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An investigation into the performance of three cloud droplet activation parameterisations

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Received: 16 January 2014 – Accepted: 27 January 2014 – Published: 10 February 2014

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Cloud droplet number concentration prediction is central to large scale weather and climate modelling. The benchmark cloud parcel model calculation of aerosol particle growth and activation, by diffusion of vapour to aerosol particles in a rising parcel of air experiencing adiabatic expansion, is too computationally expensive for use in large scale global models. Therefore the process of activation of aerosol particles into cloud droplets is parameterised with an aim to strike the optimum balance between numerical expense and accuracy. We present the first systematic evaluation of three cloud droplet activation parameterisations that are widely used in large-scale models. In all cases, it is found that there is a tendency to overestimate the fraction activated aerosol particles when the aerosol particle “median diameter” is large in a single lognormal mode simulations. This is due to an infinite “effective simulation time” of the parameterisations compared to a prescribed simulation time in the parcel model. In some cases when the “median diameter” is small in a single lognormal mode the fraction of activated drops is underestimated by the parameterisations. Secondly it is found that in dual-mode cases there is a systematic tendency towards underestimation of the fraction of activated drops, which is due the methods used by the parameterisations to approximate the maximum supersaturation with respect to water vapour.

1 Introduction

Clouds are important components in understanding climate change and therefore must be accurately represented in large-scale (regional and global) weather and climate models so that we can make realistic future climate predictions. The effective radiative forcing of aerosol and cloud interactions (including cloud albedo enhancement and cloud lifetime effect) have some of the largest uncertainties of all considered components of radiative forcing as reported in the IPCC 5th Annual Report (Myhre et al., 2013, page 123, Fig. 8.20). Aerosol particles interact with clouds by acting as nuclei

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on which water vapour can condense under liquid water supersaturated conditions. A change in the concentration of the subset of aerosol particles that act as cloud condensation nuclei (CCN) within a cloud will lead to a change in cloud droplet number concentration. The albedo of a cloud is dependent on the number concentration of cloud drops (Twomey, 1974) as is the cloud life-time (Albrecht, 1989). It is therefore key to understanding the role of clouds in climate that the activation of cloud droplets is well represented in numerical models.

The ability of a particle to act as a CCN is dependent on its size, composition and the ambient conditions – most notably the supersaturation (Pruppacher and Klett, 1997). Köhler theory determines the size to which an involatile deliquesced particle must grow in order to activate as a cloud drop, which is determined by the particle dry size, composition and the ambient supersaturation.

The presence of large (typically larger than 1000 nm dry size) aerosol particles may suppress the number concentration of activated drops. This is due to the fact that larger drops compete effectively for available water vapour such that they suppress the maximum supersaturation (S_{\max}), which results in fewer “smaller” particles activating (e.g. Sander, 1999). Some large particles do not grow quick enough to reach their critical diameter before peak supersaturation and therefore remain as unactivated particles, reducing the total fraction of activated drops. The large amount of water condensing in the growth of large particles leads to a suppression of S_{\max} .

Sectional cloud parcel models provide a physically realistic and internally consistent calculation of particle activation and droplet growth in a parcel of air undergoing adiabatic ascent. In this study we use the Aerosol-Cloud-Precipitation Interaction Model (ACPIM) (Connolly et al., 2012). ACPIM calculates both the sub-saturated growth of aerosol particles as well as their supersaturated growth by water vapour diffusion. This model makes very few simplifications of the condensation process giving confidence that the predictions are physically realistic. We acknowledge that the dynamical framework employed in this work does not allow reproduction of realistic atmospheric dynamics, nevertheless the initial formation period of clouds can often be assumed to be

adiabatic (Heymsfield et al., 1978). For a detailed description of processes represented in ACPIM, that are relevant to this paper, see the supplementary information of Topping et al. (2013).

Cloud parcel models such as ACPIM are too computationally expensive to be used in large scale global climate models. It is therefore necessary to rely on parameterisation schemes to estimate the number of activated cloud drops within large scale models. The most widely used parameterisation schemes fall into two families, those based on the work of Abdul-Razzak et al. (1998), Abdul-Razzak and Ghan (2000) and those following Fountoukis and Nenes (2005). The differences between these two sets of parameterisations are discussed in Sect. 2.1. A synopsis of these parameterisations is given in Connolly et al. (2013).

