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Interactive comment 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" *by* Daniel Rothenberg and Chien Wang

Anonymous Referee #1

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This research is a follow-up from an earlier work (Rothenberg and Wand, JAS, 73, 2016) in which an emulator of an aerosol activation parcel model is built. The emulator is built by utilizing a polynomial chaos expansion. In the current work, the authors evaluate the emulator of the aerosol activation process by using instantaneous aerosol and meteorological fields from the MARC model to drive the emulator. The evaluation is performed by comparing the emulator predictions (Maximum Supersaturation - Smax, and Number of activated aerosol particles - Nact) against the detailed parcel model simulations, as well as against commonly used activation parameterizations.

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The work shows that when the space of input parameters is sampled in the full range of potential combinations, which are also used to train the emulators, the performance of the emulators seems to be better than those of the activation parameterizations. However, when realistic input is used, the performance of the emulators is deteriorated and the activation parameterizations perform better, with lower standard deviation of the errors.

General Comments:

The manuscript presented here presents an interesting and somehow novel approach at generating emulators of aerosol activation schemes. The authors claim that the approach could be implemented in aerosol cloud interaction studies with climate models, and that it could be a good way of dealing with the increasing complexity of the aerosol schemes used in state-of-the-art global climate models. However, some serious conceptual problems are observed in several points during the manuscript, all of which need to be revised before the manuscript is suitable to be published. Main problems with the document are:

- The document has a lengthy historical introduction to the study of aerosol activation process. However, there is abundant literature on this topic, including review papers. This paper is basically a methods paper, in which a different approach is used to built and evaluate an emulator. As such, that should be the focus of the paper and should be written accordingly.

- Despite being focused on the description of the emulator development and evaluation, the document fails to discuss other, similar studies, based on the concept of a statistical emulator, eg., Patridge et al., Atmos. Chem. Phys., 11, 7269–7287, 2011, Carslaw et al., Nature 2013... etc.). - The paper is unnecessarily long, especially given the fact that it is a follow up paper and most of the details of the emulator development are described in an earlier work. It should be much more concise and focused on the evaluation procedure.

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- A reference is made in the abstract to GCCN, but no explanation for this is given. Is the focus of the study the analysis of GCCN impact on accuracy of the prediction?

- It is unclear what is the potential use the emulator presented here could have. The manuscript deals with offline evaluation of the emulator, but no plan or path forward for its online implementation and evaluation is mentioned nor discussed. The authors do not discuss the computational cost associated with the emulator compared to in-use parameterizations.

- Contrary to what the authors claim in the paper, activation schemes have been tested using complex aerosol distributions from state-of-the-art GCMs and chemical transport models. The activation parameterizations are not only tested with idealized aerosol size distributions.

Specific Comments

- The introduction should be made a lot more concise. The authors include here a very long historical review of activation parameterizations, but it could and should be done in a much more concise way. L33-page 3. "many evaluations of activation scheme performance have focused on the same set of relatively simple aerosol particle size distributions". This sentence is inaccurate. Many evaluations of activation parameterizations have been performed using aerosol fields produced from global aerosol models (such as MAM-3 from Liu et al., 2012), not only with idealized aerosol fields.

- I strongly encourage the authors to re-write some portions of the paper. Section 2, Activation Parameter Space, should be shorter. Use of a table (such as table 1) and a short explanation in the body of the manuscript should be enough to describe the aerosol populations produced by MARC.

- Line 19-page 6. "The different particle size distributions in MARC are influenced by both different emissions sources and acted upon by different physical processes. This leads to a great deal of spatial heterogeneity in the size distribution parameters.

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One aspect of this heterogeneity is depicted in Fig. 2,. . .." — This is a characteristic common to almost all current aerosol models used in general circulation models. Such general sentences should be removed.

- The usefulness of Figure 2 is not clear. I suggest removing it or using a more effective visualization of what is being said. For instance, a few global maps showing regional patterns or zonal averages could be utilized here instead.

- Section 2.2. Could be merged with section 2.3. This initial "reduction of parameter space" could be explained in just a few lines. It some modes have no hygroscopicity whatsoever, then, they cannot affect supersaturation balance, and will not be activated.

- Section 2.3, should be re-written. Although the authors presented a lengthy introduction to the aerosol activation problem, this section largely ignores what has already been learned in the topic for the last few decades. It is well known that nucleation mode particles are not important for activation (too small, too high of a critical supersaturation), coarse particles are important in suppressing supersaturation development (large, few, but kinetically limited) but not great contributors to the number concentration. Authors should refer to these facts, but there is no need to use 2 pages to explain the iterative procedure.

L31-p7 – "This strategy effectively employs a "greedy" algorithm to sort the set of available aerosol modes, ranking their influence on activation by their cumulative depression on the supersaturation maximum achieved for a given parcel ascent." – Consider removing this sentence since its redundant with the next paragraph.

L13-p8 "Thus, it would be reasonable to assume that the nucleation mode particles would "dominate" activation in this case - or that small changes in the burden of these particles could have large consequences on how many cloud droplets will form. . . . However, that doesn't happen." I would suggest to the authors removing this kind of sentences from the manuscript. In the lengthy introduction to the paper, the authors already explained some of the basics of the activation process. Only if the process

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had never been studied would it be “reasonable” to assume nucleation mode particles would play an important role in activation. It is well known this is not the case.

- The need for the paragraphs from line 10 to line 30 in page 8 is unclear. I would suggest removing or rewriting all the sentences from “The vast majority of particles exist. . .” to “. . . .which leads to a more rapid balancing between the parcel’s adiabatic cooling and warming due to latent heat release”. The latter sentence is completely inaccurate and reflects an incomplete understanding of the activation process. During the development of a maximum supersaturation adiabatic cooling is never balanced by the latent heat release. The parcel keeps on cooling as long as there is an updraft forcing it upwards. The latent heat release slows down the cooling rate, but what really controls the maximum supersaturation is the mass transfer of water from the vapor to the liquid.

L10-p9 “This is consistent with the physics of the activation problem; the presence of more aerosol surface area on which condensation can occur tends to produce a greater source of latent heat release to counter-balance adiabatic cooling in the ascending parcel, suppressing the development of a higher Smax” —- Same conceptual mistake. The phrase is incorrect or at least inaccurate. In aerosol activation, the reduction of water vapor is by far more important in reducing the rate of increase of supersaturation, than the release of latent heat.

- The same conceptual mistake is made again in lines 7 and 15 in page 10.

- Line 15 – Page 12. Authors should discuss in much more detail the implementation limitation of their proposed method. “ The trade-off here potentially lies in emulator performance, because the PCM will attempt to train the emulator to perform well for input parameters that we’ve assigned equal likelihood to, but may actually be far less likely to occur.” This kind of drawback seems to be shared between all the statistical models used, which are also discussed by the authors in the introduction.

- The authors devote too much time to the discussion of the emulation in the entire

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range of mathematically (but highly nonphysical) combination of parameter values. Figures 5, 6, 7, and 8 are used to illustrate the results for the entire range of potential set of input parameters. Most of the discussion of the discrepancies between parcel model simulations and activation parameterization is focused on regimes unlikely to be found (e.g., extremely clean environments with less than 1-10 #/cm3). Some other emulator studies have used expert elicitation to constrain the ranges of some parameters, so they can sample physically meaningful regions of the parameter space.

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