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Interactive comment on "Revisiting Twomey's approximation for peak supersaturation" by B. J. Shipway

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General Assessment

This work presents an alternative method of computing the activation fraction of cloud condensation nuclei in an adiabatic parcel, with the goal of developing a new parameterization for atmospheric models. An improved expression to the so-called Twomey's lower bound approximation for the condensation integral is developed. Although itself it is not a substantial novel contribution, the manuscript is well written and it is of interest to the atmospheric community. Aerosol activation still lies at the center of the uncertainty in the determination of the aerosol indirect effect. The paper would be suitable for publication in ACP after some clarifications regarding the limitations of the proposed

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approach are introduced.

General Comment

In general the expression developed is only useful for an idealized case where kinetic limitations to droplet growth are not significant. Other parameterizations have tried to incorporate these effects which may have an impact on the formation of cloud droplets in non-idealized conditions. Thus, besides the arguable mathematical simplicity of the proposed approach it is hard to see novelty or even a substantial new contribution in this work. Even though some of these limitations are mentioned in the work the authors should be more self-critical of their own work and explicitly mention in the abstract and in the conclusions the idealized conditions for which the proposed parameterization is applicable.

Many thanks for taking the time to review this manuscript. Your comments are much appreciated. However, it must be stressed that the current work seeks to find a simple, more efficient and more accurate numerical treatment (parametrization) of an existing, widely used and accepted physical model of the activation process. It does not aim to develop a more complex or more complete physical representation as has been the motivation of many other recent papers. This development is not in conflict with the development of a more complete representation, but notably highlights the errors introduced through Twomey's approximation, which are significantly reduced with the proposed parametrization. The sample results provided demonstrate that these errors are of a similar magnitude to the errors introduced by ignoring, for example, kinetic effects. In the context of current parametrizations widely used in climate and regional NWP models (where errors introduced through lack of subgrid variability further outweigh those of the approximations made to the physical model), the proposed parametrization offers a viable alternative which is competitive in performance and expense.

Specific Comments

Page 25902, Line 15. This line is misleading. It is true that the lookup table
is independent of chemical characteristics of the aerosol, however a multimodal
aerosol population with N species still requires 2N +1 inputs to find the activation
fraction. It must be emphasized that this statement refers only to the condensation integral.

This has been clarified in the abstract:

While multimodal aerosol with N different dispersion characteristics requires 2N+1 inputs to calculate the activation fraction, only N of these one-dimensional lookup tables are needed.

2. Page 25902, Line 17.The authors should explicit mention here that the parameterization is only applicable to cases where kinetic and inertial limitations to droplet growth are negligible and when the particles are in true equilibrium with their environment prior to activation.

See following comment.

3. Page 25903, Line 25. It must be mentioned, not only in the appendix, what the underlying assumptions are: non continuum effects are neglected, and the initial size of the particles is negligible compared to the droplet size, and all particles are in true equilibrium with their environment.

I have added the following to the main body of text following equation (1):

We note that in order to obtain Eq. (1) it is assumed that i) the initial particle size is negligible compared to the droplet size, ii) particles grow in equilibrium with the environmental relative humidity, and iii) kinetic and inertial limitations to droplet growth are ignored. These and further approximations are discussed in Ghan et al. (2011), while more detail surrounding the derivation and approximations used to obtain Eq. (1)

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is provided in Appendix B. We also note that although kinetic effects are not explicitly treated in Eq. (1), the parametrization of Fountoukis and Nenes (2005) is employed, which uses a size averaged diffusivity coefficient to account for changes in diffusivity with droplet size (c.f. Shipway and Abel, 2010).

I have not added this to the abstract, which I believe succinctly summarized the focus of the paper, i.e. the improved treatment of Twomey's approximation and the method of solution.

4. Page 25904, line 11. It must be simply.

Corrected

- Page 25904, Line 15. It must be mentioned that this only applies to those particles in true equilibrium with their environment. The expression is not applicable for cases where giant CCN or surface adsorption are significant.
 - I have added in the caveat 'and assuming droplets remain in equilibrium with their environment' to the preceding sentence.
- 6. Page 25096, line 20. The hygroscopicity parameter (kappa) is nowadays a very common way to express these relations. This should be at least mentioned.

