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Interactive comment on “A statistical-numerical aerosol parameterization scheme” by J.-P. Chen et al.

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Received and published: 14 August 2013

This manuscript describes a statistical parameterization scheme SNAP for modal aerosol models with the intention to improve the accuracy of approximate analytical solutions of the size distribution evolution. While the scientific approach presented is interesting and potentially useful, the manuscript is a very laborious to read and, because of this, I worry that it may lose some of its target audience. I give some suggestions below how the readability could be (at least partially) improved, but I encourage the authors to give this a serious thought themselves as well. My second major comment is that the manuscript would be much better suited to ACP's sister journal Geoscientific Model Development, and if it is possible without an additional review round, I strongly

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recommend it to be transferred to GMD.

Reply: 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. 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.

Specific comments: The manuscript is very heavy on the mathematical equations. Consider whether you could move some of the mathematical details to appendices.

Reply: Equations in section 3 explain the physical processes and how the SNAP scheme can be applied, so we think those details are useful for reader who want to adopt SNAP. Section 2 can be shortened. We have deleted the discussions and formulas regarding to the gamma-distribution which are not critical to this study. Also, the example on sedimentation flux is shortened and moved to Section 3.

The terminology used is often quite technical (e.g., 'offline spectral integration', 'evolution tendency is solved by in-line numerical integration') which makes the text hard to follow at times. Try and be more concrete, this will help the readers that are not directly familiar with this terminology. In addition, in many places it would helpful to focus better on the specific topic at hand, rather than discussing several related points simultaneously.

Reply: Thanks for the suggestions. We tried to revise the text accordingly. For example, the term "offline spectral integration" has been changed to "offline numerical integration"; and the paragraph containing "evolution tendency is solved by in-line numerical integration" has been deleted all together.

Introduction: It takes a quite a bit of work for anyone without extensive hands-on experi-

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ence with modal models to understand what this study aims at and why it is important. In this light, I recommend the authors to cut the majority of the general aerosol and aerosol model description on pages 12034 and 12035, and concentrate in more detail to the specific problem that they are addressing in this manuscript, i.e. simplification of the distribution evolution integrals to obtain analytical solutions. E.g., why are these analytical solutions needed and what makes obtaining them so difficult (e.g. compared to bin schemes); a general overview of previous attempts to solve this problem (also beyond BS95; some of material from section 2.2 could be moved here) and what can be considered the current state-of-the-art method; how this method improves upon the previous methods, etc.

Reply: Combining both reviewers' suggestions, we have simplified the description in the introduction and in section 2. Specific problems in modal model that introduced in earlier studies (i.e. simplification of the distribution evolution integrals to obtain analytical solutions) are briefly mentioned in the introduction and more details given in section 2.

P. 12038: I would also cut the discussion on the similarities and differences to the earlier Chen and Liu (2004) study, as it is not directly relevant here.

Reply: This paragraph has been removed as suggested.

Intro: The introduction misses lots of references e.g. to models that are discussed.

Reply: A few more relevant references are added and briefly discussed (also see comment #3).

P. 12035, l. 14-16: there are many bin models that activate only a fraction of particles in the bin; several techniques to do this as accurately as possible have been developed.

Reply: Bin models are not the focus of this study, therefore such information is probably too detailed for the introduction section. So we have removed this paragraph.

P. 12306, l. 15-17: The modal models have in-built problems with step-function type

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processes such as activation. It is therefore misleading that imply that a continuous size spectrum in the modal model would automatically treat this process more accurately than a sectional scheme.

Reply: Agree. We have removed these statements.

The gamma distribution on p. 12040 is not further discussed in the manuscript and can be moved to an appendix.

Reply: We have deleted most of the discussion and keep only a brief comment.

It might be helpful to summarize briefly and using clear terminology the essence of the four method already in section 2.2. E.g. (1) SNAP-A: assume a constant particle size ..., (2) SNAP-B: transform growth kernel by ..., etc.

Reply: They are now summarized at the end of section 2 and also in the conclusion section.

Why is SNAP-A included when it is quite clear from the start that this is not a good approximation for non-monodisperse modes (it is later called ‘no-skill reference’)? If you decide to leave SNAP-A, consider if you can condense some of the text in 2.2.1 and remove the equations which are not of direct relevance to what is done later in the study. This goes with the description of all the methods as well.

Reply: It is true that SNAP-A is not very accurate and is used only as a benchmark. But it is also used as a base for SNAP-C and SNAP-D (i.e., SNAP-C and SNAP-D prove adjustment factors for SNAP-A). In view of this, we have excluded it as a part of the SNAP scheme and instead called it “MSA”. Other SNAP schemes are also renamed, which are SNAP-KT, SNAP-IT and SNAP-OS (to replace SNAP-B, SNAP-C and SNAP-D, respectively).

