

  We have included discussion of the noted literature (Partridge et al. (2011) and Carslaw et al. (2013)) and other work on emulation as applied to atmospheric and climate processes, and made an effort to place this new work in that appropriate context. We have also worked to reduce the duplication of details already available in Rothenberg and Wang (2016).

- **GCCN**

  Although the focus of this manuscript is not on analyzing the impacts of GCCN on the accuracy of activation calculations in general, we did wish to understand how their inclusion/exclusion could influence the relative performance of our metamodels. We agree that this was not sufficiently introduced in the manuscript, and have added a brief discussion on how the presence of GCCN impacts aerosol activation in the introduction. Additionally, we added some clarifying remarks where the separate "main" and "GCCN" schemes are introduced in the manuscript to better highlight our motivation for training two separate emulators.

- **Path forward/computational cost**

  We defer the reviewer to our comments at the beginning of this document. We also include brief remarks in the conclusion of the manuscript discussing our implementation in the global model and its implications for future work.

- **Testing of activation schemes**

  We agree with the reviewer that, in fact, activation schemes have been tested against complex aerosol distributions from a variety of tools, including GCMs and CTMs. However, we stress that our remarks about testing the schemes refers more-so to inter-comparisons of different

activation parameterizations. Since the comprehensive review of Ghan et al. (2011), few works have looked at how different activation schemes perform when coupled to the same host model, or using the same set of complex aerosol size distributions. In order to more accurately represent the historical work on this issue, we've amended some of our comments on this topic in the manuscript, especially in the Introduction (e.g. the first *Specific Comment* below).

Specific Comments

The following responses match exactly to each of the comments provided by the reviewer, in the order provided. We have included reference line numbers (used in the review) to help keep track of which comment is which, where possible.

- We've rephrased **P3/L33** following the final *General Comment* above and the reviewer's comment
- Section 2 has been entirely re-written, and consolidated with Section 3.
- **P6/L19**: Removed
- **Figure 2**: We've revised our presentation of the aerosol size distribution parameters, and have replaced the original Figure 2 with a plot featuring PDFs of those parameters. This complements Table 2 and our discussion of the emulator approach later in the manuscript.
- **Section 2.2**: Replaced and consolidated with Section 3
- **Section 2.3**: Replaced and consolidated with Section 3. In the revised manuscript we have greatly shortened the discussion of the iterative procedure.
- **P7/L31**: Re-phrased and worked differently as part of the shortening mentioned previously.
- **P8/L13**: Removed, similarly to the previous comment.
- **P8/L10-30**: Removed, similarly to the previous comment. Reviewer #2 raised a similar concern about our discussion of the mechanism driving supersaturation production. We agree with both reviewers that our explanation is incomplete, and derives from a too-literal interpretation of our Equation (1). We've re-written the discussion (which occurs several times, as indicated in the next two comments) to emphasize the role of water vapor availability/reduction in controlling the maximum supersaturation.
- **P9/L10**: See previous comment
- **P10/L7,15**: See previous comment
- **P12/L15**: Re-worked the discussion on the limitations of our methodology in the consolidated Sections 2 / 3.
- **Figures 5-8**: We note that the discussion which includes Figure 5-8 still draws samples from the set of uniform distributions described in Table 2, which were constructed using the observed ranges of aerosol size distribution parameters sampled from MARC. However, the first sampling study presented in the manuscript assumes these distributions to be independent, which is where the potential for non-physical combinations arises. We've tried to re-balance the discussions of our results more towards the aerosol size distributions sampled from MARC.

Reviewer #2

Major Comments

1. **Structure and Length of the manuscript**

We have embraced all three reviewers' comments about the structure and length of manuscript and have undertaken substantial revisions and re-writing to address them. Specifically:

a) The introduction has been nearly entirely re-written. We've removed references to studies of processes irrelevant to the results we present in this manuscript.

b) We agree that the discussion of the small role of nucleation mode aerosol in activation dynamics is redundant. Section 2 has been consolidated and merged with Section 3. However, we have tried to retain some discussion and analysis of the iterative calculations, since we believe this procedure is more widely useful. Please refer to the comments at the beginning of this document for more information.

c) Similar to the previous comment, we emphasize that our attempt to consolidate Sections 2 and 3 addresses this issue pertaining to reporting results which are not new.

