

1 **Response to Reviewer 1 to the paper : Theoretical investigation of mixing in warm clouds –**  
2 **Part 2: Homogeneous mixing authored by M. Pinsky, A. Khain, A. Korolev, and L.**  
3 **Magaritz-Ronen**  
4

5 **Authors appreciate the Reviewer's time and efforts to review our manuscript.**

6 General comments to all three parts (repeated in all three reviews). I read the papers with  
7 considerable interest mostly because this seemed to be a popular topic some time ago, in both  
8 observations and modeling. I was curious to see what new these manuscripts bring. Frankly, I  
9 was disappointed. First, the analysis concerns a highly idealized problem, with little applications  
10 to real clouds. Turbulent mixing in clouds is by far more complicated than situations depicted in  
11 Fig. 1 of part 1 (and then repeated in different shapes as Figs. 1 in Part 2 and 3). Second, I am  
12 aware of a study in which the authors developed a fairly sophisticated model of microphysical  
13 evolution during turbulent stirring (Jarecka et al., JAS 2013) aiming at prediction of the  
14 homogeneity of mixing. They applied the model to LES simulations of shallow convective cloud  
15 field. The impact was surprisingly small and the authors of that paper argued why this might be  
16 so (the entrained air comes from the descending shell and is not far from saturation). So in a  
17 sense the subject is "old news". Finally, the lengthy discussions, full of unnecessary caveats  
18 and references to details of small multi-panel figures, made the reading frustrating. All three  
19 parts read like a student dissertation, not a concise scientific paper highlighting key points and  
20 leaving the rest for the reader to follow. Thus, I read the manuscripts with decreasing interest,  
21 and my comments are more detailed for the part 1, and get more general for parts 2 and 3.

22  
23 Overall, I do not believe that the subject matter deserves close to 100 pages and close to 50  
24 figures. I feel that the material deserves a single, short and concise manuscript, with new  
25 material clearly separated from what I feel has been discussed in the past, perhaps not at such  
26 a level of detail. Reading introductions to all three parts made me mad, because all three say  
27 basically the same thing with different language and organization. Part 1 is mostly trivial in my  
28 view, with some parts speculative and other repeating already published material (see detailed  
29 comments). Parts 2 and 3 have some aspects that perhaps deserve to be published, but it is not  
30 clear to me how useful these are (not very much in my opinion). References to aircraft  
31 observations are vague and missing the key aspect, which is the irrelevance of an idealized  
32 problem considered by the authors to low-spatial resolution observations of a complicated  
33 multiscale natural system.

34  
35 **® The overview sections, which were copied and pasted for all three different reviews, can  
36 be summarized by the following claims:**

37 **a) The problem of turbulent mixing in clouds "seemed to be a popular topic some time  
38 ago", but now "the subject is old news".**

39 **b) This study addresses a "highly idealized problem" and uses simplified models in order to  
40 describe cloud mixing.**

c) The results presented in the papers are not new and are “repeating already published material”.

The authors strongly disagree with the above statements of Referee 1.

In response to the first claim: the mechanism of mixing is still not well understood and continues to be a highly relevant problem in the cloud physics community, especially given the high rate of recent publications on this topic. We believe that the three papers contribute significantly to the theory of interaction of cloud droplets with turbulent environment and present novel techniques of investigating the effect of mixing both from a theoretical standpoint and through in-situ observations.

Second, in contrast to the reviewer, we support the common practice of using idealized models of complex cloud processes, in order to investigate physical mechanisms without being bogged down by the multitude of other processes involved. Idealized considerations (e.g. adiabatic assumptions) are widely used in cloud physics as well as in physics in general. The assumptions are clearly articulated at the beginning of each paper in order to let a reader judge about the level of idealization of the utilized approaches.

Third, as regards to novelty, the following new results have been obtained:

a) The first paper suggests a new technique for identifying type of mixing (homogeneous or inhomogeneous) based of the analysis of the moments of droplet size distributions. It was shown that homogeneous mixing breaks functional relationships between the moments. Nothing like that has been done before. A novel approach for identifying mixing from in-situ observations was proposed. The comments obtained by the authors from their colleagues showed that the proposed technique start to be utilized by other research groups.

b) The second paper considers *homogeneous* mixing. One of the important finding of this paper is an analytical universal solution describing the rate of evolution microphysical parameters as well as the final equilibrium state (mixing diagram). It is shown that in case of polydisperse droplet size distributions evolution of droplet spectra can lead to increase in characteristic size of droplets in contrast to widely accepted "classical" view, when the characteristic droplet size is decreasing. It was shown that evaporation time can be expressed in terms of time of phase relaxation. This is important for definition of reaction time in Damkoller number.

c) The third paper is dedicated to *inhomogeneous* mixing. A theoretical framework for a time dependent mixing of two volumes that accompanies by cloud droplet evaporation is developed. A new turbulence-evaporation model of time evolution of ensemble of droplets under different environmental parameters is proposed. In contrast to previous studies the Damkoller number is introduced as a result of re-normalization of mixing-evaporation equation, rather than empirically. It is shown that any mixing leads to droplet spectrum broadening. For

the first time the scientifically grounded demarcation between homogeneous and inhomogeneous mixing in the space of environmental parameters is performed.

The authors regret that Referee 1 overlooked all these novelties.

The authors also believe it is impossible to follow the recommendation of Referee 1, to combine all papers into one single, summary paper. While the papers all consider the same subject, they perform completely different functions with regard to investigating the issues of mixing.

35

36 General comment. Part 2 provides detailed analysis of the temporal evolution of the  
37 thermodynamic homogenization after the mechanical mixing quickly homogenizes the initially  
38 separated cloudy and cloud-free (i.e., sub-saturated) volumes. I feel Part 2 presents some relevant  
39 results, but the paper is way to long for the outcomes it provides. I hope my comments below  
40 will help the authors to convert this part into a section of the rewritten  
41 new manuscript.

42 (R) We appreciate this comment of Reviewer. However, we do not see the possibility to  
43 compress all parts of the study into one paper. The topics discussed in these parts as well as the  
44 approaches used are too different to combine them into one paper.

45 (C) 1. Towards the end of the introduction, the paper (once again) introduces the relevant  
46 time scales. What I find interesting and worth pursuing in my view is that the time scale  
47 describing droplet evaporation, taken as the phase relaxation time scale in the current  
48 study, is not the only possibility. Others (including the Jarecka et al. paper I think) have  
49 taken this time scale as the time required to evaporate a droplet with the sub-saturation  
50 of the cloud-free volume. I think some studies considered both and took the smaller  
51 (or the larger?) of the two (Feingold?). Note that the phase relaxation time scale has  
52 no information about the sub-saturation of the initially cloud-free volume, but the other  
53 definition does. Is this relevant? It should be for the extremely inhomogeneous mixing  
54 I think (I will comment on that in my review of Part 3).

55

56 (R) We know that some authors indeed use evaporation time of individual drop under given sub-  
57 saturation as a characteristic time of mixing process. In our opinion only phase relaxation time  
58 should be used as a characteristic time scale of the process since we should consider behavior of  
59 a large amount of droplets. In the paper (Pt 2) is shown that the phase relaxation time is a natural

60 time scale of mixing problem. This is clearly seen from universal renormalized evaporation  
 61 equations, in which the phase relaxation time plays the role of time unit. Adiabatic consideration  
 62 of the total volume in which mixing takes place leads to strict relationships between the changes  
 63 of supersaturation (or sub-saturation) and liquid water content in the volume, that makes  
 64 absolutely impossible to consider the changes in the size of individual droplet and in  
 65 supersaturation independently. These two quantities (supersaturation and liquid water content)  
 66 are tightly linked.

67 Note that supersaturation (or sub-saturation) is not in the list of parameters that determine phase  
 68 relaxation time. Thus, the consideration of evaporation of individual droplet (under unchanged  
 69 supersaturation) is not physically grounded and cannot be a time scale related to the mixing  
 70 process. This comment is added to conclusion of Pt2.

71

72 2. Top of p. 30274. The Damkoehler number was introduced much earlier that  
 73 Lehmann (2009), not using the name. Old paper by Latham, Baker and others should  
 74 be cited here.

75 (R) Done

76

77 3. P. 30275. I think the fact that S can be taken as a linear combination of the supersaturations  
 78 between the two volumes after the end of the mechanical mixing is an  
 79 interesting observation. However, the small temperature difference assumption may be  
 80 valid for the cumulus, but it is likely invalid for the subtropical stratocumulus. The inversion  
 81 jump there is typically around 10K and the very top of the cloud may be exposed  
 82 to several degree temperature differences between cloudy and cloud-free air.

83

84 (R) Very high temperature jumps like 10C above cloud top of stratocumulus cloud indicate,  
 85 supposedly, the absence of intense turbulent mixing between cloud and dry air. In many cases  
 86 temperature jumps is a few degrees C (H. Riehl). Penetration of clouds from BL to the inversion  
 87 layer and their evaporation within this layer indicates the existence of strong interaction between  
 88 Sc and the inversion layer.

89 A comment is included: At temperature differences of 5-10°C, the deviation from the analytical  
 90 solution (6) increases, which requires using more precise formulas for supersaturation (see Pt. 1).

91 4. Section 3 should be compressed. Section 3.1 shows formulas already introduced and used  
 92 earlier in the paper (Part 1).

93 Formulas presented in Appendix A of Pt 1 which are the same as used in 3.1 of Pt 2 are deleted  
94 (since they are not used in Pt 1).

95

96 (C) Section 3.2 should be combined with 3.3 focusing on the key outcome (shown in Fig. 3).  
97 Section 3.3 provides a universal (i.e., using nondimensional parameters) analysis of the problem.  
98 As much as it is interesting, it is not very useful in my opinion. My suggestion is to compress it  
99 into an appendix in the rewritten paper.

