

1 **Response to Referee #2:**

2
3 We are grateful for the careful review and instructive comments. We have revised the
4 paper carefully based on the reviewer's comments. A point-by-point reply to the
5 comments is described as follows (*italic text in blue color is from the reviewer*).

6
7 **Overall Appraisal:**

8 *This work develops a quasi-parcel model approximation to describe the activation of*
9 *aerosol into cloud droplets near the cloud base of warm stratocumulus. The authors*
10 *compile observations from several field campaigns around the world and use them to*
11 *investigate the performance of their model. These closure studies reveal a good*
12 *approximation of the parameterization to the observed cloud droplet number*
13 *concentration at cloud base. This work adds to the existing pool of droplet activation*
14 *parameterizations. The attempt of writing the parcel model equations on a*
15 *dimensionless basis could help future development.*

16 *The authors place less emphasis on trying to obtain a closed analytical solution and*
17 *rather use a semi-analytical integration. However, current models may be able to*
18 *handle the associated computational cost. On the other hand, the exposition of the*
19 *theoretical basis and rationale behind the authors approach is flimsy and, in some*
20 *cases, inaccurate. These should be clarified and corrected before the work could be*
21 *published.*

22 **Response:**

23 We thank the reviewer for the positive confirmation to the main goal of our work.

24 We agree that computing power has rapidly increased in recent years. Yet the
25 computational cost of GCMs is still quite substantial. According to Balaji et al. (2017,
26 doi: 10.5194/gmd-10-19-2017), the ASYPD, defined as the Actual Simulated Years
27 Per Day for the GCMs in a 24 h period on a given platform, of models in CMIP6
28 ranges from 0.04 to 25.2 (median 4.9) years. The faster CPU or parallel computation
29 helped to enhance the model efficiency, but the physical parameterizations (such as
30 schemes to solve the cloud and radiation processes) in the large-scale grid are still the
31 most time-consuming parts of the climate model. On the other hand, it is much more
32 expensive for using a parcel model (currently the most accurate tool to solve the
33 activation process) than a parameterization scheme. The computing time of a parcel
34 model to obtain the S_{max} is about several minutes, but using the QDGE scheme
35 costs only about 0.1 seconds, and other physically-based parameterizations (such as
36 the four state-of-the-art schemes used in Ghan et al., 2011,
37 doi:10.1029/2011MS000074) would take even less time. For this reason, the parcel

38 model is not practical for applying in long-term (decades or centuries) global
39 simulations (Ghan et al., 2011). Therefore, it is still necessary to develop
40 parameterized schemes to solve the aerosol activation in GCMs at present.

41 In addition, we elaborated our descriptions on the QDGE scheme in more detail in
42 Sect. 2.1. and included the comparison between results from the QDGE scheme and a
43 parcel model (Sect. 2.2 of the revised paper), to better explain the rationale behind the
44 QDGE scheme.

45

46 **General Comments:**

47 *a) My main concern in this work is the lack of rationale behind the proposed*
48 *approach. There is very little discussion regarding the approximations taken or*
49 *the validity of the assumptions. Although an acceptable closure is achieved*
50 *against observations, this does not guarantee that the approach is theoretically*
51 *sound. Particularly as the evaluation of the scheme seems tightly constrained by*
52 *observations.*

53 **Response:**

54 Thanks for the constructive suggestion. We agree that the closure with
55 observational cloud cases cannot be regarded as evidence for the theoretical
56 rationality of the QDGE scheme. We revised Sect. 2 to improve the
57 methodological description of the QDGE scheme.

58 We included more details about the fundamental rationale of the QDGE scheme
59 in Sect. 2.1, explaining each approximation or assumption we have made. A
60 schematic diagram (Fig. 1 in the revised paper) is added to show the major steps
61 of the QDGE scheme. A flow chart (Fig. 2 in the revised paper) is added to
62 describe the iterative calculation to solve supersaturation (S) in each sub-level in
63 the QDGE scheme.

64 We also compare the results from the QDGE scheme and a parcel model
65 (following the experimental setup in Ghan et al. (2011)) to verify the performance
66 of the QDGE scheme (Fig. 3 in the revised manuscript). The theoretical
67 rationality and practical advantage (for future high-resolution GCM) of the
68 QDGE scheme are summarized at the end of Sect. 2.2.

69

70 *b) The assumption of a constant saturation ratio, even over a short time step, is*
71 *unfounded. S changes over a very short time scale and it is not likely that it would*
72 *ever remain constant. Did the authors perform a timescale analysis to show*
73 *under what conditions their approximation would be acceptable?*

74 **Response:**

75 We are sorry for the misleading description in the previous version of the
76 manuscript. We assumed that S was constant locally (that is, within a sub-level
77 with a typical height of 1~10 m) but varied with time/height throughout the host
78 grid of GCMs. We clarify the assumption (Sect. 2.1) and show the S of each
79 sub-level (i.e. S_i in Fig. 1b, where $i = 1, \dots, N_{sub}$) in a schematic diagram (Fig.
80 1 in the revised paper).