2. **Comparison to other activation schemes**

We agree with the reviewer that there is an inherent "unfairness" in evaluating the ARG and MBN schemes against our own parcel model and our sets of input parameters, and as the reviewer notes, we allude to this in the Conclusion. In the revised manuscript we bring up this point of discussion earlier. However, we do wish to emphasize that regardless of how a given scheme has been developed, to predict online CDNC in a global model, it must be coupled to some representation of an aerosol size distribution which may fall far outside the parcel model evaluations used to initially fit the scheme. As a result, it is critical to document the performance of these schemes in these extreme scenarios, which can and do arise in numerical models. We have attempted to clarify this discussion in the manuscript.

3. **Features of the aerosol size distribution in MARC**

Table 2 provides some perspective on the distribution of MOS hygroscopicity - in the model, it is restricted to vary between 10^{-10} and 0.507 (volume-weighted mixtures of OC and SO4). In practice, it takes a mean value of 0.27 ± 0.04 (1 standard deviation). We've clarified in the manuscript that the relative abundance of OC and SO4 dominates the small variation in κ_{MOS}. In MAM3, primary organic carbon is assumed to have a κ of 0.1, which would tend to increase this average value, although we note that the presence of SOA, BC, and dust - all of which have smaller κ than sulfate - would tend to reduce the average value away from that of pure sulfate. Additional analysis using a variance-based decomposition and the derived metamodels (following Rothenberg and Wang (2016)) suggests that the hygroscopicity of MOS plays a very small role in influencing droplet nucleation given our particular aerosol size distribution, and instead the size and abundance of sulfate is more important.

4. **Previous work (Rothenberg and Wang, 2016)**

We agree with the reviewer that deferring the reader to our previous study helps to streamline the present one, and have undertaken to do so while revising the manuscript. The new content here - which we emphasize in the revised manuscript - is the application to a more complex aerosol size distribution, and the training of a metamodel for use in predicting online cloud droplet number concentration in a global model which uses that aerosol size distribution.

5. **Role of gCCNs**

We've expanded our discussion of Figure 10 to put this result in the context of previous work on role of gCCN, especially in clean environments.

6. **Applicability of the emulator**

   We wish to emphasize that the emulators were in fact trained to be able to cover extremely clean conditions (see Table 2, which indicates the ranges of size distribution parameters included in the training). Figures 5 and 7 further illustrate that the emulators perform at least as well as the ARG and MBN schemes in extremely clean conditions, too. We have focused the discussion in the revised manuscript more towards the activation of aerosol size distributions sampled from MARC directly to clarify these points. We have also included a discussion of the computational performance of the schemes.

Minor Comments

1. **P1/L3-4**: We agree on this detail, and have added this caveat.
2. **P2/L17** and **P18/L35**: Changed "processes" to "effects"
3. **P6/L18**: The minimum is set to 0.2 m/s and the maximum to 10 m/s; we've tried to make this clearer.
4. **P8/L30**: This point was also raised by Reviewer #1. We've re-worded this discussion to de-emphasize the role of latent heat release and instead focus on water vapor reduction, which is a more important influence on supersaturation development.
5. **P10/L18**: Re-phrased as just "cloud droplets"
6. **P10/L19,27**: We've adopted the the phrasing recommended by the reviewer.
7. **P12/L25,26**: We've re-written this to express our intended point, which was that in Rothenberg and Wang (2016), predicting $S_{\max}$ and then diagnosing N_{act} tended to be more accurate than predicting N_{act} directly as an emulated response.
8. **P17/L29**: Rephrased with more specifics on how activation is part of a fundamental relationship between aerosol and CDNC, and how this influences the indirect effect.
9. **P18/L27**: Clarified that Gantt et al. (2014) used different activation schemes to simulate cloud radiative effect and in the global average, these effects differed by 0.9 W/m^{-2} depending on the scheme used.
10. **P19/L1**: Rephrased sentence to avoid this vagueness.
11. **P19/L5**: We agree that this point should be made much earlier in the manuscript, and have included it towards the end of our revised Introduction as a motivation for further development of activation schemes.
12. **Figure 1**: Added a gap on the y-axis as recommended, and revised figure aesthetics.
13. **Figure 3**: We've adopted this recommendation and replaced the figure with a simple description in our revised Sections 2/3.
14. **Figure 4**: Labels are now defined in the caption.
15. **Figure 5**: Removed "one-one plot comparing" from caption.