100 (R) We disagree with Reviewer. We believe that obtaining of universal dependencies is one of  
101 the most important results of the study. This universal solution clearly shows the comparative  
102 role of different microphysical and thermodynamical parameters. This role is hardly possible to  
103 get from numerical studies. Note that Formula (6b) and Fig. 6b show actually the analytical  
104 expression for widely used mixing diagrams. Thus, we cannot shorten this section or move it to  
105 an appendix.

106

107 (C) The conclusion at the end of section 3.3 that the time scale is the phase relaxation time scale  
108 is trivial. Is the phase relaxation time scale about a response of the system to the supersaturation  
109 perturbation? The end of the mechanical mixing is exactly such a perturbation. So what is new  
110 here?

111 (R) As was mentioned in our response to Comment C1 of Reviewer, we wanted to stress that the  
112 phase relaxation time is a natural time scale of mixing problem in contrast to other time scales  
113 considered in literature.

114

115 (C) Figs. 3 and 4 and their discussion are not needed. Of course the time scale based on the  
116 initial radius changes as droplets evaporate and the mean radius decreases.

117

118 (R) Figs. 3 and 4 (and corresponding discussions) show:

119 a) time dependencies of microphysical characteristics during process of homogeneous  
120 evaporation under different initial parameters. Note that in standard mixing diagrams only final  
121 equilibrium states are considered. The figures show that the droplet relaxation time is the  
122 characteristic time scale.

123 b) the figures clearly demonstrate the existence of two different possible final regimes which  
124 depend in the initial relative humidity (or on universal non-dimensional parameter). These

125 regimes are: total droplet evaporation and termination of evaporation by reaching saturation  
 126 state; curves separating these two regimes are also presented.

127 c) it is shown that the theory developed in the study agrees well with the numerical solution of  
 128 more complicated non-linear equation for supersaturation.

129 Thus, these figures hardly can be excluded.

130

131

132 (C)5. Section 4 considers a polydisperse case. As before, I did not attempt to follow the  
 133 derivations, but as I stated before I consider this problem trivial: you know how much water has  
 134 to be left at the end (i.e., after the parcel is brought to saturation), so the only problem is how  
 135 much shift of the spectrum is needed. I am not sure if obtaining detailed formulas for the  
 136 evolution of various quantities is of significance in this highly idealized problem.

137

138 (R) This part illustrate non-trivial features of homogeneous mixing in case of initial polydisperse  
 139 DSD. It is shown that (in contrast to general opinion)

140 a) Homogeneous mixing can be accompanied by a decrease in droplet concentration;

141 b) Homogeneous mixing may lead to an *increase* in the mean volume and an increase in the  
 142 effective radii. The increase or decrease in the effective radii depend on the characteristic of  
 143 initial DSD.

144 Since in real clouds DSDs are polydisperse, the widely accepted believes that homogeneous  
 145 mixing always decreases effective radius keeping droplet concentration unchanged is wrong.

146 These conclusions are main ones of the Pt2.

147 We would like to stress that consideration of such "highly idealized problem" showed results  
 148 that were not recognized by investigators for many years. This study shows, therefore, that even  
 149 such simplified model has direct relation to real clouds in which DSDs are polydisperse and time  
 150 dependent.

151

152 6. Section 5 does not belong to this paper, not in this form. First, the authors talk about "parcels".  
 153 Which fluid flow model applies parcels in its formulation? I think the authors mean "grid  
 154 volumes", not "parcels". Second, finite-difference fluid flow models (with some exceptions)  
 155 typically assume that a grid volume is homogeneous. Any flux in and out (advection, turbulence,  
 156 etc) leads in instantaneous homogenization of such a grid volume after completion of the time  
 157 step. So obviously such a model does "homogeneous mixing". This is trivial. One can design a

158 model (or scheme) that includes (i.e., assumes) a subgrid-scale structure of model-predicted  
159 variables, but this is a different story. Overall, section 5 is perhaps a start for a new scheme  
160 development, but it does not belong to this paper.

161

162 (R) This is not so trivial as reviewer assumes. The mixing that is simulated in any model is  
163 inhomogeneous at resolvable scales (the text is clarified) and typically homogeneous at the  
164 subgrid scale. Since we consider the understanding of this point is actual for cloud community,  
165 we decided to keep this section as Discussion, because it is important for cloud modelers.

166

167 7. Conclusion section is again short. As I said before, I consider point 2 a trivial consequence  
168 of the bulk evaporation in case when small droplets in the initial distribution  
169 have to evaporate completely.

170

171 (R) We suppose that if we would follow all recommendations of Reviewer concerning  
172 shortening and deleting entire sections, the conclusions would be indeed short. We believe,  
173 however, that in our responses we managed to show that results obtained are new and not trivial.  
174 As regards to specific comment of Reviewer to point 2 of conclusions, we agree that indeed the  
175 result is natural and easy to understand. The problem, however, is that during many years  
176 investigators believe (see, for example, comments of reviewer 2) that homogeneous mixing  
177 always decreases drop size, mean volume radius, but keeps droplet concentration constant. This  
178 is a wrong believe, because in contrast to the classical concept, the DSDs in real clouds are  
179 polydisperse.

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188 **Response to comments and remarks of reviewer 2**

189 **We are grateful to Reviewer for this high evaluation of the study**

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192 **Comments on “Theoretical investigation of mixing in warm clouds – Part 2: Homogeneous**  
 193 **mixing”**

194

195 The significant contribution in this paper is the development of analytical results for the time  
 196 evolution of a droplet size distribution for both monodisperse and polydisperse droplets in the  
 197 limit of homogeneous mixing. The novelty in this, as far as I am aware, is that previous work has  
 198 focused on the final stage of the distribution, such as commonly portrayed in the mixing  
 199 diagram. Although it is idealized work, the non-dimensional equations and parameters in  
 200 Equations 16 to 19 provide nice insight into cloud response to mixing. In particular, the finding  
 201 that polydisperse droplet populations can show signatures typically interpreted as due to  
 202 inhomogeneous mixing is insightful.

203 **Ⓜ We are grateful to Reviewer for this high evaluation of the study.**

204

205 General criticisms:

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207 ©1. The introduction should provide better and clearer motivation for why a study of the time  
 208 evolution of the droplet size distribution is of interest. This would be more valuable than  
 209 additional discussion of general mixing concepts that already are covered in part 1.

210 **Ⓜ The introduction is improved and necessary comments are included.**

211

212 ©Also, as with part 1, the link between this paper and the other two parts needs to be clearly  
 213 defined. What are the assumptions and limitations, and what is the area of application for the  
 214 present work, especially relevant to the other two parts? With regards to the connection to other  
 215 work, is Figure 1 in this part 2 really necessary, given that a similar version already is at the  
 216 foundation of part 1 (part 1 Figure 1)?

217 **Ⓜ We did not introduce any additional assumptions as regards to homogeneous mixing. In**  
 218 **contrast to typically used basic concept of homogeneous mixing we consider a) time evolution of**  
 219 **droplet ensemble in a volume, where all droplets experience the same sub-saturation and b) we**  
 220 **also consider the case when initial DSDs are polydisperse. Corresponding comment is added into**



221 the introduction. We also increase the number of cross-references between different parts of the  
 222 paper.

223

224 ©Finally, it is absolutely necessary that the notation between the three papers should be uniform.  
 225 This currently adds a lot of confusion in trying to understand the results of part 2 in the context  
 226 of the results obtained in part 1.

227 ® Notations are corrected and brought into line.

228

229 ©2. Overall I would say that, while I view this as insightful work, there is a general need  
 230 throughout the paper to discuss more about the physical interpretation rather than just writing  
 231 down equations. For example, Eq. 23 is too complicated.

232 ® Comments to eq. 23 are given

233

234

235

236

237 Specific comments

238

239 ©1.  $S$  is supersaturation in Part 2 and 3, but it is saturation ratio in Part 1 and most textbooks.  
 240 Choose one symbol and be consistent in all parts.

241 ® The correction is made.  $S$  is supersaturation in all three parts

242

243 ©2. Liquid water content and liquid water mixing ratio are different. In the text,  $q$  is liquid water  
 244 mixing ratio, but the authors sometimes refer to it as LWC. For example, section 3.2 “Time  
 245 evolution of supersaturation and LWC”, section 4.3 “Evolution of LWC and supersaturation”.  
 246 The authors need to check the whole text and make it consistent.

247 ® Corrected. Note we prefer to write down formulas using liquid water mixing ratio (kg/kg), but  
 248 in the figures we show LWC ( $\text{g/m}^3$ ). This is why we use word LWC in the names of sections

249

250 ©3. Page 30279, line 14: missing space between “parameter” and “gamma”.

251 ® corrected

252

253 ©4. Equation 22 on page 30280: should have a “ $\sim$ ” over “ $t$ ” for “ $S(t)$ ”.

254

255 ® corrected

256

257 5. Page 30288, line 20-22: I don't understand "the concept of homogeneous mixing, according to  
258 which the mean and effective radii do not change." Please clarify.

259 ® The sentence is corrected. An increase in the value of the effective radius contradicts the  
260 concept of homogeneous mixing, according to which both the mean radii and the effective radii  
261 decrease in the course of mixing.

262

263 ©6. Appendix B: Equation number in the text (for example page 30298 Line 15) should be "BX"  
264 not "AX"

265 ® Corrected

266

267 ©7. Figure 3 caption: should be "LWC=0.29g/m<sup>3</sup>" not q

268 ® Corrected

269

270 ©8. Figure 6b: should be normalized liquid water mixing ratio not LWC

271 ® Corrected

272

273

274 ©9. Figure 7a: x axis should be 0, 5 10 15?

275 ® Corrected

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278 ©10. Figure 7 caption: also q should be LWC.

279 ® Corrected

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286 **Theoretical investigation of mixing in warm clouds. Part 2: Homogeneous**  
287 **mixing**

288

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310 **Abstract**

311 Evolution of monodisperse and polydisperse droplet size distributions (DSD) during  
312 homogeneous mixing is analyzed. Time-dependent universal analytical expressions for  
313 supersaturation and liquid water content are derived. For an initial monodisperse DSD, these  
314 quantities are shown to depend on a sole non-dimensional parameter. The evolution of moments  
315 and moment-related functions in the course of homogeneous evaporation of polydisperse DSD is  
316 analyzed using a parcel model.