Ghan et al. (2011) provide an evaluation of two of the parameterisations evaluated in this study, Abdul-Razzak and Ghan (2000) and Fountoukis and Nenes (2005) with Barahona et al. (2010) extension. In this work we explore the performance of the parameterisations over a larger parameter space and run many more simulations. In general our results are similar to those of Ghan et al. (2011): Abdul-Razzak and Ghan (2000) consistently underestimate the fraction of activated drops in a dual-mode aerosol size distribution and Fountoukis and Nenes (2005), with Barahona et al. (2010) extension, underestimates the fraction of activated drops to a lesser extent than Abdul-Razzak and Ghan (2000), in simulations where the total number concentration of aerosol in a dual-mode size distribution is $> 2000 \text{ cm}^{-3}$. Ghan et al. (2011) make a suggestion for further work to provide a comparison of parameterisations against different numerical models such as the results presented here.

2 Method

ACPIM is a detailed bin-resolving cloud parcel model and is taken as the “ground-truth” to compare the parameterisation methods to. It is therefore used as the reference model for this study. The three widely used parameterisation schemes are evaluated for

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their ability to reproduce accurate values of the fraction of activated drops, for a large parameter space. We now explain the salient points of the parameterisations.

2.1 Description of parameterisations

Each of the parameterisation schemes used in this study can be used to represent the activation of single or multiple lognormal aerosol size distributions or “modes”. A lognormal distribution, describing the number of aerosol particles per natural logarithm of the bin width, $\frac{dN}{d\ln D_p}$, is described by the following equation:

$$\frac{dN}{d\ln D_p} = \frac{N_{\text{ap}}}{\ln \sigma \sqrt{2\pi}} \exp \left[-\frac{\ln^2 \left(\frac{D_p}{d_m} \right)}{2 \ln^2 \sigma} \right] \quad (1)$$

where N_{ap} is the total number concentration of aerosol particles, $\ln \sigma$ is the natural logarithm of the geometric standard deviation and d_m is the median diameter (Jacobson, 1999). The values for the median aerosol diameter of a lognormal mode, d_m , are given in Table 2 along with the breadth of the mode, $\ln \sigma$.

The first scheme, originally described in Abdul-Razzak et al. (1998), is further developed in Abdul-Razzak and Ghan (2000) to include multiple modes, hereafter referred to as ARG. The second scheme is Fountoukis and Nenes (2005), hereafter referred to as FN, and the third is an extension of FN that includes the effects of large (giant) CCN described in Barahona et al. (2010), hereafter referred to as FN GCCN.

The two parameterisations find approximate values for the maximum supersaturation achieved by a rising parcel of air, S_{max} , in different ways. FN sets the equation for the rate of change of supersaturation to zero and then iteratively finds a value for S_{max} that satisfies the equation. This is done by using a method called “population splitting” to divide the size distribution of CCN into two groups: one with only CCN that are close to their critical diameter and the other with CCN that are not (Fountoukis and Nenes, 2005). ARG also sets the equation for the rate of change of supersaturation to zero,

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then after neglecting the effects of curvature, gas kinetics and solute (in the equation for droplet radius growth rate) an approximate expression for S_{\max} is derived (Abdul-Razzak et al., 1998). Adjusting coefficients, calculated from numerical simulations, are applied to account for the errors made by simplifying the droplet growth rate (see Abdul-Razzak et al., 1998, for details).

In both parameterisations the number of CCN that activate given the maximum supersaturation is then calculated by applying Köhler theory to make a change of variable of S_{\max} to D_p , which is then used in conjunction with the prescribed lognormal size distributions to calculate the number of activated particles. The number of activated aerosol determined by both types of schemes, ARG and FN, is considered to be the number of aerosol with diameter greater than the smallest activated aerosol diameter (Abdul-Razzak et al., 1998; Fountoukis and Nenes, 2005).

Barahona et al. (2010), further develop the FN method to account for the fact that giant CCN – i.e. CCN with dry aerosol diameters greater than approximately 500 nm – may have insufficient time to grow to their activation size.