81 In large-scale stratus clouds, the maximum supersaturation (usually less than
82 0.2 %) in the cloud appears about 100m above the cloud base, that is, the rate of S
83 change is $0.002 \% \text{ m}^{-1}$ or so (Pandis et al. 1990). According to this
84 characteristic of aerosol activation, we assume that the supersaturation is
85 approximately constant in the sub-grid scale (1~10m) for the QDGE scheme. We
86 added the description in the revised paper (Lines 112-116, Sect. 2.1).

87

88 *c) It is also not clear that this model can be called quasi-steady state since the*
89 *environment and the droplet sizes are clearly changing, and none of their*
90 *derivatives is negligible. What are the rigorous expressions from where the*
91 *parameterization is derived?*

92 **Response:**

93 The quasi-steady state refers to the following two assumptions in each sub-level:
94 1) the constant environmental supersaturation; 2) the conservation of total water
95 mass mixing ratio and liquid water static energy. In the revised paper, Eq. (1) is
96 the rigorous expression and Eq. (4) is the numerical expression for the particle
97 size growth.

98 The rationality of assumption 1) has been explained in our answer comment b)
99 above. For assumption 2), we assume that the air parcel ascends adiabatically in
100 each sub-level, which is the same as the assumption of the parcel model.
101 Correspondingly, the total water mass mixing ratio and liquid water static energy
102 are conservative.

103

104 *d) The proposed model resembles a Euler integration of the regular parcel model*
105 *where the differential equation describing the evolution of supersaturation was*
106 *replaced by an iteration over an algebraic expression. The authors should*
107 *explain the rationale behind such approach and compare it against a more*
108 *rigorous model where the evolution of the supersaturation is computed explicitly*
109 *using a differential equation.*

110 **Response:**

111 Yes, it is a good suggestion. We added more detailed explanations on the iterative

112 calculation in Sect. 2.1 of the revised paper. The Euler method was used to obtain
113 S along sub-levels for approaching an S profile (as shown in Fig. 1b and 1c of the
114 revised paper). While the iteration is to calculate the S value in each sub-levels.
115 We included more details about the fundamental rationale of the QDGE scheme
116 in Sect. 2.1, explaining each approximation or assumption we have made.

117

118 *Specific Comments:*

119 1) *Line 27. "in affecting" does not sound correct. Better say "determining"*

120 Have corrected (Line 29 in the revised paper, similarly hereinafter).

121

122 2) *Lines 52-53. This is a confusing sentence. Please clarify.*

123 Have rewritten (Lines 53-54).

124

125 3) *Lines 58-63. This is misleading and inaccurate. Most theoretical*
126 *parameterizations are approximate solutions to the parcel model equations.*
127 *Hence they must be evaluated against the rigorous solution first. Then, they can*
128 *be evaluated against observations. These are not "alternatives". Both*
129 *approaches aim to elucidate a different aspect of the parameterization accuracy.*

130 Yes, we agree that the evaluation by comparing against the rigorous solution
131 (such as the parcel model) is necessary before the validation against observations,
132 thus we changed "Alternatively" to "However" (Line 62).

133

134 4) *Line 70. Is the closure experiment the same as the evaluation? Please rephrase.*

135 The repeated part has been removed (Line 72).

136

137 5) *Line 75. Remove "that are"*

138 Have corrected (Line 78).

139

140 6) *Line 80. Aerosol is plural already.*

141 Have corrected (Line 83).

142

143 7) *Lines 82-84. This is an awkward sentence. Please rephrase.*

144 Have rewritten (Lines 85-86).

145

146 8) *All equations. Please choose either supersaturation or saturation ratio, but not*
147 *both. Changing between s and S makes things very confusing.*

148 Have corrected. We use S to represent supersaturation uniformly (Lines 88-89).

149

150 9) *Line 89. Sp is the droplet equilibrium saturation ratio.*

151 Have corrected (Line 91).

152

153 10) *Line 92. Rephrase. "The parameters A, B and C account for ... , given by,"*

154 Have rewritten (Appendix A).

155

156 11) *Line 104. Please clarify what water content means in this context.*

157 Here the "water content" means "aerosol water contents", that is the amount of
158 water vapor uptaken by hygroscopic growth of aerosol particles, defined as the
159 ratio of the wet aerosol volume to the dry one. We added the explanation in
160 Appendix A.