P. 12045-12046: Indicate in text whether you are talking about Figure 2a or 2b.

Reply: Thanks for pointing out the ambiguity. Both the “Fig. 2” on p. 12045 and 12046

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should be Fig. 2a (note: now renumbered as Fig. 3a).

Section 2.2.2: It is confusing that sedimentation velocity is discussed as an example here but no such examples are given for the other methods. Remove it, or alternatively choose a simple process that you can work through with all the four methods (this would illustrate the differences between the approaches in a concrete way).

Reply: Agree. The paragraphs on sedimentation is shortened and moved to section 3.2. This will keep section 2.2 simple and easier to read. Also, ice nucleation in section 3.1 now becomes the first process discussed, and we use it to work though all four methods.

Eq. 23: what does ‘modal-value approximation’ mean? Is it the same as eq. 11? If so, call it by the same name; if not, change the notation to something less confusing.

Reply: As we defined in Eq. (9) {note: Eq. (12) in the original version}, there are different ways of defining the “mean size”. Modal-value approximation takes the modal value μ to represent the mean size as given in Eq. (11) {note: Eq. (14) in the original version}. We also indicated later in the text that the modal-size approximation is a special case of $n=0$ in Eq. (12). We added “cf. Eq. (11)” after the term “modal-value approximation” to refresh the readers’ memory.

Is Eq. (32) used for both f terms in equation (25)? If not, won’t this be inconsistent?

Reply: Eq. (32) is use only for f in the exponential term in Eq. (25), whereas Eq. (29) is used for “ f ” in the square root term. Our calculation indicated that using different parameterization formulas for the same variable in different part of the equation won’t cause any problem (as indicated in new Fig. 4).

Figure 4 & P. 12502: Visually it is close to impossible to see whether SNAP-D (or any of the other methods) performs better - contrary to what is claimed in the text. What are the error values for I2 and I3 with the other methods (now one cannot judge whether SNAP-D performs better)?

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Reply: Most of the SNAP-D dots are packed along the 1:1 line (i.e. relatively few scattered dots), so we thought this would tell that SNAP-D is better. But the symbol color did not come out as clear as we wished. We have modified the figure and hope the clarity has been improved. But to be safe, we deleted this sentence so that our claim is based on the error values, not the figure. In addition, we noticed that SNAP-A (now called MSA) results were not presented correctly here because in the earlier calculation we did not apply the modal size to approximate the r^k term (i.e., it is kept in the integral which can be solved as M_k). Strictly speaking, this is not a pure MSA. Thanks for leading us to find this problem. We have corrected it and redraw the figure. The error values for I2 and I3 are provided. The paragraph now reads as: “The mean errors in I0 are 317% for MSA, 22% for SNAP-KT, 63% for SNAP-IT, and 16% for SNAP-OS. These errors tend to increase toward higher moments. They are They are 2800%, 25%, 60%, and 12%, respectively, for I2, and are 15100%, 34%, 73% and 22%, respectively, for I3.”

P. 12053, last line: Vapour density at the surface is not size independent (Kelvin term)! I also disagree with the statement later in the text that the Kelvin effect is often ignored in aerosol models. What is this claim based on?

Reply: Yes. Therefore we stated that “If we assume that . . . is independent of size”. We make the simplification here to facilitate the discussion as the complete equation is too cumbersome. We also emphasize this by modifying the latter sentence as “However, in reality the size dependence of f_g and $\gamma(v,p)$ cannot be ignored, particularly for small aerosol particles”. In several places in later text we further stressed the importance of the Kelvin effect. Regarding to the Kelvin effect, we meant to say that it is commonly ignored in “modal aerosol models” not all aerosol models. Thanks for pointing this out. There are indeed a few modal models that do consider the Kelvin effect, but models such as CMAQ and WRF-Chem (at least the publicly available versions) do not. To clarify this, we removed the statement “the Kelvin effect is often ignored in aerosol models” and added the sentence: “Yet, many modal aerosol models considered only

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water uptake due to the Raoult effect (e.g., Jacobson, 1997; Mann et al., 2010). A few that did take the Kelvin effect into account (e.g., Ghan et al., 2001) need to utilize a convenient form of the Kelvin equation which is applicable only for sufficiently dilute solutions.”

P. 12058, l. 20: 'selected amount of data' what is this selection based on?

Reply: We systematically take one out of every 5 or 10 consecutive points, depending on the amount of data. Explanation is added in the text.

Throughout the text: change appendices A and B to Table A1 and Table A2.

Reply: Modified as suggested.