317 It is shown that the classic conceptual scheme, according to which homogeneous mixing  
318 leads to a decrease in droplet mass at constant droplet concentration, is valid only in cases of  
319 monodisperse or initially very narrow polydisperse DSD. In cases of wide polydisperse DSD,  
320 mixing and successive evaporation lead to a decrease of both mass and concentration, so the  
321 characteristic droplet sizes remain nearly constant. As this feature is typically associated with  
322 inhomogeneous mixing, we conclude that in cases of an initially wide DSD at cloud top,  
323 homogeneous mixing is nearly indistinguishable from inhomogeneous mixing.

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327 Key words: *turbulent mixing, homogeneous mixing, mono-and polydisperse droplet size*  
328 *distributions*

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## 334 1. Introduction

335 Turbulent mixing at cloud edges and cloud tops accompanied by phase transitions has been  
 336 the focus of numerous studies, beginning with the pioneering works of Baker and Latham  
 337 (1979), Baker et al. (1980), Blyth et al. (1980) and Baker and Latham (1982). Laboratory  
 338 experiments by Latham and Reed (1977) showed that after mixing with sub-saturated air some  
 339 droplets completely evaporate while others remain unchanged. This finding gave rise to the  
 340 concept of two types of turbulent mixing: homogeneous and inhomogeneous. A recent  
 341 description of the classical concepts of homogeneous and inhomogeneous mixing can be found  
 342 in study by Korolev et al. (2016), hereafter referred to as Pt1.

343 **Figure 1** presents a conceptual scheme of homogeneous mixing between saturated cloud  
 344 volume  $V_1$  containing droplets and sub-saturated droplet-free volume  $V_2$  (Fig. 1a) in case of  
 345 initially monodisperse droplet size distribution (DSD). This scheme is the base of further  
 346 analysis. According to the concept of homogeneous mixing, the air within the volumes mixes at  
 347 a rate much higher than the characteristic rate of droplet evaporation. So, the fields of  
 348 temperature and humidity (and, therefore, the fields of the relative humidity and supersaturation)  
 349 are rapidly homogenized throughout the entire volume, and all the droplets experience the same  
 350 supersaturation (Fig. 1b). At the end of the first stage, the droplet concentration decreases due to  
 351 dilution down to  $N_{m0} = N_1 \frac{V_1}{V_1 + V_2}$ , where  $N_1$  is the initial droplet concentration in the cloud  
 352 volume. At the second stage (Fig. 1c), droplets change supersaturation and temperature through  
 353 their evaporation. There are two possible scenarios for the final equilibrium states. In the first  
 354 one, illustrated in Fig. 1c, droplets continue evaporating until they saturate the environment. The  
 355 size of all droplets decreases, but they evaporate only partially, so the droplet concentration  $N_{m0}$   
 356 remains unchanged. In the second scenario, when the initially droplet-free volume is very dry,

357 the droplets penetrated from cloud volume evaporate completely. In case of polydisperse initial  
358 DSD, both partial and complete evaporation of droplets determine the final DSD.

359 In contrast to homogeneous mixing, spatial homogenization during *inhomogeneous* mixing is  
360 a relatively slow process. According to the concept of extremely inhomogeneous mixing, some  
361 droplets are transported by the turbulent eddies into the dry environment and experience  
362 complete evaporation, whereas other droplets remain unchanged. As in the case of homogeneous  
363 mixing, the process of droplet evaporation continues until either the environment is saturated or  
364 all the droplets evaporate. According to the classical concept, during extremely inhomogeneous  
365 mixing the shape of DSD is conserved, however, the total droplet concentration decreases (see  
366 review by Devenish et al., 2012 and Pt 1).

367 The classical concepts analyze only the final equilibrium states which are based on the mass  
368 conservation consideration. Strictly speaking, the size distributions in the final states, assumed in  
369 the classical concepts, are hypothetical and cannot be reached. This is because the classic  
370 concepts do not take into account the mixing-induced DSD broadening. Detailed simulation of  
371 time evolution of DSD and other microphysical parameters is necessary not only to better  
372 determine the final states, but also to evaluate time periods during which such final states are  
373 reached. The analysis of time evolution is practically important because many DSDs measured  
374 in-situ correspond to the transient state, but not to the final equilibrium state. Besides, it is  
375 necessary to determine the evolution of initially polydisperse DSD, that may substantially differ  
376 from the evolution of monodisperse DSD.

377 We analyze the time evolution of the DSD during homogeneous mixing in cases of  
378 monodisperse and polydisperse initial DSDs. The analysis is based on new equations and  
379 methodology developed by Pinsky et al. (2013, 2014).

380 First, we need to evaluate conditions at which mixing can be considered homogeneous. The  
381 characteristic spatial scale of homogeneous mixing can be estimated by comparing the

382 characteristic times of two processes providing thermodynamic equilibrium inside a mixing  
 383 volume. The first process is mechanical mixing (diffusion) governed by turbulence. Turbulent  
 384 mixing leads to homogenization of temperature, humidity (and, thus, of supersaturation) as well  
 385 as of droplet concentration within the volume  $V = V_1 + V_2$ . The second process is evaporation of a  
 386 droplet ensemble, which leads to an increase in relative humidity and to the thermodynamical  
 387 equilibrium in the mixing volume.

388 The process of mixing that is accompanied by droplet evaporation is characterized by two  
 389 time scales. The first time scale is the characteristic mixing (homogenization) time  $\tau_{mix}$  of an  
 390 entrained volume with linear scale  $L_{mix}$  can be evaluated from the relationship (Monin and  
 391 Yaglom, 1975)

$$392 \quad \tau_{mix} = \varepsilon^{-1/3} L_{mix}^{2/3}, \quad (1)$$

393 where  $\varepsilon$  is the turbulent kinetic energy dissipation rate. The estimation (1) suggests that the size  
 394 of the volume falls within the inertial interval of turbulence. Therefore, after the time  $\tau_{mix}$ ,  
 395 volume with a linear scale of about  $L_{mix}$  will be mechanically homogenized and all the droplets  
 396 in the volume will experience the same supersaturation.

397 The second time scale characterizes rate of droplet evaporation and corresponding changes of  
 398 supersaturation. In this study, as well as later in Part.3, it will be shown that the characteristic  
 399 evaporation time is the *phase relaxation time*  $\tau_{pr}$ , that determines the rate of change of  
 400 supersaturation during condensation or evaporation (Mazin, 1968; Korolev and Mazin, 2003)

$$401 \quad \tau_{pr} = (4\pi\mathcal{D}\bar{r}N)^{-1}, \quad (2)$$

402 where  $N = N_{m0}$  is the concentration of droplets in the mixing volume,  $\bar{r}$  is the mean radius of  
 403 droplets and  $\mathcal{D}$  is the diffusivity of water vapor. The spatial scale at which the mixing time is

404 equal to the phase relaxation time is called a phase relaxation scale  $L_{pr}$  (Mazin, 1968). This scale  
 405 can be calculated from Eqs. (1) and (2) as

$$406 \quad L_{pr} = \varepsilon^{1/2} \tau_{pr}^{3/2} \approx \varepsilon^{1/2} (4\pi \mathcal{D} \bar{r} N)^{-3/2} \quad (3)$$

407 The type of mixing is often characterized by the value of the Damköhler number which in  
 408 regard to atmospheric mixing is defined as the ratio  $\tau_{mix} / \tau_{pr}$  (Baker et al., 1980; Jeffery, 2007;  
 409 Lehmann et al., 2009):

410

$$411 \quad Da = \frac{\tau_{mix}}{\tau_{pr}} = \frac{4\pi \mathcal{D} \bar{r} N L_{mix}^{2/3}}{\varepsilon^{1/3}} \quad (4)$$

412 The case  $Da \ll 1$  corresponds to homogeneous mixing, when mechanical homogenization  
 413 occurs much faster than does droplet evaporation. The case  $Da \gg 1$  corresponds to extremely  
 414 inhomogeneous mixing. It is reasonable to consider the value  $Da = 1$  as a boundary separating  
 415 the two types of mixing. This condition is equivalent to the condition

416

$$417 \quad L_{mix} = L_{pr} \approx \varepsilon^{1/2} (4\pi \mathcal{D} \bar{r} N)^{-3/2} \quad (5)$$

418

419 Expression (5) determines the maximum spatial scale at which mixing can be considered as  
 420 homogeneous. The evaluation of the spatial scales at conditions typical of different cloud types  
 421 is presented in **Table 1**. One can see that the characteristic volume size at which mixing can be  
 422 considered homogeneous ranges from 0.2 m to 0.6 m. At larger scales, supersaturation within the  
 423 mixing volume is non-uniform and droplets in the volume experience different values of relative  
 424 humidity. In this case, the mixing should be considered inhomogeneous.

425 The further paper structure is the following. In Section 2 we calculate the thermodynamic  
 426 characteristics of the resulting volume at the end of the first stage of mixing (see Fig. 1b).



427 Section 3 presents an analytic solution of homogeneous droplet evaporation in the monodisperse  
 428 DSD case. In Section 4 effects of polydispersivity on the DSD evolution are described. The  
 429 problem of turbulent mixing representation in numerical cloud models is discussed in Section 5.  
 430 The main results of the study are presented in Conclusion Section 6.