2.2 Model inputs

ACPIM allows the size distribution and any variation of composition of aerosol to be defined in addition to the particle mixing state. Updraft velocity is prescribed and the $\frac{dP}{dt}$ dependence is determined assuming an atmosphere in hydrostatic balance. Importantly, ACPIM is time dependent and the maximum simulated ascent time in the model is controlled. This ensures that the height that the parcel rises and hence the cloud depth for the simulation (for a given updraft velocity) is held within atmospherically reasonable bounds. The model assumes moist adiabatic ascent, no mixing with ambient air and outputs the supersaturation and the number distribution of activated and unactivated aerosol particles as a function of time. Only aerosol with diameters larger than their critical diameter are recorded as activated drops.

An evaluation of all of the mentioned parameterisation schemes against detailed numerical parcel models are reported in their respective studies. In this study we use

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a similar evaluation method to Barahona et al. (2010). However to avoid any bias in our results we use a Monte Carlo sampling technique to explore the parameter space over an atmospherically relevant range of conditions.

Barahona et al. (2010) use a very wide parameter space, where the number concentration of the two lognormal modes and the median aerosol diameter in the first mode are chosen from Seinfeld and Pandis (1998) to be atmospherically representative (page 2470 Barahona et al., 2010). The range of median aerosol diameters investigated in their study is chosen to represent all possible sizes of aerosol, from that of newly nucleated particles to giant CCN (Pruppacher and Klett, 1997, as described by).

Here we have chosen a large parameter space for the single-mode case similar to the one used by Barahona et al. (2010) to enable us to demonstrate the accuracy of the parameterisations under many conditions.

In the dual-mode case the parameter space is also similar to that used by Barahona et al. (2010). In the Supplement of this paper the parameter space used in dual-mode experiments has been reduced to avoid extreme concentrations of small and large particles (that are rarely found in the atmosphere) and a smaller range of updraft velocities so experiments only represent cloud depths more reasonably likely to exist. Such parameter space reduction reduces potential biases in the parameterisations that would be driven by unphysical parameter combinations.

The values in Table 1 are similar to the conditions used in the evaluations of ARG, FN and FN GCCN in their respective studies. Similarly the values in Table 2 were chosen to be within the same parameter space as was used to initially evaluate ARG, FN and FN GCCN. A value of 1 is used for the mass accommodation coefficient of water as accordance with the latest experimental evidence (Miles et al., 2012).

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3 Results

3.1 Demonstration of time dependency in model simulations

First, to explore and illustrate the time dependency in the growth of larger aerosol, ACPIM was run for two cases: small aerosol median diameter (100 nm, the “small aerosol” case) and large aerosol median diameter (1500 nm, the “large aerosol” case), both with total number concentrations of 500 cm^{-3} . The results from the parameterisations and ACPIM are shown in Fig. 1, for the same initial conditions.

In the “small aerosol” case the parameterisations reproduce the fraction of activated drops and maximum supersaturation well. However, in the “large aerosol” case, the parcel model does not reach the maximum possible supersaturation because it takes too long for the larger aerosol to reach the size required for activation. The result is that no aerosol activate in the simulated time of 2000 s for the large aerosol case. The results from the parameterisations effectively have no run time limit and therefore activate nearly all of the large aerosol. This would be equivalent to running ACPIM for an unrealistically long time such that the parcel of air reaches an unrealistic height before activating the large particles into cloud drops. We refer to this as an “infinite effective simulation time” artefact for the case of the parameterisations. Figure S1 shows that the actual amount of time the parcel model requires to activate the large aerosol is over 66 000 s. This equates to 19.8 km for the updraft velocity of 0.3 ms^{-1} that was used. Unperturbed ascent of this extent does not happen in the atmosphere; hence, the parameterisation appears to activate an unphysical fraction of the particles under these conditions (albeit with a population of unphysically large particles). Elimination of unrealistically high number concentrations of large particles from our simulations ensures that such obvious biases are not introduced in our evaluation, but the effect of overestimated activated fractions with unrealistic “effective simulated time” will still occur. This is a feature throughout the comparisons presented in the following sub-sections.

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(400 cm⁻³: 2400 cm⁻³). In many cases (mostly at updraft velocities above 2 m s⁻¹) the parameterisations activate all of the second mode and none of the first mode. At updraft velocities between 2 m s⁻¹ and 6 m s⁻¹ this results in an overestimation of the fraction of activated drops and at updraft velocities larger than 6 m s⁻¹ an underestimation. This is a feature of the time independent nature of the parameterisations, which can be demonstrated by increasing the run time of the parcel model from 2000 s to 8000 s – see Fig. S8, which shows that the feature at 0.16 fraction activated, present in Fig. 3, is less pronounced with a longer run time.