161

162 12) *Line 108. Different from what? Also why would this be important near water
163 saturation, when the droplet activates?*

164 We now move this part to Appendix A.

165 κ is a parameter introduced by Petters and Kreidenweis (2007) to represent the
166 hygroscopicity of aerosol with a variety of chemical compounds. Whenever the
167 chemical composition of aerosol is determined, the value of κ can be determined.
168 However, Petters and Kreidenweis (2007) and Kreidenweis et al. (2008) found
169 that the calculated aerosol water content (the ratio of the wet aerosol volume to
170 the dry one) based on κ biased at low relative humidity for some compounds.
171 Therefore, the QDGE scheme accounts for the variations in κ with relative
172 humidity to avoid the possible biases at low relative humidity in calculating the
173 growth of aeroso particle.

174

175 13) *Line 112. The system is missing equations describing the evolution of the
176 saturation ratio, the temperature, and the droplet size distribution. So direct
177 numerical solution would not be only expensive but impossible.*

178 We added Eq. (3) in Sect. 2.1 of the revised paper to describe the variation of
179 environmental S . In the QDGE scheme, we calculated the variation of
180 supersaturation with time/height by dividing the vertical grid of the host model
181 (large scale climate model) into sub-levels, producing a supersaturation profile in
182 the grid. More details of the major steps of the QDGE scheme are shown in Fig. 1
183 of the revised paper. The supersaturation in each sub-level was iteratively
184 calculated based on temperature and total water mass (integration over the
185 activated particle size distribution). Fig 2 in the revised paper shows a flow chart

186 of the iterative calculation for the supersaturation in each sub-level.

187

188 14) *Line 114. I am not sure what the “non-linear behavior of the water vapor*
189 *saturation ratio vertical profile” means.*

190 That means supersaturation S is non-linear varied with height, as schematically
191 plotted in Fig 1c in the revised paper. We modified the sentence accordingly
192 (Line 106).

193

194 15) *Line 116. This is contradictory to the previous statement. If S can be assumed*
195 *constant, how then is it that time steps much smaller than 1 s are needed?*
196 *Supersaturation is relaxed quickly in cloudy parcels, so this would be wrong. The*
197 *authors should add more explanation and justification to their assumptions. As it*
198 *stands it seems very ad-hoc and possibly incorrect.*

199 We largely modified Sect. 2.1 and 2.2 to clarify the assumptions for the QDGE
200 scheme. The constant supersaturation was assumed in each sub-level (typically 1
201 to 10 m in height) of the host model grid. An iterative calculation was conducted
202 in each sub-level to obtain the supersaturation. Finally, a vertical profile of
203 supersaturation was produced to represent the variation of S with height in the
204 host model grid. Figs. 1 and 2 in the revised paper show the major steps and the
205 iterative calculation in more detail.

206

207 16) *Line 156. What are the advantages of this calculation over writing a differential*
208 *equation for S ?*

209 A key for solving the differential equation for S (Eq. 3) is to determine dq_w/dt
210 by integrating wet particle size distribution calculated by Eq. (1). Whereas,
211 solving Eq. (1) needs the solutions of Eq. (2) and (3). Therefore, there is no
212 analytical solution at present for the differential equation for S .

213 Our iterative calculation is trying to use a numerical method to solve this issue
214 and makes the S in each sub-level available. We have tested that the iterative
215 method can converge to the desired value quickly, so it is efficient (Fig. 2).

216

217 17) *Line 164. Where exactly can you set the entrainment rate?*

218 The entrainment is considered to have a direct impact on the total water mass
219 mixing ratio r_t and the liquid water static energy h , as shown in Eq. (13) and (14)
220 in the revised paper. Both the total water mass mixing ratio and the liquid water
221 static energy are used to calculate the sub-level supersaturation (Fig. 2 and Eqs.
222 8-12 in Sect. 2.1 of the revised paper).

223

224 *18) Line 166. Couldn't find any mention of this scheme in those papers.*

225 Since there was no paper describing the QDGE scheme before, we could not
226 directly mention QDGE in the Arctic research. The description "A numerically
227 efficient solution of the condensational droplet growth equation" in Mahmood et
228 al. (2019) stands for the QDGE scheme. But there is no description in Arora et al.
229 (2015). Thus, we have removed this sentence in the revised paper.

230

231 *19) Figure 2. Is the observed LWC used to drive the model?*

232 Yes. LWC is converted to q_w for calculating the initial total water mass mixing
233 ratio r_t and liquid water static energy h (Fig. 2 and Eqs. 8-11 in the revised
234 paper), which are used to calculate S_i in the sub-level (Fig. 1b in the revised
235 paper).