431

## 432 **2. Thermodynamic characteristics of the mixing volume at the end of the first stage**

433 At the first stage (see the scheme in Fig.1) homogeneous mixing is considered as an isobaric  
 434 process that is not accompanied by phase transitions (Korolev and Isaac, 2000). Let us consider  
 435 mixing between a cloud volume with mass  $m_1$ , supersaturation  $S_1 = 0$  and temperature  $T_1$  and a  
 436 droplet-free volume with mass  $m_2$ , supersaturation  $S_2 < 0$  and temperature  $T_2$ . The cloud  
 437 volume also contains droplets with concentration  $N_1$  and liquid water mixing ratio  $q_1$  (Fig. 1).  
 438 For the sake of simplicity, we assume that the mass of both volumes is equal to one. Let us  
 439 further assume that  $\mu$  is the mass fraction of the cloud air that mixes with the mass fraction  
 440  $(1 - \mu)$  of the droplet-free air. In this case, the air mass in the mixing volume will be equal to  
 441  $m_1\mu + (1 - \mu)m_2 = 1$ . Isobaric mixing leads to an approximate linear dependence of droplet  
 442 concentration  $N_{m_0}$  on  $\mu$ .

443 After an instantaneous homogenization of the two volumes, the intermediate temperature  
 444  $T_{m_0}$ , droplet concentration  $N_{m_0}$  and liquid mixing ratio  $q_{m_0}$  are (see Pt.1):

$$445 \quad N_{m_0} = N_1\mu; \quad q_{m_0} = q_1\mu; \quad T_0 = T_1\mu + T_2(1 - \mu) \quad (6a)$$

446 If the temperature difference  $|T_1 - T_2|$  does not exceed a few degrees, the intermediate  
 447 supersaturation  $S_{m_0}$  can be approximated by a linear dependence on  $\mu$

$$448 \quad S_{m_0} = S_2(1 - \mu) \quad (6b)$$

449 **Figure 2** shows, that in case  $|T_1 - T_2| < 2^\circ C$ , the deviations of supersaturation from the linear  
 450 dependence (6b) are small enough and can be neglected. In cases when the temperature of the  
 451 dry volume substantially differs from the temperature of the cloud volume, the dependence of the  
 452 resulting supersaturation on parameter  $\mu$  becomes non-linear (Fig. 2). At temperature  
 453 differences of  $5-10^\circ C$ , the deviation from the analytical solution (6) increases, which requires  
 454 using more precise formulas for supersaturation (see Pt. 1). Values of  $N_{m0}$ ,  $q_{m0}$ ,  $T_{m0}$  and  $S_{m0}$   
 455 determine the initial conditions for the second stage of homogeneous mixing. This is actually  
 456 homogeneous evaporation of droplets, which leads to a thermodynamic equilibrium between  
 457 water vapor and liquid water.

458

### 459 **3. Analysis of homogeneous droplet evaporation in the monodisperse DSD case**

460

#### 461 **3.1. Basic assumptions and equations**

462 The second stage of mixing consists in homogeneous evaporation of droplets. The evolution  
 463 of DSD during the second stage is considered here under the following assumptions: a) the  
 464 processes inside the mixing volume are adiabatic, b) the droplet size distribution is  
 465 monodisperse, c) the vertical velocity of the volume  $u_z = 0$  and d) the sedimentation of droplets  
 466 is neglected and their concentration remains constant.

467 The liquid water mixing ratio can be expressed as

$$468 \quad q(t) = \frac{4\pi\rho_w}{3\rho_a} Nr^3(t) \quad (7)$$

469

470 where  $r(t)$  is the radius of droplets and  $N$  is the droplet number concentration. Closed  
 471 equations describing condensation/evaporation in a moving adiabatic air volume were obtained

472 by Pinsky et al. (2013). In an unmoving adiabatic volume, evaporation is described by the  
 473 equation for supersaturation (e.g., Korolev and Mazin, 2003)

$$474 \quad \frac{1}{S+1} \frac{dS}{dt} = -A_2 \frac{dq}{dt} \quad (8)$$

475 and droplet evaporation is described by the simplified equation (Pruppacher and Klett, 2007)

$$476 \quad r \frac{dr}{dt} = \frac{S}{F}, \quad (9)$$

477 where  $S$  is the supersaturation over a flat water surface. For cloud droplets Eq. (9) is valid with  
 478 high accuracy. We do take into account formation of haze particles resulting from droplet  
 479 evaporation. This allows us to neglect the curvature term and chemical term in the evaporation  
 480 equation. Coefficients  $A_2$  and  $F$  in Eqs. (8, 9) are slightly dependent on the temperature and the  
 481 pressure

$$482 \quad A_2 = \frac{1}{q_v} + \frac{L^2}{c_p R_v T^2} \quad (10)$$

$$483 \quad F = \frac{\rho_w L^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_s(T) \mathcal{D}} \quad (11)$$

484 We assume that coefficients  $A_2$  and  $F$  do not change in the course of droplet evaporation.

485 The physical meaning and units of other variables are given in **Appendix A**.

486

### 487 **3.2. Time evolution of supersaturation and of liquid water content**

488 The closed differential equations for the liquid water mixing ratio  $q$  and supersaturation  $S$

489 to be used in the analysis are derived in **Appendix B**.

$$490 \quad \frac{dq}{dt} = BN_{m0}^{2/3} \left[ (S_{m0} + 1) \exp\{-A_2(q - q_{m0})\} - 1 \right] q^{1/3} \quad (12)$$

$$491 \quad \frac{1}{S+1} \frac{dS}{dt} = -A_2 BN_{m0}^{2/3} S \left( q_{m0} - \frac{1}{A_2} \ln \frac{S+1}{S_{m0}+1} \right)^{1/3} \quad (13)$$

492 where

$$493 \quad B = \frac{3}{F} \left( \frac{4\pi\rho_w}{3\rho_a} \right)^{2/3} = const \quad (14)$$

494 The solutions of these equations depend on the values of  $N_{m0}$ ,  $q_{m0}$ ,  $T_{m0}$ , and  $S_{m0}$ , obtained after  
 495 the first stage of mixing. Eqs. (12) and (13) are rigidly connected by the following equation  
 496 directly following from Eq. (8):

497

$$498 \quad \ln[S(t)+1] = -A_2q(t) + C \quad (15)$$

499

500 where  $C = \ln[S_{m0} + 1] + A_2q_{m0}$  is determined by the initial conditions at  $t = 0$ .

501 Since in this study we assume  $S(t) \leq 0$ , it is convenient to use the relative humidity  $RH$  and  
 502 the saturation deficit  $SD$  to characterize the thermodynamic state of the mixing volume. Both  
 503 quantities are easily related to  $S(t)$ :  $SD(t) = -S(t)$ ,  $RH(t) = 1 + S(t)$ . **Figure 3** demonstrates  
 504 dependencies  $S(t)$  and  $q(t)$ , calculated at an initial relative humidity  $RH_{m0}$  that varies from 72%  
 505 to 91.6%, and an initial LWC of  $0.6 \text{ gm}^{-3}$ .  $RH_{m0}$  corresponds to the relative humidity in dry  
 506 volume  $RH_2$ , that ranges from 43% to 83% at  $\mu = 0.5$ . The results of solving Eqs. (14-15) were  
 507 compared with those obtained using a parcel model (Korolev, 1995) in which evaporation is  
 508 described using equations with temperature-dependent parameters, and were found to be in  
 509 excellent agreement. This agreement can be attributed to the fact that temperature changes that  
 510 occurred in the course of the mixing are relatively small, validating the assumption about the  
 511 constancy of  $A_2$  and  $F$ .

512 As seen from Fig. 3, the final equilibrium state may be reached within several seconds. Fig. 3  
 513 shows the possibility of the two final states mentioned above: a) complete droplet evaporation

514 reached at different time instances depending on the initial value of  $S_{m0}$ , and b) partial droplet  
 515 evaporation at  $RH_{m0} > 82\%$ . In the latter case, the final supersaturation is equal to zero.

516

### 517 3.3. Universal dependencies of supersaturation and of LWC on time

518 In order to simplify the further analysis we introduce the following non-dimensional

519 parameters: normalized liquid water mixing ratio  $\tilde{q} = \frac{q}{q_{m0}}$  which is equal to normalized liquid

520 water content, normalized supersaturation  $\tilde{S} = \frac{S}{A_2 q_{m0}}$ , and non-dimensional time  $\tilde{t} = t / \tau_{m0}$ ,

521 where  $\tau_{m0} = (BA_2 N_{m0}^{2/3} q_{m0}^{1/3})^{-1}$  is the time scale. Then the set of non-dimensional equations

522 describing changes of supersaturation and liquid mixing ratio can be written as (see Appendix B

523 for detail)

$$524 \quad \tilde{S}(\tilde{t}) = -\tilde{q}(\tilde{t}) + \gamma \quad (16)$$

$$525 \quad \frac{d\tilde{q}}{d\tilde{t}} = \tilde{q}^{1/3} (\gamma - \tilde{q}) \quad (17)$$

$$526 \quad \frac{d\tilde{S}}{d\tilde{t}} = -(\gamma - \tilde{S})^{1/3} \tilde{S}, \quad (18)$$

527 where

$$528 \quad \gamma = 1 + \frac{S_{m0}}{A_2 q_{m0}} = 1 + \frac{1 - \mu}{\mu} \frac{S_2}{A_2 q_1} \quad (19)$$

529 is dimensionless parameter which depends on the initial supersaturation  $S_{m0}$  and the initial liquid

530 water mixing ratio  $q_{m0}$ . The value of this parameter can be either positive or negative. Eqs. (16-

531 18) are strictly valid if  $|S_{m0}| \ll 1$ , i.e. when the value of supersaturation  $S$  is negligible in

532 comparison with unity in the factor  $(S + 1)^{-1}$  on the left-hand side of Eq. (8). However, a detailed

533 comparison of solutions of Eqs. (16-18) with those obtained using a numerical model showed

534 that Eqs. (16-18) provide accurate solution at  $RH_{m0}$  as low as 30-40%. Eq. (17) should be  
 535 solved with the initial condition  $\tilde{q}(0) = 1$ , and Eq. (18) should be solved with the initial condition  
 536  $\tilde{S}(0) = \frac{S_{m0}}{A_2 q_{m0}} < 0$ . Therefore, solutions of both equations depend on the sole parameter  $\gamma$ . Eqs.  
 537 (17) and (18) are rigidly connected by balance equation Eq. (16).

