Reply to ACPD-13-C4190-2013 (reviewer #2) comments

The authors present a method for parameterising the size distribution of a modal model with improved accuracy via statistical fitting of numerical results. The methods avoid the need to simplify the growth kernel and are mostly computationally more efficient than numerical integration. The methods introduced here are potentially useful to model developers and as a result might be better placed in GMD after some revisions. My main comments are centred around the introduction which could be improved to improve readability and focus the reader on what is addressed in the remainder of the paper. I will provide more detailed comments but mainly the use of examples to explain all the problems is distracting and the comparison of bin and modal models could be improved.

We appreciate the reviewer's thoughtful and constructive comments to help us improve the manuscript. We followed your suggestions as close as possible in our revision, with point-by-point responses given below (in blue bold fonts). As for the suggestion of GMD, we acknowledge that it may be a better place for this type of manuscript. But for some academic issues in our country, we much prefer to publish the manuscript in ACP. Also, we feel that this topic is still suitable for ACP. Hope the reviewers and editor will agree to it.

Page 12035, Line 8: The references here could include more recently developed aerosol models. Are the methods here an improvement of even the most up-to-date models?

Reply: The following references are now added: sectional models -- Yu and Luo, 2009; Bergman et al., 2011; modal models -- Vignati et al., 2004; Mann et al., 2010; Pringle et al., 2010; Liu et al., 2012. We do not have the opportunity to test all current modal aerosol models. But we believe these models applied either lookup tables or simplified kernels and thus share similar weakness. Our approach is unique and should provide improvement in computation accuracy (but not necessarily computation efficiency as some of the modal models are highly simplified for faster computation).

Page 12035, Line 16: I find the interchange between aerosol and cloud models distracting. Perhaps add in some information about which types of model this is applicable to and how it will improve them but don't interchange throughout the text.

Reply: Agree. We believe the best and easiest way is to delete most comments about cloud models.

Page 12035/6: The details of specific model processes as examples is unnecessary in the introduction since the same can be applied to further processes. Perhaps list a few of the processes that will benefit from improving current models but don't detail them here.

Reply: Agree. We deleted such details on p.12035 line 12-16, and p.12036 line 14-17 and 20-23.

Page 12036, Line 4: Mann (2012) also compare a modal and bin aerosol model version. What models are compared?

Reply: Zhang et al. (1999) compared modal versus bin modules in several air quality models, whereas Mann et al. (2012) did a similar comparison for global chemical transport models. We have updated the discussions regarding to these studies.

Page 12036, Line 10: Some more recent modal models should be cited.

Reply: We have included several more recent sectional and modal models on p. 12035. The paragraph regarding to CMAQ and WRF-Chem has been removed to make the text more concise.

Page 12036, Line 12: What is the significance of this example? Is this the model that is the focus of this study? Please clarify.

Reply: We mentioned them here because they are quite popular community models, and the BS95 modal scheme is one of their advanced options. In addition, in the later part of the manuscript we chose the CMAQ model to test our scheme, and that is why we mentioned it as an example. But in order to shorten the text, we decided to remove this paragraph. But we mention somewhere else that the CMAQ model will be used to test our scheme in Section 4.2.

Page 12036, Line 14: ’avoid some of the problems’ - this seems a bit vague. A more concise comparison of the differences between the modal and bin schemes would be of benefit here (perhaps this will result when specific examples are removed).

Reply: Same as the point below, we have removed such details. A more concise comparison between modal versus sectional models has been added in the paragraphs above (i.e., Zhang et al., 1999 and Mann et al., 2012).

Page 12036, Line 23: ’Therefore...’ - it sounds like this study is aimed at improving the Brownian coagulation but this is an improvement on all processes where the kernel has to be simplified. Again, perhaps a list of a few processes that would be improved rather than a specific example would be better.

Reply: Agree. Instead of giving specific examples, we changed it to more a general statement: “The main weakness of modal parameterization is that analytical solutions are needed for calculating the evolution of size distribution, but the exact solutions are not always available due to complicated mathematical forms of the growth equations.