236

237 *20) Line 262. How does this compare against integrating over the full aerosol size*
238 *distribution?*

239 As described in Lines 318-318 we weighed the total fitted aerosol number
240 concentration by the observed aerosol number to ensure the conservation of total
241 number concentration (i.e., the total N_a integrated over the QDGE sections in
242 Fig. 6c is the same as the aerosol number integrated over the observed PSD in Fig.
243 6a).

244

245 *21) Line 276. Internally mixed aerosol is defined as a population where all particles*
246 *with the same size have the same composition. Please correct.*

247 Have corrected (Line 328).

248

249 *22) Line 310. Please explain where this comes from. W_{sub} and W_+ represent similar*
250 *things. That is, each parcel moves with a given vertical velocity. A rigorous*
251 *approach would integrate the parameterization over the distribution of W . In*
252 *absence of that, a mean (in the sense of the mean value theorem) could be used.*
253 *That would be either W_+ or W_{sub} , but not both.*

254 As illustrated by Ghan et al. (2011) (doi:10.1029/2011MS000074), updrafts are
255 not adequately resolved in global models, so subgrid variations in updraft
256 velocity must be taken into account. Most climate models (e.g. Lohmann et al.,
257 2007; Ming et al., 2007; Gettelman et al., 2008; Wang and Penner, 2009) often
258 represent the grid updraft velocity using the sum of the large-scale grid-mean
259 updraft velocity (w_+) and the subgrid variation in updraft velocity (w_{sub}) within

260 the grid cell (See Ghan et al. (2011) P16 for more details). Here we use a similar
261 approach, w_+ and w_{sub} are obtained from the average and the standard
262 deviation of the probability density of function (PDF) of the sampled vertical
263 velocity from aircraft measurement on clouds (Sect. 3.2.3), as derived in Peng et
264 al. (2005) and Meskhidze et al. (2005). Therefore we regarded w_+ and w_{sub} as
265 the correspondences to the large-scale grid mean and the subgrid variation of the
266 updraft velocity.

267

268 *23) Line 322. This sounds awkward. Maybe use, “using Eq.(21) into Eq. (20) we*
269 *obtain”*

270 Have rewritten (Line 374).

271

272 *24) Line 334. Awkward sentence. Maybe just say TKE is given by...*

273 Have rewritten (Line 386).

274

275 *25) Line 382. Please explicitly define $CDNC_M$ and $CDNC_O$*

276 We have explained the $CDNC_M$ in more detail and explicitly defined $CDNC_O$ in
277 Sect. 3.3.

278

279 *26) Line 392. Is R^2 this the Pearson correlation coefficient?*

280 R^2 is the square of the Pearson correlation coefficient in our research. We
281 modified the sentence to clarify it (Line 447).

282

283 *27) Line 418. This agreement is somehow unexpected. Given the assumptions made,*
284 *my suspicion is the observed LWC is used to drive the parameterization which*
285 *along with the total aerosol number provides a strong constraint to CDNC.*
286 *Please clarify whether this is the case.*

287 In the closure experiment, LWC is used to calculate the initial r_t (total water
288 mass mixing ratio) and h (liquid water static energy) by converting LWC to
289 q_w (Fig. 2 and Eqs. 8-12 in the revised paper), which is used to calculate S in
290 the sub-level (Fig. 1b). However, LWC has no direct impact on N_{CCN} (Fig. 1e).
291 Therefore, the decent performance of the QDGE scheme in the closure
292 experiment is not determined by using the input LWC from observation.

293

294 *28) Line 450. As written, Eq. (1), i.e., the droplet growth equation, does not imply this.*
295 *The supersaturation balance is missing.*

296 This is our fault, the sentence “This is consistent with the droplet growth equation”

297 should be “This is consistent with the change of environmental supersaturation
298 (Eq. (3))”. We have corrected it (Line 504).

299

300 29) *Line 476. How efficient? It would be appropriate to include some timing*
301 *benchmarks (against rigorous solutions or other commonly used*
302 *parameterizations) to assess the applicability of the scheme in large scale*
303 *atmospheric models.*

304 Yes, thanks for the good suggestion. We added some descriptions about the time
305 consumption for the QDGE scheme and the parcel model in Sect.2.2 in the
306 revised paper (Line 210). The time of a parcel model to obtain the S_{max} for a
307 cloud case is several minutes, but it is only about 0.1 seconds for the QDGE
308 scheme. We also added a comparison between the results of the QDGE scheme
309 and a parcel model for different aerosol and environmental conditions (Fig. 3 and
310 Sect. 2.2 in the revised paper), it confirmed the good performance and acceptable
311 accuracy of the QDGE scheme.

312