Arabas and Pawlowska
Adaptive Method of Lines for CCN Activation
Geoscience Model Development Discussions

Reviewed by Jeff Snider, University of Wyoming

Overview

Arabas and Pawlowska contribute significantly to a long-standing problem in cloud physics. At issue is the simulation of cloud droplet nucleation on a discrete population of particles (nuclei) and the particle size bifurcation that results as the humidity reaches it maximum value. By way of the scheme developed by Arabas and Pawlowska, the concentration of nucleated droplets and the particle size bifurcation are better resolved. Both of these properties need refinement in numerical aerosol-cloud studies.

My main criticism is that the paper does not provide a justification for the computational expense of tracking the droplet temperature. In Figure 2 we are shown that growing droplets are warmer (~0.1 K warmer than the air), as is expected, but we are not told what the ramification is. Since this aspect of the problem is often approximated by evoking the Maxwell-Mason formulation (e.g., Equation 7.18 of Rogers and Yau) it would be useful to have a comparison of two model simulations - one with the droplet heat budget analyzed (Equation 6) and one with the Maxwell-Mason treatment.

The "tolerance", as defined by Arabas and Pawlowska, is a bin-threshold concentration that triggers splitting. Presentation of a "concentration" with dimension of count per unit mass is common, but I was confused by the presentation on P1293. For the marine accumulation mode, the correct value should be 0.3 per milligram (0.005*60 per milligram) not 0.3 per microgram. The authors need to check carefully for this mistake. Also, see the X-axis in Figures 3 and 7; these should be labeled with per milligram not per microgram (I think).

My final criticism is about the comparison to Twomey in Figure 8. As the authors point out, this is complicated because the spectrum of nuclei (cumulative N versus SS) is different, and because "transition" regime effects, particle composition, etc. are different. It is my opinion that a useful comparison requires the same CCN activity spectrum, numerically and in Twomey's analytic scheme, and proper accounting of the transition effects. As an example, see Table A1 in Snider et al. (2003) and our discussion of the effect of a varied condensation coefficient.

Detailed Comments

P1273L8
The *difference* between the ambient and the surface vapor pressures is commonly referred to as the "driving factor."

P1273L29
What is a "…positive-defined algorithm" ?   I see this defined on P1285, perhaps you should reference parenthetically to that section of the paper.

P1274L11
The Snider et al. (2003) model is described in detail, it is "moving-sectional", it treats the growth of particles differently depending on their response time and compares to Twomey's analytic formulation and the Young formulation.   In my opinion, Snider et al. (2003) is an appropriate reference here.

cloud-drop -> droplet

drop -> droplet

P1276L15-16
This sentence is confusing.   We are told that the model is "…aimed at describing the growth of aerosol at high humidity, and corresponding high relative water contents within particles of the wet spectra."   Would it be simpler, and sufficient, to state "…is designed to described the time dependence of the wet particle spectrum."? Furthermore, you can ameliorate my criticism about the drop density (see below) with discussion, written here.   Specifically, it needs to be stated that for the conditions modeled (RH>95%) the mass of the salt is small in comparison to the mass of the water. As a result, the drop density in Equation 11 and the drop surface tension (Equation 12) can be assumed independent of drop composition.   Related to this, you have the drop temperature as a derived parameter.   Why not allow the temperature/size dependence to influence the Kelvin term in Equation 12?   I.e., σ / T_w is temperature dependent and the numerator's temperature dependence is amplified by the temperature dependence of the denominator.

P1276L20
Tracking of the temperature of each size and each chemical component is admirable, but is the sensitivity of the calculation to drop temperature significant and therefore worth the computational expense?   Please see my general comment in the introduction.

P1277L10
The vapor diffusivity is also pressure dependent.

P1278 Equation 11
1) Since the "total mass" has contributions from water and salt (P1276L16-L17), and the drop density has contributions from water and salt (see P1277L5), the drop density needs to be behind the time derivative. If not, then you are assuming that the drop density is a constant for all sizes and chemical classes. If you are not going to make the assumption that the drop density is constant, then a constitutional parameter is needed behind the time derivative. See Equation A8 in Snider et al. (2003)
2) Parenthesis should go around the product of the third moment and the sum of number concentrations.

P1278 Equation 12
1) You have a droplet temperature for every size and for every chemical class (P1276L20), but the Petters and Kreidenweis (2007) Koehler equation assigns all drops the same temperature, surface tension and drop density. Two of the effects (temperature and surface tension in the Kelvin part of the PK07 Koehler equation) are significant; they can shift a particle's critical supersaturation by as much as 0.1 % (Sc going from 1.0 to 1.1%, for example). There is discussion of this in Section 4.5 of this reference:

http://www-das.uwyo.edu/~jsnider/snider_jgr_2010.pdf

2) In Equation 12, I suggest you state the assumptions (e.g. constant surface tension in Equation 12, constant drop density in Equation 11).

3) In Equation 12, I suggest that you show the water activity explicitly as a function of wet and dry radius.

4) "Water activity" is not the "Raoult term." "Raoult" also appears somewhere else in the paper.

5) Show explicitly the multiplication in the second term of the denominator.

P1279L9
A semantic issue, but it seems that a real gas can have temperature-dependent heat capacities and still behave "perfectly." In fact, you are making the common assumption that the heat capacities are constants for all temperatures and pressures.

P1280 Equation 17
Parenthesis should go around the product of the third moment and the sum of number concentrations.

P1282 Equation 19
1) The vapor diffusivity is also pressure dependent.
2) The second term on the right hand side comes from the coupled heat/mass budget equations (Equations 4, 5, 6, 7, 8, 9 10).   The path from Equations 4-10 to Equation 19 is not clear. Since this coupling is novel (apparently), it is important to develop the path.   Please don't leave the algebra for the reader.
3) Thanks for pointing out Maxwell-Mason in lines 4-6.

P1283 Equation 20
Here you are developing complexity with the subscript "m", yet "m" and "c" seem to be the same thing. Is the subscript "m" necessary?

P1283 Equation 21
Classically, the equilibrium state is defined by the Koehler equation at the initial RH.   Is the second of these two equations stating that?   The drop size initialization is detailed on the next page; perhaps that text should be merged with this text.

P1283L17
This is the first discussion of the "dilute" approximation for the solution. In my opinion, this discussion needs to be in Section 2.1, and clearly articulated.   In that case, my criticism about the drop density and drop surface tension would be addressed.   My point is this: For RH in excess of ~95%, the mass of water is large in comparison to the mass of salt. Hence, the drop density and the surface tension can be assumed independent of composition, although they may still be functions of temperature. As we discussed in Snider et al. (2010), the effect of temperature on $\sigma(T_w)/T_w$ is significant for some nuclei. Also relevant is this paper:

Growth of Cloud Drops by Condensation: Effect of Surface Tension on the Dispersion of Drop Sizes, R.C.Srivastava, Journal of the Atmospheric Sciences, Volume 48, Issue 13 (July 1991) pp. 1596-1599

P1285L23
Why limit moment calculations to droplets smaller than 25 um?

P1287L21
built -> built

P1289L2
Given that the response time of supermicron seasalt particles to is quite long (>1 second), is it appropriate to start the simulation at this large of RH?

P1290L9
The phrase "The boundary of the very bin…." is confusing.   Also see P1289L16, similar comment.

P1292L4
stronger that due – > stronger than that due

P1293L19
(suggested change)
Both being the focus of studies of cloud albedo and climate impacts

P1295L1-L4
I do not understand the point of this paragraph.

Figure8
The green and red fields for Twomey is confusing.   Why not use a line?