Section 3.4. is quite confusingly formulated. Condense the text and focus on the aspects directly relevant to this study. Regarding nucleation, there is clear consensus (e.g. based on the CLOUD experiments, which are the best nucleation experiments to date; see Kirkby et al., 2011) that binary nucleation alone does not explain observed nucleation rates. Regarding Kelvin effect: this effect causes the small particles to absorb less water but is negligible for larger particles; indicate the size of particles in Fig. 8 in the text; what is 'r₀'?

Reply: Section 3.4 has been revised significantly to make it smoother for reading. We believe now all content are directly related to this study, as the parameters discussed are all relevant to aerosol modeling. As for the question that binary nucleation is too weak to explain the observation, we are aware of such conclusions. However, as mentioned in the text, it is possible that earlier calculations of binary nucleation did not consider the variation of surface tension with size, which is quite strong at the sizes of nucleation embryos. When this effect is considered, the nucleation rate can reach a realistic level. In any case, we did not deny the importance of other nucleation mechanisms. Perhaps our method can be applied to other nucleation mechanisms in the future. Regarding Kelvin effect, the size dependence is clearly demonstrated in Fig.

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8 (new Fig. 9). We have indicated the size of the particle (0.01 micron) in the figure caption. The term r_0 should be r_{eq} . Thanks for pointing out this typo.

P. 12062 bottom -12063 top: are you all the time verifying the sectional model here? “and found similar results.” what are these results (the readers shouldn’t need to go back to the earlier study).

Reply: Yes, the verifications in section 4.1 are all against sectional model results. As for the “similar results”, we explained it in the revision as: “We re-conducted the verification for aerosol size scales and found that the bin model acquired similar high accuracy and conservation of the moments.”

Figure 9: The text is not entirely fair and seems to favour SNAP. E.g. at 6 h, SNAP does not capture the smallest particles better than the other two methods. “at somewhat larger sizes to the left of the modal size.” what does this mean?

Reply: Agree, the text is now modified as “For example, the BS95 and GHQ distributions deviate more obviously at the small end at 1 hr as well as at the large end at 6hr, whereas the SNAP distribution deviates more at the small end at 6 hr.”

End of section 4.1: Give values also for the intra-modal coagulation test.

Reply: We added this sentence: “For SNAP-IT, GHQ and BS95, the errors in M_0 are 0.028%, 0.092% and 0.090%, respectively; whereas for M_2 the errors are -0.03%, 0.103% and 0.103%.” The time evolutions of M_0 and M_2 are given in the attached figures, but we will not show them in the manuscript.

Section 4.2 incorporates SNAP into regional models only in terms of some specific processes while (apparently) simulating the rest of the processes with the old formulation. I do not see the point of such a comparison and would remove this section. However, if the editor disagrees, see also the following comments.

Reply: Quite a few processes or mechanisms (transport, sedimentation, Kelvin effect and more) cannot be analyzed with the parcel model in section 4.1. So we wish to

address some of them in Section 4.2 using regional models. We intentionally separate the processes, because errors from different processes might offset each other. Also, it is easier to explain the model differences with single-process sensitivity tests.

Figure 11: would be clearer to show the following two panels: baseline and difference to baseline.

Reply: Thanks for the suggestion. They have been modified accordingly, with the addition of 6-bin results (see below).

P. 12064, bottom: it is clear that the modal model with 6 tracers is faster than the bin scheme with 12 tracers; however, do you need 12 bins to obtain this good a solution or could the number of bins be reduced without losing accuracy?

Reply: This is an interesting question. We answer this by running an additional simulation using only 6 bins. The revised figure shows that results using 6 bins are indeed less accurate than those using the 6-parameter modal approach (assuming that the 12-bin results are exact). We also performed another analysis to investigate the dependence of accuracy on bin number. A new figure (Fig. 2) shows that the error decreases by more than two orders of magnitude for an order increase in bin resolution. The example used here is the intra-modal coagulation rate, and the error may be different for other processes (generally smaller for processes/equations that involve only single integral). We briefly mention this analysis in the beginning of section 3.

Typos: - p. 12037, l. 19: 'gas'??? Reply: Thanks. It should be "gap". Note that we have deleted the whole paragraph.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 12033, 2013.