538 Defining  $x(\tilde{t}) = (\tilde{q}(\tilde{t}))^{1/3}$  and  $\chi = |\gamma|^{1/3} \text{sgn}(\gamma)$ , the solution of Eq. (17) with the initial  
 539 condition  $x(0) = 1$  is

$$540 \quad 2\chi\tilde{t} = \ln \left[ \frac{(1-\chi)^2}{(x-\chi)^2} \frac{x^2 + \chi x + \chi^2}{1 + \chi + \chi^2} \right] + 2\sqrt{3} \left[ \text{atan} \frac{2\sqrt{3}\chi(1-x)}{3\chi^2 + (2+\chi)(2x+\chi)} \right] \quad (20)$$

541 The solution for the normalized supersaturation can be obtained from Eq. (20) and the balance  
 542 equation (16)

$$543 \quad \tilde{S}(\tilde{t}) = -\tilde{q}(\tilde{t}) + \gamma = -x^3(\tilde{t}) + \chi^3 \quad (21)$$

544 **Figure 4** demonstrates time dependencies  $\tilde{S}(\tilde{t})$  and  $\tilde{q}(\tilde{t})$  calculated at different  $\gamma$  at  
 545  $S_{m0} > -10\%$  (i.e.  $RH_{m0} > 90\%$ ). As seen from Fig. 4, the analytical solution is quite close to the  
 546 numerical one. The deviation increases with an increase in parameter  $\gamma$ . At  $\gamma = -0.5$ , the error  
 547 in the final  $RH$  is about 15%. The initial  $RH_{m0}$  in this case is about 90%.  $RH_{m0}$  is the relative  
 548 humidity in the mixing volume  $V$  after the first stage of mixing. As mentioned above,  $RH_{m0}$   
 549 may be substantially higher than  $RH_2$  in the initially droplet-free volume. *In-situ* measurements  
 550 (Gerber et al., 2008) and remote measurements of aerosol humidification (Knight and Miller,  
 551 1998; Bar-Or et al., 2012) indicate the existence of zones of high  $RH$  along cloud edges. **These**  
 552 **observations and results of numerical simulations indicate that the analytical solution (20-21) is a**  
 553 **universal one and applicable at any RH values in cloud surrounding.**

554 The amplitude of the deviation of the analytical solution for supersaturation from the  
 555 modeled result decreases with the decrease of  $\gamma$ . The cause of the deviation is neglecting term  
 556  $(S + 1)$  on the left-hand side of Eq. (8).

557 There are two types of solutions determined by parameter  $\chi$  (or parameter  $\gamma$ ), separated by  
 558 value  $\chi = 0$  (Figs. 3 and 4). Condition  $\chi = 0$  corresponds to  $\mu = \mu_{cr}$  (see Pt 1) and indicates  
 559 complete evaporation of all the droplets and the relative humidity increasing up to 100 %.  
 560 Condition  $\chi > 0$  corresponds to solutions  $\tilde{q}(\tilde{t})$  with asymptotic behavior at  $\tilde{t} \rightarrow \infty$ :  $\tilde{q} \rightarrow \gamma$  and  
 561  $\tilde{S} \rightarrow 0$ , which means that droplets do not completely evaporate. Condition  $\chi < 0$  means that all  
 562 the droplets completely evaporate.

563 At  $\chi = 0$ , the analytical solution is:

$$564 \quad x = \frac{3}{\tilde{t} + 3}; \quad \tilde{q}(\tilde{t}) = \left( \frac{3}{\tilde{t} + 3} \right)^3; \quad \tilde{S}(\tilde{t}) = - \left( \frac{3}{\tilde{t} + 3} \right)^3 \quad (22)$$

565 Both LWC and supersaturation tend to zero at  $\tilde{t} \rightarrow \infty$ .

566 At  $\chi < 0$ , the duration of the evaporation is limited in time (Figs. 3, 4). Normalized  
 567 evaporation time  $t_e$  depends on parameter  $\chi$  only. This dependence can be obtained from Eq.  
 568 (20) at  $x = 0$ :

$$569 \quad t_e = \frac{1}{2\chi} \ln \frac{(1-\chi)^2}{1+\chi+\chi^2} + \frac{\sqrt{3}}{\chi} \operatorname{atan} \frac{\sqrt{3}}{2\chi+1} \quad (23)$$

570 Here the time is counted in the relaxation time scales. This fact indicates that the phase  
 571 relaxation time is the time scale that should be used in the analysis of mixing. The choice of a  
 572 time scale will be discussed elaborated in greater detail below.

573 At  $\chi > -1/2$ , one have to use the values  $\text{atan}\left(\frac{\sqrt{3}}{2\chi+1}\right) - \pi$  in Eq. (23) instead of  $\text{atan}\frac{\sqrt{3}}{2\chi+1}$ .

574  $t_e$  is the time needed for supersaturation to reach its maximal value (Figs. 3a, 4a). This value is

575 calculated from Eqs. (16) and (6)

$$576 \quad \tilde{S}_{\max} = \gamma = \chi^3 = 1 + \frac{S_{m0}}{A_2 q_{m0}} \approx 1 + \frac{S_2}{A_2 q_1} \frac{1-\mu}{\mu} = 1 + R \frac{1-\mu}{\mu} \quad (24)$$

577 where  $R = \frac{S_2}{A_2 q_1}$  is a dimensionless parameter describing the ratio of  $S_2$  in the initially dry air

578 and the reserve of liquid water available for evaporation. (This parameter can be determined

579 more precisely using Eq. (2) or Eq. (4) given in Pt. 1).

580 The dependence  $t_e(\chi)$  is shown in **Figure 5**. One can see that at large sub-saturation values

581 ( $\chi < -0.4$ ) all the droplets evaporate within the span of a few relaxation times, and the analytical

582 results agree well with the model (benchmark) results. At a high initial  $RH$ , droplet evaporation

583 increases the humidity to nearly the saturation value, at which evaporation becomes extremely

584 slow. In this case, simplified analytical formulas overestimate the evaporation time.

585 At  $\chi > 0$ , droplets partially evaporate,  $q$  reaches the minimal value and the thermodynamic

586 equilibrium is reached when  $S \rightarrow 0$ . Minimal normalized equilibrium LWC can be found from

587 Eqs. (21) and (6)

$$588 \quad \tilde{q}_{\min} = \gamma = \chi^3 = 1 + R \frac{1-\mu}{\mu} \quad (25)$$

589 Eq. (25) is similar to Eqs. (4-5) (see Pt.1.).

590 Dependences of  $\tilde{S}_{\max}$  and  $\tilde{q}_{\min}$  on cloud air fraction, calculated at different initial conditions

591 using Eqs. (24) and (25) are shown in **Figure 6**. The diagram in Fig. 6b is actually a kind of

592 widely used mixing diagram for homogeneous mixing, and Eq. (25) is an universal analytic

593 equation for calculation of this diagram depending on the non-dimensional parameter  $R$ . Panel



594 (a) shows that the higher the cloud fraction of air parcels involved in mixing, the lower the  
 595 saturation deficit, i.e. the higher the final  $RH$  is. At small  $R$ , saturation is reached at a low  $\mu$ .  
 596 Similar diagrams in physical units are shown in Pt.1 in Figs. 4a,g.

597 The equations presented above allow to predict the results of homogeneous mixing both for  
 598 partial and complete evaporation of droplets (**Table 2**). One can see that at low temperatures,  
 599 the environment air volume becomes cloudy at relatively low values of liquid water content in  
 600 the cloud volume even if  $RH$  of the environment volume is as low as 50%. The reason is that the  
 601 saturation ratio at low temperatures is low and only a small amount of liquid should be  
 602 evaporated to make the initially dry volume saturated.

603 The diagram in Fig.6b allows to calculate the final liquid water content remaining in the  
 604 entire volume after saturation reaches 100%. This diagram also shows that for each value of  $\mu$   
 605 there is a certain value of  $R$  at which full evaporation takes place. The remaining  $\tilde{q}_{\min}$  increases  
 606 with decreasing  $\mu$  and increasing  $R$ , as shown by the arrow.

607 The natural time scale of the evaporation and, therefore, homogeneous mixing was  
 608 determined as  $\tau_{m0} = (BA_2 N_{m0}^{2/3} q_{m0}^{1/3})^{-1}$ . This value is inversely proportional to  $N_{m0} r_{m0}$  and actually  
 609 coincides with the initial phase relaxation time  $\tau_{pr}$ . The equality of the characteristic time scales  
 610 of variations in supersaturation and in liquid water content directly follows from the equation  
 611 (15). Indeed, in case of  $|S| \ll 1$  the equation can be rewritten as  $S(t) = -A_2 q(t) + C$  thus  
 612 establishing a linear relationship between the supersaturation and the liquid water mixing ratio.

613 It should be emphasized that  $\tau_{m0}$  is not the time of total droplet evaporation or the time of  
 614 reaching saturation (when evaporation is over). The time of complete evaporation of all droplets  
 615 can be substantially longer than phase relaxation time  $\tau_{m0}$ .