Technical Comments

1. **P2/L2**: Included this grammatical change.
2. **P2/L18**: Corrected
3. **P2/L19**: Sentence does not appear in revised Introduction.
4. **P3/L10**: Fixed in BibTeX file with references.
5. **P5/L6**: Corrected
6. **P5/L17**: Corrected

7. **P6/L33**: Added definition
8. **P10/L10**: Equations are removed in revised Sections 2/3 in lieu of a reference to Rothenberg and Wang (2016) and standard texts
9. **P12/L21-22**: Corrected. "Activate" was missing
10. **P15/L34**: Corrected
11. **P17/L23**: Rephrased to avoid awkwardness
12. **References**: We use the provided BibTeX bibliography format and will consult with the editor on an appropriate course of action; Updated the Morales Betancourt and Nenes (2014) reference.

Reviewer #3

General Comments

1. **Computational Expense**

   We've included an assessment of the computational cost of running the CESM/MARC with the different activation schemes compared in this work, and our new emulators.

2. **New developments in this study**

   In line with the previous general comment, we indicate in the manuscript that the emulators derived here are implemented in the CESM/MARC to calculate online CDNC. We further clarify that the work necessary to accomplish this implementation (documented in the manuscript) is major advance beyond Rothenberg and Wang (2016), which only attempted to apply the PCM to emulate a parcel model using idealized, single-mode aerosol distributions.

3. **Length of manuscript**

   Following the comments made at the beginning of this document, we have substantially revised the manuscript by re-writing the introduction, removing material redundant to Rothenberg and Wang (2016), and consolidating Sections 2 and 3. We defer including a table of GCMs and their activation schemes, instead referring the reader to Table 3 of Ghan et al. (2011). In lieu of a pros/cons table for each scheme, we have simply truncated the discussion of each, referring the reader again to Ghan et al. (2011) and other works for more information.

4. **Exclusion of aerosol modes**

   In revising our discussion of the iterative calculations, we have removed the example figure which was the source of aerosol mode confusion here. To the broader point about justifying exclusion of some of the aerosol modes and parameters, we disagree here. The OC and BC modes are excluded by assumption (they are hydrophobic with $\kappa \approx 0$). Reviewers #1 and #2 strongly emphasize that nucleation mode aerosol do not contribute significantly to activation, which we agree with. No other modes are immediately excluded. The basis for excluding the Aitken and larger dust and sea salt modes follows the results of our iterative calculations. We've added some discussion on this point to the consolidated Sections 2 and 3, but for brief reference here: in general, the largest dust and sea salt modes exceedingly rarely have number concentrations greater than 1 per cm^3, which explains why they appear as a "dominant" mode in no case of our iterative calculations. The Aitken mode in MARC is generally small (see Table 1), which limits its influence on activation, although the reviewer's comments about the nucleation (or Aitken) mode particles being large enough in number concentration to

influence $S_{\max}$ by depleting available water vapor is still relevant. Still, we believe that the results of the iterative calculations themselves provide a defensible basis for neglecting the MARC Aitken mode in the ultimate emulation parameter set.

5. **MBS hygroscopicity**

   In MARC, we assume MBS is constructed as a black carbon core fully covered by a sulfate shell. Therefore, the surface of such particles is assumed to be sulfate. In MARC, MBS forms from aging of external black carbon and continually grows from sulfate condensation and coagulation. Based on model results, the core-shell mass ratio is normally sufficiently small to support the assumption that the particle takes a hygroscopicity value comparable to that of sulfate.