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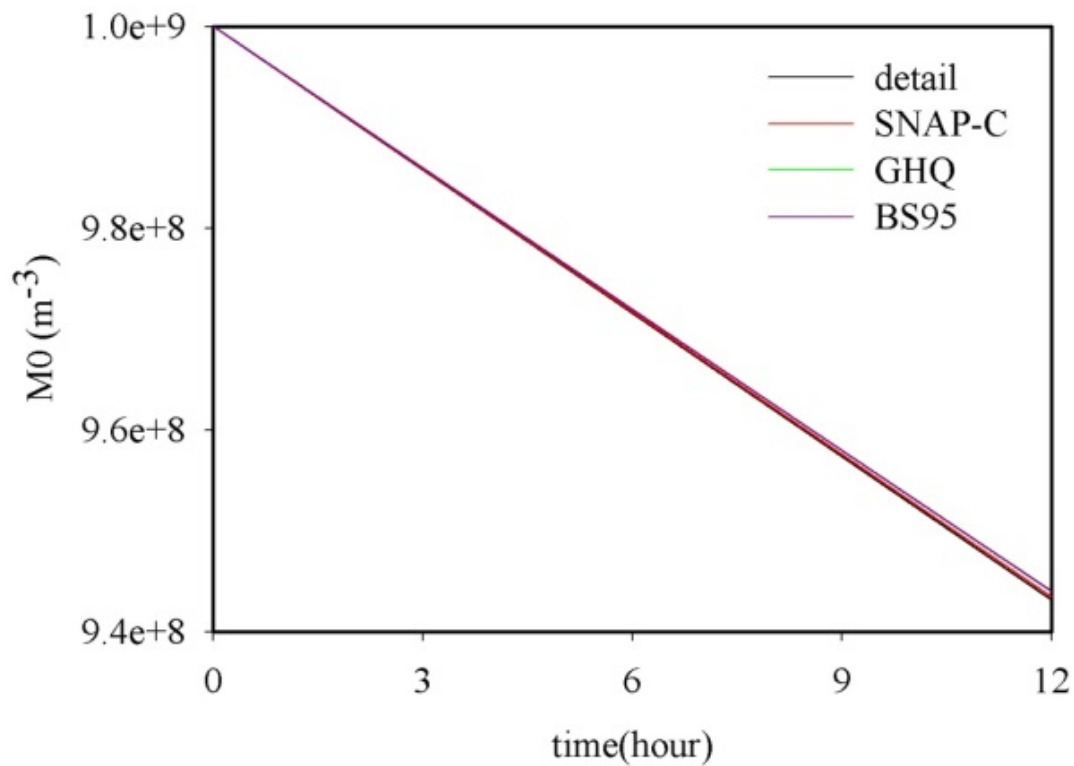
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Fig. 1.

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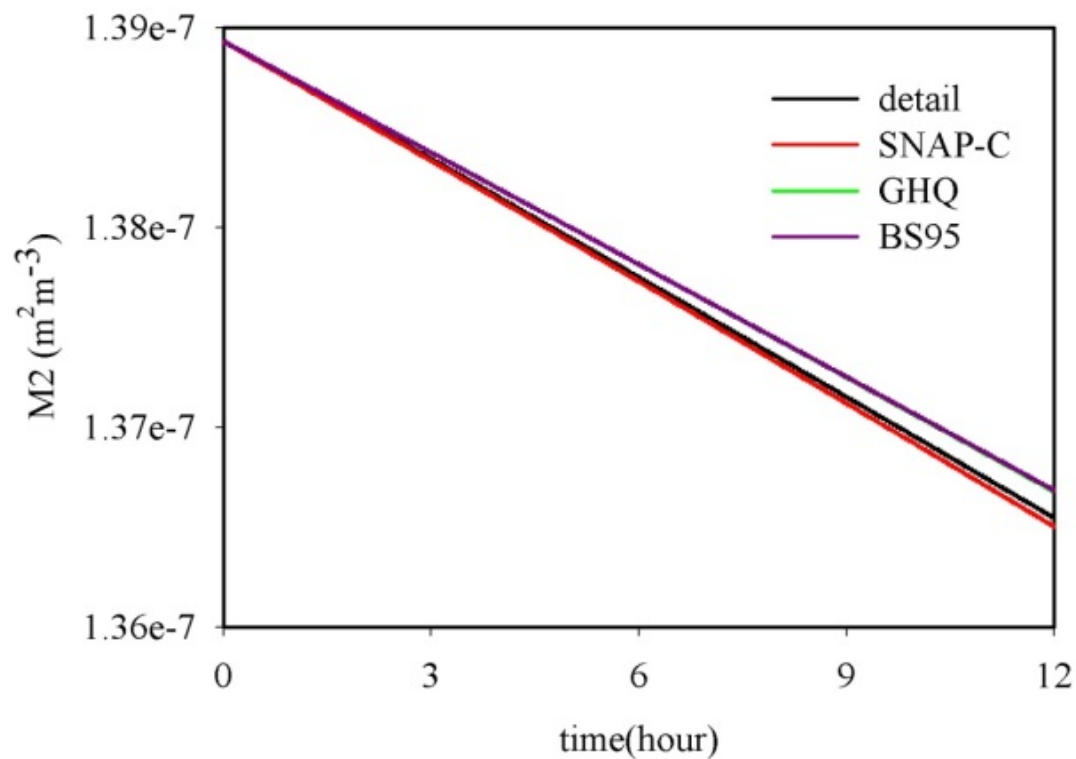
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Fig. 2.

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