616

#### 617 **4. Analysis of homogeneous droplet evaporation in case of a polydisperse DSD**

618 **4.1. DSD evolution in the course of droplet evaporation**

619 To analyse polydisperse DSD evolution during droplet evaporation at the second stage of  
 620 homogeneous mixing, we use the same equations for diffusional growth (9) and supersaturation  
 621 (8) as those used in case of a monodisperse DSD. The solution of Eq. (9) can be written in the  
 622 form

$$623 \quad r^2(t) = r_0^2 - Q(t) \quad (26)$$

624 where the non-negative function  $Q(t) \geq 0$  is proportional to the supersaturation integral

$$625 \quad Q(t) = -\frac{2}{F} \int_0^t S(t') dt' \geq 0 \quad (27)$$

626 This function characterizes a decrease in the square of droplet radii. At  $t=0$ ,  $Q(0)=0$ . Let  
 627  $f_0(r_0)$  be an initial DSD immediately following the first stage of homogeneous mixing. This  
 628 distribution obeys the normalization condition

$$629 \quad N_{m0} = \int_0^\infty f_0(r_0) dr_0 \quad (28)$$

630 where  $N_{m0}$  is the initial droplet number concentration after the first stage of mixing. Using the  
 631 inverse transformation  $r_0 = \sqrt{r^2 + Q(t)}$  alongside with condition  $r \geq 0$  and the relation between  
 632 the distribution functions  $f(r, t) = f_0(r_0) \frac{dr_0}{dr}$  we get

$$633 \quad f(r, t) = \begin{cases} \frac{r}{\sqrt{r^2 + Q(t)}} f_0\left(\sqrt{r^2 + Q(t)}\right), & r \geq 0 \\ 0, & r < 0 \end{cases} \quad (29)$$

634 Eq. (29) shows that the time changes in DSD and in its moments depend both on the initial DSD  
 635 at  $t=0$ ,  $f_0(r_0)$  and on the time-dependent function  $Q(t) \geq 0$ .

636 To illustrate the DSD evolution using Eq. (29), we assume that the initial distribution  
 637 immediately following the first stage of mixing can be represented by a Gamma distribution:

$$638 \quad f_0(r_0) = \frac{N_{m0}}{\Gamma(\alpha)\beta} \left(\frac{r_0}{\beta}\right)^{\alpha-1} \exp\left(-\frac{r_0}{\beta}\right) \quad (30)$$

639 where  $N_{m0}$  is an intercept parameter,  $\alpha$  is a shape parameter and  $\beta$  is a slope parameter of  
 640 distribution. Different sets of parameters allow approximations of both narrow and wide DSD.  
 641 The parameters of the initial Gamma distribution used in this study are presented in **Table 3** and  
 642 are chosen so that the modal radii of DSD and the LWC would be the same for both  
 643 distributions.

644 Combining Eqs. (29) and (30) yields an equation for DSD evolution as a function of  $Q(t)$

$$645 \quad f(r,t) = \begin{cases} \frac{N_{m0}}{\Gamma(\alpha)\beta^\alpha} (r^2 + Q)^{\frac{\alpha}{2}-1} r \exp\left(-\frac{\sqrt{r^2 + Q}}{\beta}\right), & r \geq 0 \\ 0, & r < 0 \end{cases} \quad (31)$$

646 This DSD (31) depends on four parameters, wherein parameter  $Q(t)$  increases with time  
 647 according to Eq. (27). Examples of evolutions of an initially narrow DSD and an initially wide  
 648 DSD are shown in **Figure 7**. All the calculations were performed using a parcel model (Korolev,  
 649 1995).

650 There is a significant difference between evolution of DSDs in cases of monodisperse and  
 651 polydisperse DSDs. At a monodisperse DSD, droplet concentration remains unchanged until the  
 652 final stage of evaporation when droplets become small and then all evaporate rapidly. At a  
 653 polydisperse DSD the droplet concentration decreases simultaneously with the decrease in  
 654 LWC. Evaporation of a the narrow size distribution is consistent with the concept of  
 655 homogeneous mixing (Fig. 7a). However homogeneous evaporation of droplets with a wide  
 656 DSD may be mistakenly taken for inhomogeneous mixing.

657

## 658 **4.2. Evolution of DSD moments and related functions**

659 Eq. (29) also allows evaluation of droplet concentration, DSD moments and related  
 660 functions. Droplet concentration corresponds to the zero moment of DSD and can be expressed  
 661 as

$$662 \quad N(t) = \int_0^{\infty} f(r, t) dr = \int_0^{\infty} \frac{r}{\sqrt{r^2 + Q(t)}} f_0\left(\sqrt{r^2 + Q(t)}\right) dr = \int_{\sqrt{Q(t)}}^{\infty} f_0(r_0) dr_0 \quad (32)$$

663 Since function  $Q(t)$  monotonically increases with the time, the right-hand integral in Eq. (32)  
 664 decreases, indicating a decrease in droplet concentration with time. If the initial distribution of  
 665 droplets is described by a Gamma distribution, the decrease of droplet concentration with the  
 666 time is evaluated using Eqs. (30) and (32) as

$$667 \quad N(t) = \int_{\sqrt{Q(t)}}^{\infty} f_0(r_0, t) dr_0 = \int_{\sqrt{Q(t)}}^{\infty} \frac{N_{m0}}{\Gamma(\alpha)\beta} \left(\frac{r_0}{\beta}\right)^{\alpha-1} \exp\left(-\frac{r_0}{\beta}\right) dr_0 =$$

$$668 \quad \frac{N_{m0}}{\Gamma(\alpha)} \int_{\frac{\sqrt{Q(t)}}{\beta}}^{\infty} x^{\alpha-1} \exp(-x) dx = N_{m0} \frac{\Gamma(\alpha, \eta)}{\Gamma(\alpha)}. \quad (33)$$

669

670 where

$$671 \quad \eta(t) = \frac{\sqrt{Q(t)}}{\beta} \quad (34)$$

672 is a non-dimensional function of time and  $\Gamma(\alpha, \eta(t))$  is an upper incomplete Gamma function  
 673 (Korn and Korn, 2000). It follows from Eqs. (32) and (33) that  $N(t) \leq N_0$ . The dependencies of  
 674 the normalized droplet concentration  $\frac{N(t)}{N_{m0}}$  on time for an initially narrow DSD and an initially  
 675 wide DSD are shown in **Figure 8**.

676 Fig. 8 shows that in case of an initially narrow DSD, droplet concentration does not change  
 677 during the first 20 s when  $RH_{m0} = 91.6\%$  because the DSD does not shift strongly enough  
 678 toward smaller droplet radii. At lower initial supersaturations, droplet concentration decreases

679 with time. This decrease may take place rapidly and may last several seconds only. The red line  
 680 separates two different evaporation scenarios. The curves above the red line correspond to partial  
 681 evaporation and reaching the saturation state, whereas the curves below the red line correspond  
 682 to complete evaporation of droplets and the environment remains subsaturated.

683 At an initially wide DSD, droplet concentration decreases at any initial subsaturation value  
 684 because the DSD contains small droplets that start evaporating regardless the subsaturation  
 685 value. In the particular example shown in Fig. 8, the droplet relaxation time is shorter in case of a  
 686 wide DSD, so droplet concentration decreases slower than at a narrow DSD. Droplet  
 687 concentration decreases substantially during a few tens of seconds, but does not reach zero due  
 688 to a significant concentration of large droplets in the wide DSD. The equilibrium state is not  
 689 reached within 20 s.

690 Fig. 8 also demonstrates an excellent agreement between the results of analytical calculations  
 691 performed using Eq. (33) and results obtained using the parcel model.

692 A normalized moment of the  $k$ -th order is evaluated as

$$\begin{aligned}
 \overline{r^k}(t) &= \frac{1}{N(t)} \int_0^\infty r^k f(r,t) dr = \frac{1}{N(t)} \int_0^\infty \frac{r^{k+1}}{\sqrt{r^2 + Q(t)}} f_0\left(\sqrt{r^2 + Q(t)}\right) dr = \\
 693 \quad &\frac{1}{N(t)} \int_{\sqrt{Q(t)}}^\infty (r_0^2 - Q(t))^{k/2} f_0(r_0) dr_0 \\
 694 \quad &\overline{r^k}(t) = \frac{\int_{\sqrt{Q(t)}}^\infty (r_0^2 - Q(t))^{k/2} f_0(r_0) dr_0}{\int_{\sqrt{Q(t)}}^\infty f_0(r_0) dr_0} \quad (35)
 \end{aligned}$$

696 In case when the initial distribution is given by the Gamma distribution (30), Eq. (35) leads to  
 697 the following equation

$$698 \quad \overline{r^k}(t) = \frac{\beta^k}{\Gamma(\alpha, \eta(t))} \int_{\eta(t)}^\infty (x^2 - \eta^2(t))^{k/2} (x)^{\alpha-1} \exp(-x) dx \quad (36)$$

699 The even moments can be represented using incomplete the Gamma functions.

700 **Figures 9 and 10** show the time dependencies of quantities typically used for characterizing

701 the DSD shape, namely the mean radius  $\bar{r}(t)$  (panel (a)), the effective radius  $r_{eff}(t) = \frac{\bar{r}^3(t)}{r^2(t)}$

702 (panel (b)), the RMS width of DSD  $\sigma(t) = \sqrt{r^2(t) - \bar{r}^2(t)}$  (panel (c)) and the dispersion

703 coefficient  $\delta(t) = \frac{\sigma(t)}{\bar{r}(t)}$  (panel (d)). The dependencies corresponding to an initially narrow DSD

704 are shown in Fig. 9 and those corresponding to an initially wide DSD are shown in Fig. 10.

705 At an initially narrow DSD, the mean radius and the effective radius decrease with time in  
706 agreement with the concept of homogeneous mixing. Formation of plateaus in the mean radii, in  
707 the effective radii and in droplet dispersion over long time periods is caused by the existence of  
708 droplets in the tail of DSD distributions. While the concentration of such droplets is negligibly  
709 small, their evaporation takes a significant amount of time.

710 At an initially wide DSD (Fig.7b), complete evaporation of the smallest droplets starts at the  
711 very beginning of the second stage of mixing. This evaporation leads to an increase in the  
712 effective radius and in the mean radius that changes non-monotonically. **An increase in the**  
713 **value of the effective radius contradicts the concept of homogeneous mixing, according to which**  
714 **both the mean radii and the effective radii decrease in the course of mixing.** This increase is  
715 explained by the fact that subsaturation at an initially wide DSD leads to a significant rapid  
716 decrease in the concentration of small droplets, while the changes in LWC whose value is  
717 determined by larger droplets, do not occur that quickly.