In cases where the fraction of activated drops is greater than 0.16 in the parcel model FN GCCN performs well with a generally small underestimation. ARG underestimates the fraction of activated drops significantly more than FN GCCN and the underestimation increases with updraft velocity: there is implicitly too much competition for water vapour in its formulation. This is the opposite effect to that exhibited by FN, implying too little competition in this scheme. The method for approximating the integral for S_{\max} in ARG is therefore too negative and in FN, too positive. FN GCCN corrects this with an additional term in the integral, (Barahona et al., 2010).

As expected ARG also underestimates the peak in RH in the majority of cases (see Fig. S6 that shows results of peak RH achieved in each simulation in the dual mode case), which also shows that FN GCCN performs best out of the two parameterisations at predicting the peak RH. Figure S6 also shows that the spread of peak RH values calculated by the parameterisations increases with updraft velocity, rather than a strong systematic offset.

The difference between FN GCCN and FN can clearly be seen in Fig. 3. Without the inclusion of effects of large aerosol in the parameterisation FN overestimates the number of activated drops and this overestimation increases with median diameter of second aerosol mode (see Fig. S7) and updraft velocity. Since the FN GCCN scheme shows a marked improvement when compared with FN in cases where large aerosol particles are present we have excluded the FN results in the comparison between the single and dual mode experiments below.

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3.4 Comparison between single- and dual-mode cases

Figure 4 shows the ratio of the number of activated drops calculated by the model to the number calculated by the parameterisations is most often close to unity for the monomodal cases. The majority of the rest of the data for the monomodal case are found below 1 showing a general overestimation by the parameterisations of the fraction of activated drops. All data in the first bin show where the parcel model does not activate any drops but the parameterisations do. The bimodal case shows a tendency to underestimate the number of activated drops using both ARG and FN GCCN, but with ARG clearly performing less well.

4 Conclusions

While the parameterisations evaluated in this paper perform well under a range of atmospherically relevant conditions, they also produce results that differ notably from the results of the parcel model under a wide range of conditions. Such conditions could provide the input distributions for the parameterisations when used within GCMs, producing significantly unphysical estimates of activated drop number concentrations.

The main conclusions from this study are as follows.

- First, there is a systematic tendency in the parameterisations towards overestimating the activated fraction of drops. This is a result of parcel models considering the time required for activation and cloud development, whilst the parameterisations implicitly allow an infinite “effective simulation time”.
- Second, the estimation of S_{\max} within the parameterisations leads to apparent opposite behaviour between the ARG and FN families of parameterisations. FN GCCN performs better with the additional term in the S_{\max} integral approximation. There is a small residual tendency towards underestimation of the fraction of activated drops, but with less of a low bias than ARG. The ARG parameterisation

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represents too much competition for water vapour resulting in an underestimation of the fraction of activated drops.

Hence, in a general sense, the parameterisations evaluated here tend to overestimate the number of activated drops in a single lognormal aerosol size distribution and under-
5 derestimate the number of activated drops in dual lognormal aerosol size distribution. The overestimation in the single-mode case is a result of an infinite “effective simulation time” in the parameterisations. The underestimation in the dual-mode case results from the methods used to approximate S_{\max} within the parameterisations.

It should be noted that the performance of the parameterisations is very dependent
10 on the parameter ranges chosen for the comparisons as illustrated throughout the Supplement for both single and dual mode simulations.

Supplementary material related to this article is available online at <http://www.geosci-model-dev-discuss.net/7/1317/2014/gmdd-7-1317-2014-supplement.pdf>.

15 *Acknowledgements.* This work was funded by the NERC ACID-PRUF programme, grant code NE/I020121/1, and a NERC studentship.

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**Table 1.** Initial conditions for the ACPIM simulations.

	ACPIM only		
Temperature	Pressure	Runtime	RH
290.15 K	950 hPa	2000 s	0.90

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Table 2. Parameter space investigated for both ACPIM and the parameterisations. Two evaluations were done: single mode and dual mode experiments.