Therefore, the growth equation must be simplified to get an analytical solution; however, this simplification can lead to large uncertainties."

Page 12037: The comparison to Chen and Liu seems oddly place. A discussion of their work would be better placed in the introduction along with a review of other current methods and a discussion of how this work aims to improve upon these.

Reply: Thanks for the suggestion. This paragraph is not critical and is mostly removed to make the text more concise.

Page 12037, Line 15: This is a comparison of bin and modal models so should be earlier in the introduction.

Reply: As above. We have removed this paragraph.

Page 12037, Line 20: The mention of modes here is confusing when talking about the bin model - can you reword this somehow?

Reply: As above. We have removed this paragraph.

Page 12037, Line 24: 'If not designed properly...' - if it is designed properly this is not a weakness, is there some reason why designing the experiment properly is not possible?

Reply: As above. We have removed this paragraph.

Page 12038, Line 12: With a more concise discussion of the advantages/disadvantages of using a bin or modal scheme I think the reference back to bin modals is unneccesary here.

Reply: As above. We have removed this paragraph.

Equation 1: Make sure all parts are defined (n) and state whether they related to the particular mode or the entire distribution (n, N and _).

Reply: Yes. We have defined "*n*", and added the statement "The whole aerosol size distribution may be composed of several of such modal functions."

Equation 2: Is n(r) = n(ln r)?

Reply: They are slightly different. If expressed as n(r), there should be an extra "r" in the denominator on the first right-hand-side term.

Page 12039, Line 14: Why have you picked 3 tracking variables? Explain why 3 are necessary.

Reply: Mann et al. (2012) showed that 2 tracking variables may produce significant bias. We added such statement on the previous page.

Page 12039, Line 14: What's the relevance of the current CMAQ model here? Earlier you mentioned CMAQ used BS95? Is this the model you have used? Which scheme does it use?

Reply: Yes, in later sections we will use CMAQ to test the SNAP scheme. Also, BS95 and the CMAQ documentations provided sufficient details about the modal aerosol parameterization, so it is mentioned here in case the readers want to compare the different approaches. Some of the details of CMAQ/BS95 have been mentioned in Section 4.2.

Page 12040: The description of the gamma distribution is unneccesary. It is fair to say that the method is applicable to it but the extra maths is hard on the reader when it's not directly relevant. Perhaps it could be worked through in an appendix.

Reply: Agree. We have removed most of the discussions on the gamma-type distribution.

Page 12041, Line 1: With the gamma distribution description it is necessary here to remind the reader that what follows is a result of the log normal distribution.

Reply: Same as above.

Page 12041, Line 6: Can you explain more about the growth kernel, what it typically looks like and why it's important?

Reply: We have added the sentence "This growth kernel represents the fundamental growth equation for each process. A few examples of the growth kernel will be discussed in detail in section 3." The readers will see quite a few examples later.

Page 12041, Line 9: I would remove the word 'the' from 'the parameterisation as it sounds like you are referring to a particular parameterisation.

Reply: Thanks. It has been removed.

Page 12401, Line 25: 'four methods'? It's not clear that the methods you are investigating are new.

Reply: Since the mean-size approximation (now abbreviated as MSA) is not totally new, and is considered as a no-skill benchmark in this study, we have now revised the text and removed it from the "SNAP". Also, the other three methods are renamed as SNAP-KT, SNAP-IT and SNAP-OS.

Page 12042, Line 9: Is this the usual assumption? Is it sensible?

Reply: Theoretically, it is possible to represent any function with a polynomial with

infinite terms. But we are not making such an assumption in the actual parameterization. This section is just for a mathematical analysis for errors. We have changed the statement as "It is mathematically possible to approximate the growth kernel by a polynomial function of *r* with a sufficient number of terms (i.e.,$K = \sum_{i=0}^{\infty} a_i r^i$). We apply such a polynomial function here just to demonstrate the error associated with the mean-size approximation."