718 The opposite behaviors of the effective radii (as well as of the other characteristic droplet  
719 sizes) at a narrow DSD vs. a wide DSD, illustrated in Figs. 9 and 10, suggest the existence of a  
720 large number of DSDs with initial shapes at which the evaporation of droplets leads to a decrease  
721 in droplet concentration, but does not change significantly the effective radius. So the constancy

722 of the effective radius at varying droplet concentrations does not allow to distinguish the mixing  
 723 type with full confidence. Even at a narrow DSD, the decrease in the value of the effective radius  
 724 does not exceed 20% at the an initial saturation deficit of 8.4 %.

725 In any case the evolution of DSD and of their parameters is determined by the competition  
 726 between two effects. First, this is the effect of partial droplet evaporation which shifts the DSD  
 727 toward smaller sizes and leads to a decrease in the mean radii and in the effective radii, as well  
 728 as widens the DSD. Second, this is the effect of complete evaporation of the smallest droplets,  
 729 which increases both the mean radii and the effective radii. The relative contribution of these two  
 730 effects depends on the initial DSD width and the value of the mean radius. The best indicators of  
 731 these two effects are the DSD width,  $\sigma$  and the DSD dispersion  $\frac{\sigma}{\bar{r}}$ .

732 At an initially narrow DSD and an initially low  $RH$ , partial evaporation initially dominates  
 733 and the DSD width increases due to the appearance of smaller droplets (Fig. 9c). Afterwards,  
 734 when complete droplet evaporation becomes the dominant factor, the DSD shifts significantly to  
 735 small sizes and the DSD width decreases. At an initially large  $RH$ , complete droplet evaporation  
 736 is not efficient, and the DSD gets continuously wider. Since the mean radius decreases, the DSD  
 737 dispersion tends to constant values at any initial  $RH$ . However, the largest DSD dispersion takes  
 738 place at an initially low  $RH$ , when evaporation substantially decreases the mean droplet radius.  
 739 As seen in Figs. 9d and 10d, the initial DSD dispersion at a narrow DSD is 0.1, while the initial  
 740 dispersion at a wide DSD is 0.5.

741 Figs. 10c and 10d show that at an initially wide DSD, homogeneous evaporation leads to an  
 742 increase in both DSD width and DSD dispersion. The increase in the DSD width indicates that  
 743 formation of the smallest droplets by partial evaporation is the main mechanism of the DSD  
 744 shape evolution. The DSD dispersion increases with time and rapidly reaches quasi-stationary  
 745 values of about 0.56 that are typical of real clouds.

746

747 **4.3. Evolution of LWC and of supersaturation**

748 Using Eq. (35), the time dependence of the liquid water mixing ratio is represented as

749 
$$q(t) = \frac{4\pi\rho_w}{3\rho_a} N(t) \overline{r^3(t)} = \frac{4\pi\rho_w}{3\rho_a} \int_{\sqrt{Q(t)}}^{\infty} (r_0^2 - Q(t))^{3/2} f_0(r_0) dr_0 \quad (37)$$

750 If the initial DSD is approximated by a Gamma distribution,  $q(t)$  can be written as

751 
$$q(t) = \frac{4\pi\rho_w}{3\rho_a} \int_{\sqrt{Q(t)}}^{\infty} (r_0^2 - Q(t))^{3/2} \frac{N_{m0}}{\Gamma(\alpha)\beta} \left(\frac{r_0}{\beta}\right)^{\alpha-1} \exp\left(-\frac{r_0}{\beta}\right) dr_0 =$$

$$\frac{4\pi\rho_w}{3\rho_a} \frac{N_{m0}\beta^3}{\Gamma(\alpha)} \int_{\eta(t)}^{\infty} (x^2 - \eta^2(t))^{3/2} (x)^{\alpha-1} \exp(-x) dx \quad (38)$$

752 Since at  $t=0$ ,  $\eta=0$  and the initial liquid water mixing ratio is equal to

753 
$$q_{m0} = \frac{4\pi\rho_w}{3\rho_a} N_{m0}\beta^3 \frac{\Gamma(\alpha+3)}{\Gamma(\alpha)},$$
 hence the normalized liquid water mixing ratio is calculated as

754 
$$\frac{q(t)}{q_{m0}} = \frac{1}{\Gamma(\alpha+3)} \int_{\eta(t)}^{\infty} (x^2 - \eta^2(t))^{3/2} (x)^{\alpha-1} \exp(-x) dx \quad (39)$$

755

756 **Figure 11** shows the time dependencies of LWC for different initial  $RH_{m0}$  in the resulting  
 757 volume (Fig.11a: an initially narrow DSD; Fig.11b: an initially wide DSD). One can see that at  
 758 an initially narrow DSD the LWC rapidly decreases, either to zero (full evaporation) or, as in the  
 759 monodisperse case, to an equilibrium value during a time period of the phase relaxation time (see  
 760 Section 3 for comparison). At an initially wide DSD, the LWC decreases slowly and  
 761 monotonically. In general, at an initially narrow DSDs the time dependencies,  $q_w(t)$  are quite  
 762 close to those for monodisperse DSD (Fig. 3b). The higher the initial  $RH$ , the smaller the change  
 763 in the LWC is.

764 To calculate the time dependencies of supersaturation, one can use the full equation (8) or  
 765 Eq. (15) written in the form:



$$766 \quad S(t) = (S_{m0} + 1) \exp\{-A_2 [q(t) - q_{m0}]\} - 1 \quad (40)$$

767 In Eq. (40),  $S_{m0}$  and  $q_{m0}$  are the initial supersaturation and the liquid water mixing ratio,  
 768 respectively, at  $t=0$ . The corresponding time dependencies are shown in **Figure 12**. The  
 769 analytical results of Eq. (40) are compared with the exact numerical solution obtained by a parcel  
 770 model, showing a good agreement. The behavior of supersaturation at an initially narrow DSD is  
 771 similar to that of a monodisperse DSD (Fig. 3a). As in Fig. 3a, the equilibrium non-zero values  
 772 of subsaturation correspond to complete droplet evaporation. At an initially wide DSD, the  
 773 saturation deficit monotonically decreases as a consequence of the monotonic decrease in the  
 774 LWC.

775 **Figure 13** shows the dependencies of normalized LWC on normalized droplets  
 776 concentrations calculated at different values of the initial relative humidity  $RH_{m0}$  in the mixing  
 777 volume ((a): the initially narrow DSD and (b) the initially wide DSD). Each point on the curves  
 778 corresponds to a certain time. Since the dependencies are plotted in non-dimensional  
 779 coordinates, the time instance  $t=0$  corresponds to coordinates (1, 1). The numbers along the  
 780 curves denote the points corresponding to time instance  $t=20$ s. At lower  $RH_{m0}$ , the curves  
 781 reach lower values of LWC and of droplet concentration. In case of an initially narrow DSD the  
 782 curve can be divided into three sections. The first section corresponds to high  $RH_{m0}$   
 783 ( $RH_{m0} > 90\%$ ). Within this section, the droplet concentration does not decrease with decreasing  
 784 LWC, which is in line with the conceptual scheme of homogeneous mixing (Fig. 1). This section  
 785 corresponds to the straight horizontal line in Fig. 8a. At lower relative humidity, the droplet  
 786 concentration begins to decrease with a decreasing LWC, in line with Fig. 8a. When the LWC  
 787 reaches small values, the dependence of the normalized LWC on the normalized droplet  
 788 concentration becomes close to linear. At an initially wide DSD (Fig. 13b), the dependence of

789 the LWC on droplet concentration is close to linear at all the values of  $RH_{m0}$ . This linear  
 790 dependence means that the mean volume radius varies only slightly during droplet evaporation.

791 It is noteworthy that all the curves plotted for different initial values of  $RH_{m0}$  coincide.  
 792 This coincidence can be explained by the fact that all the DSDs used depend on four parameters,  
 793 namely, the three parameters of the initial Gamma distribution and on  $Q(t)$  (Eq. 31). The  
 794 parameters of Gamma distributions are identical for each panel in Fig. 13. Parameter  $Q(t)$   
 795 monotonically increases with time. At different  $RH_{m0}$ , the values of  $Q(t)$  reach the same values  
 796 at different times. When the values of  $Q(t)$  are the same, the DSDs and all the DSD moments  
 797 are equal.

798 To sum up, at an initially narrow DSD and a comparatively large initial relative humidity  
 799  $RH_{m0} > 87\%$  phenomenon demonstrates properties typically attributed to homogeneous mixing,  
 800 when a decrease in both the LWC and the effective radius takes place at unchanged droplet  
 801 concentration (Figs. 8a, 9b, 11a, 13a). In contrast, in the case of an initially wide DSD, the  
 802 evolution of the DSD and its moments is close to that typically attributed to inhomogeneous  
 803 mixing, when droplet evaporation leads to a decrease in LWC and in droplet concentration,  
 804 while the effective radius remains unchanged. During the changes of a wide DSD, its shape  
 805 remains similar to itself (Fig. 7b), which is also considered typical of inhomogeneous mixing.

806

## 807 **5. Discussion: Application of the concept of homogeneous mixing in numerical modeling**

808

809 The procedure of mixing in any cloud model involves two steps. At the first step, the changes  
 810 in microphysical values in each grid point are calculated using the turbulent flux divergences. In  
 811 case mixing takes place between any two volumes represented by neighbouring grid points, the  
 812 mixing volume containing both grid points never becomes homogeneous (*a fortiori*, the

813 microphysical values in these grid points do not become identical during one time step), so the  
 814 spatial gradients of the microphysical variables remain between neighbouring grid points. This  
 815 step represents inhomogeneous mixing at resolving scales. Mixing algorithm in models does not  
 816 operate with "final" equilibrium values, as assumed in the classical mixing concepts, but rather  
 817 with current time-dependent values. In contrast, the changes in the microphysical and  
 818 thermodynamical variables in the volumes represented by one grid points are often considered  
 819 uniform at each time step, and therefore, the modelled subgrid mixing is treated as  
 820 homogeneous. Therefore, in most numerical models mixing is inhomogeneous at resolved scales,  
 821 but homogeneous at subgrid scales.