6. **Sensitivity of emulator to training parameter set**

   We emphasize that our training dataset does not randomly sample from the distributions in Table 2; the probabilistic collocation method provides a particular algorithm for choosing the sample parameter sets, which are far more concentrated in the "center" of the high-dimensional set of input parameter distributions (that is, near the mean of each individual parameter distribution). The over-sampling of training parameter sets we choose is a compromise to include more of the very-high and very-low parameter values in the training dataset.

   The resulting emulator, though, should not be very sensitive to the exact choice of parameter set ranges. Following the recommendation of Rothenberg and Wang (2016), the final emulator is constructing using a least squares fit of the training parameters and the evaluated parcel model responses using a particular polynomial basis. However, the key ingredient here is the set of parcel model responses computed from the input parameter sets, which will not change unless the parameter space is dramatically altered.

   We do note, as the reviewer mentions, that some GCMs include a minimum threshold CDNC value as a tuning parameter; one reason to use activation schemes is to reduce the bias towards too-low CDNC in remote maritime regions which such a threshold aims to correct.

Minor Comments

1. **P2/L14-18**: This is removed in the revised Introduction.
2. **P2/L19**: We agree in principle, but also note that the representation of sub-grid scale processes in a GCM grid box also includes a distribution of updraft speeds, and merely an average aerosol size distribution (for instance, a grid box straddling a coastal area could include a large city and open water, and have vastly different aerosol populations in either side of the grid box depending on whether or not the flow is on/offshore). Robustly assessing activation in such a diversity of cases would require something akin to the multi-scale modeling frameworks which embed a cloud-resolving model in each GCM grid box, which we briefly mention in the manuscript.
3. **P4/L33**: Unfortunately, there is not yet a reference for the implementation of MARC in CAM5; our two references refer to the CAM3.5 implementation. We have revised the phrasing "extends".
4. **P5/L23-25,28-30**: MARC is just the aerosol model. We use the default CESM cloud microphysics.

5. **P5/L32-33**: We refer the reviewer to West et al. (2014), which includes a comprehensive assessment of the role of sub-grid scale vertical velocity on activation and cloud radiative effects, and note this in the manuscript.
6. **P6/L11**: Added emissions citations
7. **P6/L33**: Added definition
8. **P7/L1-7**: Deleted as part of the consolidated Sections 2 and 3.
9. **P7/L18-19**: We agree, but emphasize that this is an assumption and limitation in MARC - that MARC does not include a detailed representation of organic aerosol.
10. **P7/L21-22**: The NUC mode does not have a fixed size, but takes the range indicated from Table 2, which is restricted to be quite small. When larger sulfate particles form, mass and number is shifted to the larger modes, so the NUC mode can't contain CCN-relevant (by size) particles.
11. **P8/L26-30**: This discussion was simplified in the revised manuscript, and these statistics aren't discussed in the same way.
12. **P8/L31-32**: Added a note that the parameter sets were sampled from instantaneous MARC output between 70S-70N and below 500mb.
13. **P9/L11-13**: This is an interesting point, but we do not consider changes in parcel buoyancy in our parcel model simulations, which assume an adiabatic ascent.
14. **P12/L13-15**: We rephrased this entire discussion in response to another reviewer's comments, and this particular point is no longer emphasized.
15. **P12/L25**: Same as previous comment.
16. **P13/L19**: Changed sentence to read, "*Both the ARG and MBN schemes include some parameters fit to parcel model simulations conceptually similar to the one emulated here.*"
17. **P14/L8-13**: We've revised the discussion to focus more on the parameter sets sampled from online MARC simulations.
18. **P15/L32-33**: We refer to Table 2, which includes (in parentheses on the righthand columns) to percentile at which the lower/upper bounds chosen to train the model occur in the distribution of those parameters as sampled from MARC. The model does predict these very small values in some cases, although they are not physically significant.
19. **Figure5-8**: Added units to figure caption
20. **Table 2**: It's correct that κ_{MOS} can not exceed that of sulfate (0.507), but we train the model with a slightly larger range to better resolve that upper-most limit case. However, we note that the mean value of κ_{MOS} is 0.27 with a standard deviation of 0.04, so our range captures nearly the entire variability in its value.

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