Variable	1 Mode Experiment	2 Mode Experiment	
		MODE 1	MODE 2
Number conc. of aerosol	50–2000 cm ⁻³	2000 cm ⁻³	400 cm ⁻³
ln σ	0.2–0.8	0.46	0.46
Median Diameter	50–1000 nm	80 nm	5–5000 nm
Updraft Velocity	0.01–10 ms ⁻¹	0.01–10 ms ⁻¹	0.01–10 ms ⁻¹

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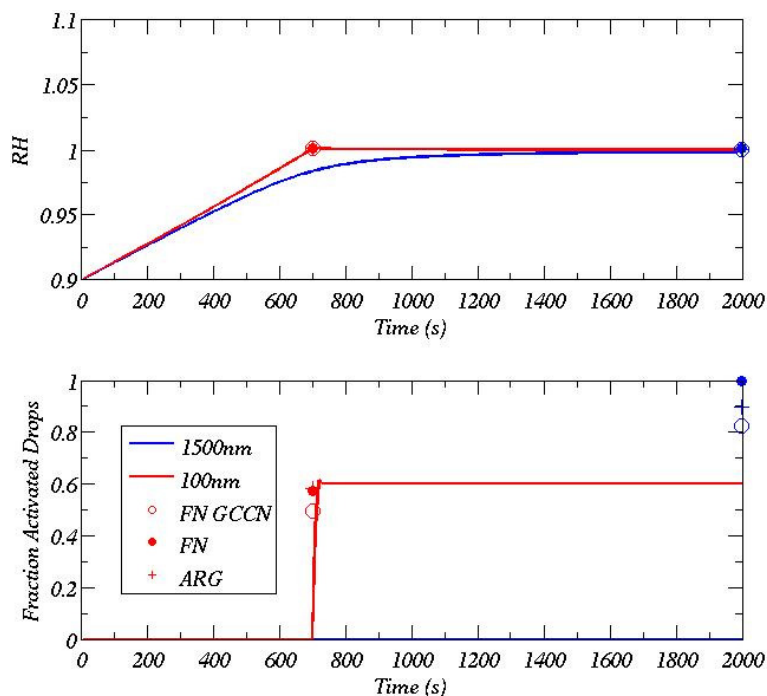


Fig. 1. Time series of RH (top panel) and fraction of activated drops (bottom panel) as calculated by ACPIM with initial conditions described in Table 1, a number concentration of aerosol 500 cm^{-3} and median aerosol diameters 100 nm (red) and 1500 nm (blue). Using the same initial conditions results from the parameterisations are plotted as single points at the time of maximum supersaturation (which is calculated from the parcel model).

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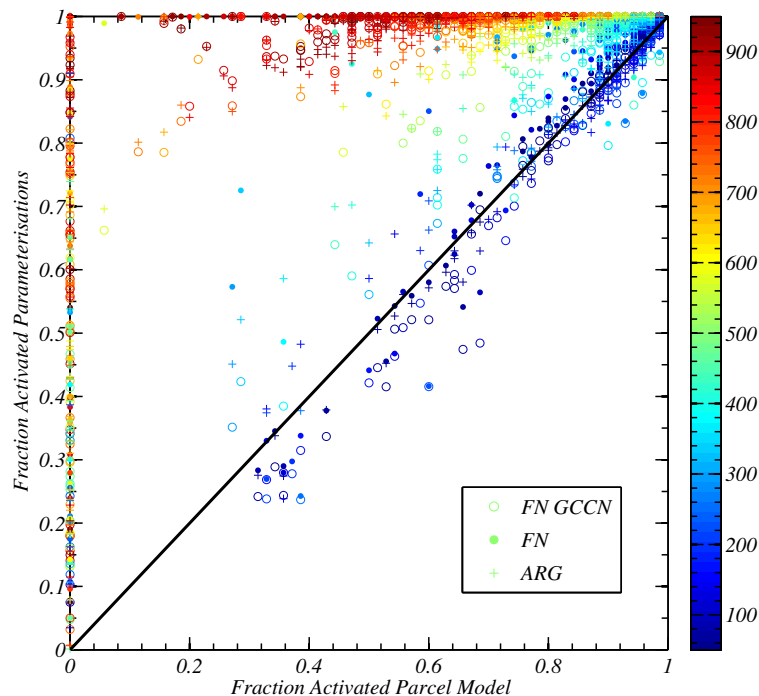


Fig. 2. Results from 1500 runs with 1 lognormal mode of ammonium sulphate aerosol with randomly sampled variable values as detailed in Table 2 and initial conditions described in Table 1. Symbols are coloured by median aerosol diameter (nm).