The details of the values used for b and β were also omitted, thus I have added the following:

In essence, b represents the solubility of the soluble fraction, while the representation of the distribution of this soluble fraction within the particle volume is determined by β . In the application demonstrated here, we assume the soluble fraction of the aerosol is uniformly distributed within the particle volume and so $\beta=0.5$. Under this as-

sumption, the parameter b is equivalent to the more commonly used hygroscopicity parameter κ (Petters and Kreidenweis, 2007)

7. Page 25907, line 24. Using t as a dummy variable and also as time just brings confusion. Please clarify.

I have replaced t with $\hat{\sigma}$ for clarity.

8. Page 25909, line 6-8. "Accurate" here is quite ambiguous. Since the only way to achieve the form of Eq. 14 is by introducing assumptions that other authors do not use and that have been proven to lead to systematic biases (see for example Betancourt and Nenes, 2014 and Simpson et al. 2014), it is hard to see whether the proposed approach is indeed more accurate.

In general, I agree that discussion of the accuracy of the scheme in relation to other schemes or the true solution is hard to demonstrate with a limited set of models and a limited input parameter space. No attempt was made in this paper to carry out such a comparison, rather the ongoing work of Partridge et al. will hopefully elucidate this aspect. In this instance, however, the accuracy is in relation to Twomey's approximation to the inner integral for equation (1) and the improved accuracy of the approximation is not ambiguous (c.f. Figure 1).

9. Page 25912, line 5. This is agreement by design as the same data was used to fir Eq. 16.

See following comment.

10. Page 25913, line 6. Again this is agreement by design.

I believe this is a good representative sample covering a wide range of updrafts and aerosol concentrations. Thus the agreement demonstrates it is a reasonable parametrization, whether by design or not. Confidence in this parametrization is further consolidated through the analysis in Appendix B which comes from an entirely different and independent approach.

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- 11. Page 25914, line 15-20. It is no clear how the proposed approach could be modified in the future to account for these effects.
 - Agreed. This would be a question for follow up research, but beyond the scope of this work.
- 12. Page 25915, line 9. It is not clear that the proposed parameterization is much more computationally efficient than others. For example, the (Abdul-Razzak and Ghan, 2002) parameterization is simply an algebraic expression that does not require iteration.

Numerical cost is also difficult to evaluate precisely since different codes will optimize differently on different platforms with different compilers. However, in this regard, the current method will be considerably quicker than those which require the evaluation of the condensation integral either through quadrature methods or using mathematical functions such as the error function or hypergeometric functions. Even with iteration the method is of a similar cost to Abdul-Razzak Ghan, since each iteration only requires the a simple linear interpolation of values from the lookup table.

13. Page 25915, line 10. The accuracy of the method is relative. The proposed approach may be better than other parameterizations only under highly idealized conditions. For other cases it is expected that parameterizations that account for lack of equilibrium and kinetic limitations would perform better.

As stated above, I agree that discussion of the accuracy of the scheme in relation to other schemes or the true solution is hard to demonstrate with a limited set of models and a limited input parameter space. Again it is hoped that the ongoing work of Partridge et al. will provide a comprehensive assessment of this and other schemes, and moreover highlight the importance of both numerical accuracy and physical complexity. I have removed accuracy from this particular statement. I have also removed the reference to accuracy from the abstract.

14. Page 25915, line 18. As mentioned above, the authors should comment on the idealized conditions used in the development of the new parameterization.

I have commented on the use of physical limitation in the final paragraph.

The underlying equation set on which the parametrization is built makes a number of physical assumptions, which are common to many well used activation parametrizations (Ghan et al., 2011). These assumptions and the resulting consequences are discussed, but the accuracy and computational efficiency of the resulting parametrization make it suitable for inclusion in both GCMs and high resolution models that use a modal representation of aerosol.

Fountoukis, C. and Nenes, A.: Continued development of a cloud droplet formation parameterization for global climate models, J. Geophys. Res., 110, D11212, 10.1029/2004JD005591, 2005.

Ghan, S. J., Abdul-Razzak, H., Nenes, A., Ming, Y., Liu, X., Ovchinnikov, M., Shipway, B., Meskhidze, N., Xu, J., and Shi, X.: Droplet nucleation: physically-based parameterizations and comparative evaluation, J. Adv. Model. Earth Syst., 3, M10001, 10.1029/2011MS000074, 2011.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleusactivity, Atmos. Chem. Phys., 7, 1961–1971, 10.5194/acp-7-1961-2007, 2007

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 25901, 2014.

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