Equation 10: I got a little confused by the use of 'i' and 'k' - is it possible to clarify when one is used rather than the other?

Reply: These coefficients are indeed somewhat confusing. We try our best to make it clearer. First, "*k*" is reserved for mentioning the order of moments (e.g., the k^{th} moment). Then, "*i*" is used to represent the overall order of the growth kernel. We try to adhere to such definitions throughout the text.

Equation 16: Can you define K_k in this first instance since there is an appearance of r_k that isn't in Line 7 or Equations 1 and 9.

Reply: Yes. It is now defined as $K = \sum_{i=0}^{\infty} a_i r^i$. See earlier reply (2 points above).

Page 12045: This example seems out of place here. In the other cases you have only explained the method.

Reply: Thanks for pointing this out. We have moved this example to Section 3.

Page 12046, Line 10: How is this method better than Whitby et al?

Reply: We are not implying that our method is better. In fact, the two methods are of similar accuracy. We just want to point out that such a formulation is not new, even though the approaches in deriving them may be different.

Page 12046, Line 10: This paragraph seems very negative about the method and so it's difficult to see why this example is included.

Reply: It was meant to point out the potential use of transforming only the kernel which may be used elsewhere (e.g., the term Cc). As the whole section is now moved to Section 3.2, this paragraph is no longer needed and is thus deleted. Also, the comment about the term Cc will show up in Section 3.4.

Page 12047, Line 5: Is this a particular artefact of the software you are using and could a better software be used?

Reply: Not many statistical software packages can provide sophisticated nonlinear fittings. We have not seen any other that does a better job than the one we are using.

Unfortunately it can handle only two variables at a time.

Page 12047, Line 18: What does 'strong function' mean?

Reply: The sentence is changed to "The corrector g_1 should depend strongly on the spectral width σ.

Page 12048, Line 17: What about SNAP-B? It already sounds like it's been outdone by C and D?

Reply: Yes, for most processes. But it is still useful for deriving diagnostic formulas as mentioned later in Section 3.5. We have deleted the comments here, and put the summary statement only in the conclusion section.

Page 12049, Line 9: Why is it not included and will it be in future?

Reply: It is worthwhile emphasizing its importance here, and we are in the process of including it in the model. The paragraph is now modified as "This process is usually not considered in traditional aerosol models which do not emphasize aerosol-cloud interactions. On the other hand, current cloud models generally do not consider the emission and production of aerosol particles, so the ice nucleation process is highly parameterized due to the lack of realistic ice nuclei. Because of the importance in climate and hydrological cycle, detailed aerosol-cloud interactions have become an essential component in advanced regional and global models, for which ice nucleation is a critical mechanism that badly needs improvement (cf. Tao et al., 2012)".

Page 12049, Line 14: Explain modes here since it is unrelated to the modal scheme.

Reply: Thanks for pointing this out. To avoid confusion, "modes" is changed to "pathways".

Page 12050, line 11: What is meant by the modal size? The mode covers a range of sizes.

Reply: The modal size μ have been defined in Eq. (5). It is the same as the statistical term "mode" which means the value that appears most often in a set of data. To avoid confusion, we modified it as "the modal size μ".

Page 12051,Line 16: How long does the trial by error take? Is this valid only for this model or is it transferable?

Reply: It depends on the complexity of the equation as well as the depth of our understanding in the physics. Typically each process takes about a week of work for us. The parameterization approach in principle can be applied to any physical or chemical processes, and the derived parameterizations are transferrable (either an individual

process or the whole package). To make the point clearer, this paragraph has been modified as "Hints of the proper parameters may emerge while examining the fundamental physics and its mathematical formulation. For example, one may recognize that q in Eq. (19) is the most pertinent parameter for heterogeneous ice nucleation. On the other hand, Eqs. (3) and (9) indicate that the variance σ^2 is a key to the representation of size spectrum. So, we selected $\bar{q} \equiv \mu/r_g$ and σ^2 as the parameters for statistical fitting."