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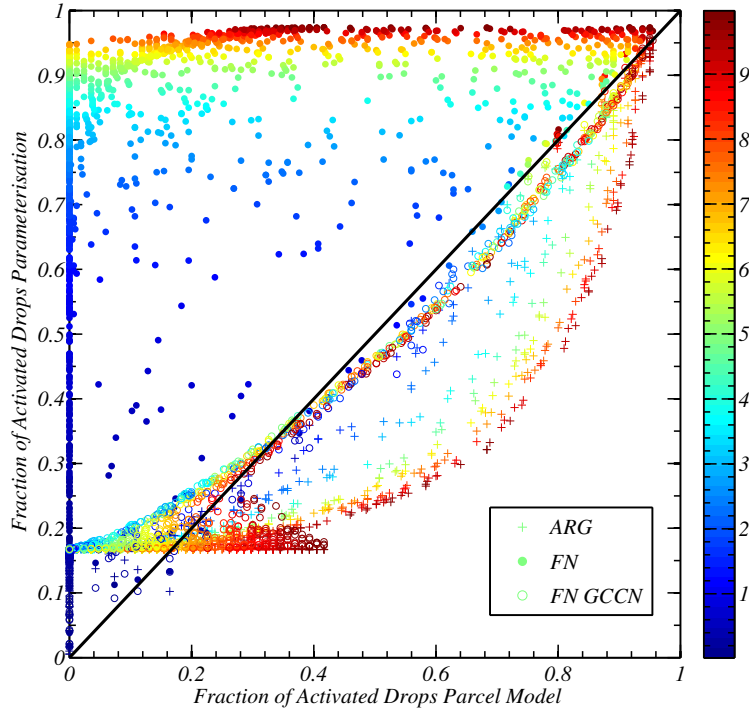


Fig. 3. Results from 1500 runs with a bimodal aerosol size distribution. Only the median diameter of aerosol in the 2nd mode and updraft velocity were changed between runs. Symbols are coloured by updraft velocity, m s^{-1} , see colour bar. For initial conditions and parameter ranges see Tables 1 and 2 respectively.

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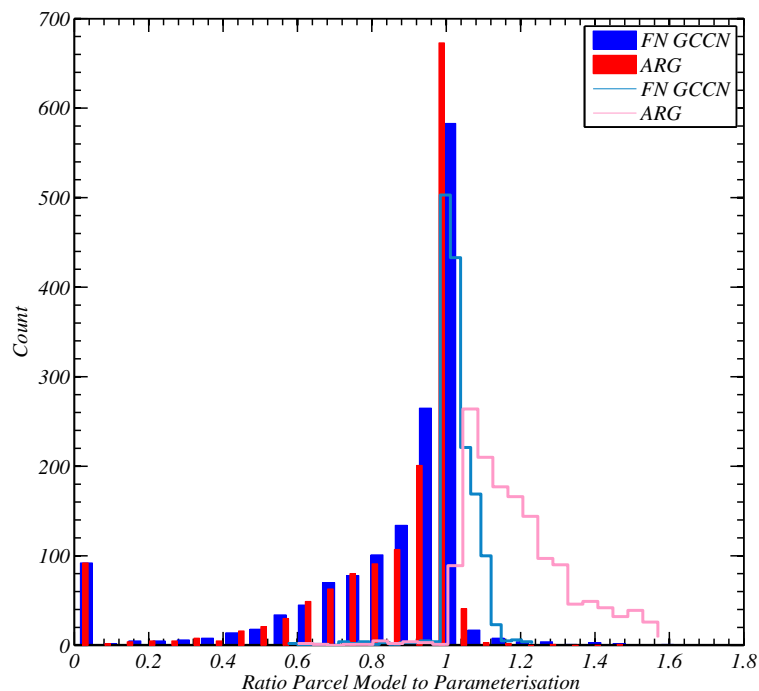
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Fig. 4. Histogram of the ratio of number of activated drops calculated by the parcel model to the number of activated drops calculated by FN GCCN and ARG parameterisations. Solid bars for monomodal runs and lines for bimodal runs.