Page 12051, Line 18: I don't understand the use of the word 'might' here. Does it?

Reply: This sentence has been modified as above.

Page 12052, Line 2: What is the 'whereas' referring to?

Reply: This word is deleted.

Page 12052, Line 9: How difficult is it to decide which formulas to use? Is it normally quite straightforward or would someone else perhaps choose a different formula?

Reply: Any formula picked up by the software will give good accuracy. But it does require some intelligent judgment to choose a form that better reflects the physics, which can aid the analysis or discussion. It also takes some programing experience to choose a mathematical form that avoids numerical problems.

Page 12052, Line 19: Can you compare the CPU times too?

Reply: Yes. We added the statement "The CPU time required for SNAP-KT, SNAP-IT and SNAP-OS are 73%, 26%, and 18% more than for the MSA method, respectively."

Page 12057, Line 16: When might SNAP-B be practical? Is there a general sense of when there is no point even trying it?

Reply: We found that either SNAP-IT or SNAP-OS will out-perform SNAP-B (now called SNAP-KT) for process equation that involve integral over the size spectrum. But for those that are not related to size distribution, SNAP-B must be used. We mentioned this in section 3.5 and the conclusion.

Page 12060, Line 18: Which SNAP method is applied here?

Reply: As above, the diagnostic equations are parameterized using SNAP-B. Text is revised accordingly.

Page 12061, Line 8: 'Apparently,....' according to who? Why?

Reply: This statement is based on the previous sentence "The size difference due to the Kelvin effect increases with humidity, reaching about 50% at 95% relative humidity and near infinity as the relative humidity approaches 100%".

Page 12063, Line 11: Will this have any impact on the robustness of the results?

Reply: "The simulations were run in parcel mode to avoid complications from other processes, such as transport and sedimentation." This should not affect our results. In fact, we think such a practice can provide a clearer picture about the accuracy of individual processes, and also avoid possible cancellation of errors from different processes. Note that the transport and sedimentation processes are examined separately in Section 4.2.

Page 12064, Line 14: Which processes was SNAP applied to?

Reply: SNAP is applied to the gravitational sedimentation and surface deposition. Text modified accordingly.

Page 12065, Line 12: Is there a reason for the specific areas with large errors?

Reply: The large relative error occurred mainly over areas that are raining and the aerosol concentration is low. This also means that the absolute errors at these locations are actually rather small. Text modified accordingly.

Page 12065, Line 14: A SNAP parameterisation of the Kelvin effect is included?

Reply: No. The modal aerosol module in CMAQ does not consider the Kelvin effect. So we also ignored it to be consistent.

Page 12066, Line 18: Do the specific formulas apply to other models or would new coefficients/formulae be required?

Reply: The formulas should be able to be used in other models without changing the coefficients (except when a different unit system is used).

Page 12067, Line 11: I would say that the conclusion regarding the Kelvin effect was reached by application of SNAP - otherwise it seems out of place and unrelated to the SNAP methodology.

Reply: Agree. The sentence is modified as "In this model, we further applied a SNAP diagnostic formula for the commonly ignored Kelvin effect, and showed that this effect cannot be ignored in aerosol modeling".

Figure 3: What's the scale of the coloured dots - it's hard to see how large/small the

deviations are.

Reply: We now explained the color dost in the figure caption: "The degree of deviation is also indicated by the color of the dots: blue, green, and yellow represent less than 1, 2, and 3 standard error, respectively, whereas red denotes greater than 3 standard error.

References: Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model. G. W. Mann, K. S. Carslaw, D. A. Ridley, D. V. Spracklen, K. J. Pringle, J. Merikanto, H. Korhonen, J. P. Schwarz, L. A. Lee, P. T. Manktelow, M. T. Woodhouse, A. Schmidt, T. J. Breider, K. M. Emmerson, C. L. Reddington, M. P. Chipperfield, and S. J. Pickering Atmos. Chem. Phys., 12, 4449-4476, 2012

Reply: Thanks for providing this useful reference.