822 The estimations in Tab. 1 indicate that mixing is homogeneous at scales lower than  $\sim 0.5$  m.  
 823 This means that to simulate homogeneous mixing explicitly, the grid spacing should be less than  
 824 0.5 m. If such grid spacing is used, the separation between mixing types could be described  
 825 explicitly. However, grid spacing in most models significantly exceeds this value. This fact  
 826 brings up two questions: "What error is introduced when the spatial scale separating mixing  
 827 types in models is much larger than 0.5 m?" and "Why are spectral microphysics models with a  
 828 resolution of 40-50 m able to reproduce observed DSD and their moments with high accuracy  
 829 (Benmoshe et al., 2012; Khain et al., 2013, 2015; Magaritz-Ronen et al., 2014)?"

830 There are several factors that compensate errors in segregating mixing types in cloud models  
 831 and allow using grid scale  $L > L_{pr}$  with little effect on DSD. The first factor is that mixing leads  
 832 to formation of cloud zones characterized by a spatial correlation scale (radius of correlation) of  
 833 temperature, humidity and droplet concentration of about 150-250 m (Magaritz-Ronen et al.,  
 834 2014). Numerical experiments with Lagrangian-Eulerian model of Sc (Magaritz-Ronen et al.,  
 835 2014) have shown that the results are not sensitive to the choice of parcel size, if this size is  
 836 substantially smaller than the spatial radius of correlation. Therefore, the mixing type has a  
 837 minor effect on the results of mixing at scales lower than the radius of correlation.

838 The second factor is that in-cloud mixing often takes place at conditions close to saturation.  
 839 At such high humidity, homogeneous and inhomogeneous mixing yield practically the same  
 840 results. The similarity of results for the two mixing types is due to the fact that mixing in clouds  
 841 is not accompanied by an appreciable phase transition.

842 The third factor was pointed out by Hill et al. (2009), who explained that stratocumulus cloud  
 843 evolution is insensitive to the type of sub-grid mixing since the rates of condensation/evaporation  
 844 caused by the resolved dynamics are by two orders of magnitude greater than the  
 845 condensation/evaporation rate caused by the sub-grid processes.

846 The fourth factor that permits us to treat sub-grid mixing as homogeneous near cloud  
 847 interfaces is that DSDs are polydisperse, which is opposite what is assumed in the conventional  
 848 mixing considerations. In the present study it was shown that for a broad DSD, the changes of  
 849  $r_{eff}$  remain small during mixing. So, a relatively small partial evaporation of droplets provide  
 850 sufficient amount of water vapor for saturation of the volume. In this case homogeneous mixing  
 851 becomes indistinguishable from inhomogeneous. The saturation of the volume may be facilitated  
 852 by entrainment of water vapor from neighboring cloud volumes.

853

## 854 **6. Conclusions**

855

856 The present study is focused on the dynamics of DSD transformation during the evaporation  
 857 stage of homogeneous mixing. The results can be summarized as follows.

858 1. Analytical equations describing time evolution of normalized supersaturation and  
 859 normalized LWC are obtained. It is found that these time dependences are universal functions

860 of a sole non-dimensional parameter  $\gamma = 1 + \frac{S_{m0}}{A_2 q_{m0}}$ . In particular, the dependences of normalized

861 LWC at the final stage on the cloud air fraction, used for plotting the universal mixing diagrams,

862 are obtained analytically. These diagrams also depend on a sole non-dimensional parameter  
 863  $R = \frac{S_2}{A_2 q_1} < 0$ , which is proportional to supersaturation in dry volume and inversely proportional  
 864 to liquid water mixing ratio in a cloud volume. This parameter is uniquely related with parameter  
 865  $\gamma$  by Eq. (24). It is shown that in many cases the major changes in the LWC take place during  
 866 the time period of the order of the phase relaxation time  $\tau_{pr}$ . The equilibrium state can be  
 867 reached after several  $\tau_{pr}$  periods.

868 2. It is shown that the phase relaxation time is a natural time scale of mixing process. This is  
 869 clearly seen from the universal renormalized evaporation equations, in which the phase  
 870 relaxation time plays the role of a time unit. In some studies (e.g. Baker and Latham, 1979;  
 871 Burnet and Brenguier, 2007; Andejchuk et al., 2009) evaporation time for an individual droplet  
 872 under given sub-saturation is considered as a characteristic time of mixing. The present study  
 873 shows that only the phase relaxation time should be used as the characteristic time scale of  
 874 mixing since we have to consider the behavior of a large amount of droplets. Supersaturation (or  
 875 sub-saturation) is not a parameter that determines the phase relaxation time. Thus, the utilization  
 876 of the evaporation time of individual droplet (at unchanged supersaturation) as the characteristic  
 877 time scale of mixing is physically ungrounded. A strict relationship between the changes in  
 878 supersaturation and in the liquid water mixing ratio makes it impossible to consider the changes  
 879 in an individual droplet size and in supersaturation independently.

880 3. An important outcome of this study is demonstration of a significant difference in the  
 881 evaporative behavior between narrow DSD and wide DSD. It is shown that homogeneous  
 882 evaporation of a wide DSD is accompanied by reduction in LWC and in droplet concentration  
 883 due to total evaporation of small droplets. Such changes of LWC and droplet concentration are  
 884 qualitatively different from those in the classic concept of homogeneous mixing. As a result,  
 885 homogeneous mixing may be erroneously interpreted as inhomogeneous one.

886 4. It is shown that the evolution of DSDs and their moments in case of polydisperse DSDs,  
887 can qualitatively differ from that predicted by homogeneous mixing concept. Evaporation of a  
888 comparatively wide DSD may even lead to an increase in the effective radii and DSD high  
889 moments. This feature is typically attributed to inhomogeneous mixing.

890 Note that the role of DSD polydispersity in the mixing process is different from that in  
891 diffusion droplet growth in ascending parcels. In an ascending adiabatic parcel the  
892 supersaturation tends to zero with height, and the DSD width decreases with height as well. In  
893 this case, it is possible to reproduce the height dependencies and the time dependencies of  
894 supersaturation and of LWC using an "equivalent" monodisperse DSD with the same droplet  
895 concentration as a polydisperse DSDs (Pinsky et al., 2014). As regards to mixing with a  
896 polydisperse DSD, it cannot be reproduced using a monodisperse DSD, with a possible  
897 exception in case of an initially extremely narrow DSD.

898

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903

904

905 **Appendix A: List of Symbols**

906

907 **Table A1 here**

908

909

910 **Appendix B. Derivation of closed equations for supersaturation and for liquid water**  
 911 **mixing ratio in the monodisperse DSD case.**

912

913 Let us consider motionless well-mixed adiabatic air volume having an initial supersaturation  
 914  $S_{m0} < 0$  and an initial liquid water mixing ratio  $q_{m0}$ .

915 1. Substitution of the formula of the liquid water mixing ratio  $q = \frac{4\pi\rho_w}{3\rho_a} Nr^3$  into the  
 916 equation for droplet radius evolution

$$917 \quad r \frac{dr}{dt} = \frac{S}{F} \quad (\text{B1})$$

918 leads to the equation for the decrease of  $q$  with the time

$$919 \quad \frac{dq}{dt} = \frac{3}{F} \left( \frac{4\pi\rho_w}{3\rho_a} \right)^{2/3} N^{2/3} S q^{1/3} = B N^{2/3} S q^{1/3} \quad (\text{B2})$$

920 where

$$921 \quad B = \frac{3}{F} \left( \frac{4\pi\rho_w}{3\rho_a} \right)^{2/3} = \text{const} \quad (\text{B3})$$

922 2. Equation for supersaturation is written as (Korolev and Mazin, 2003)

$$923 \quad \frac{1}{S+1} \frac{dS}{dt} = -A_2 \frac{dq}{dt} \quad (\text{B4})$$

924 Integration of this equation under the assumption that  $A_2 = \text{const}$  leads to the equation

$$925 \quad \ln[S(t)+1] = -A_2 q(t) + C \quad (\text{B5})$$

926 where  $C$  is determined by initial conditions at  $t = 0$

$$927 \quad C = \ln(S_{m0} + 1) + A_2 q_{m0} \quad (\text{B6})$$

928 Using Eqs. (B5) and (B6) one obtains the equation with respect to  $S(t)$

$$929 \quad S(t) = (S_{m0} + 1) \exp\{-A_2 [q(t) - q_{m0}]\} - 1 \quad (\text{B7})$$

930 3. Mutual substitution of Eqs. (B2) and (B7) leads to the closed differential equations for  
 931  $q(t)$  and  $S(t)$

$$932 \quad \frac{dq}{dt} = BN^{2/3} \left[ (S_{m0} + 1) \exp\{-A_2(q - q_{m0})\} - 1 \right] q^{1/3} \quad (\text{B8})$$

$$933 \quad \frac{1}{S+1} \frac{dS}{dt} = -A_2 BN^{2/3} S \left( q_{m0} - \frac{1}{A_2} \ln \frac{S+1}{S_{m0}+1} \right)^{1/3} \quad (\text{B9})$$

934 Eqs. (B8) and (B9) should be solved with initial conditions  $q(0) = q_{m0}$  and  $S(0) = S_{m0}$   
 935 respectively.

936 4. In case  $|S_{m0}| \ll 1$ , supersaturation is close to zero all the time  $|S(t)| \ll 1$  and Eqs. (B7-B9)  
 937 can be simplified as follows

$$938 \quad S(t) = S_{m0} - A_2 [q(t) - q_{m0}] \quad (\text{B10})$$

$$939 \quad \frac{dq}{dt} = -BN^{2/3} \left( A_2 q^{4/3} - (A_2 q_{m0} + S_{m0}) q^{1/3} \right) \quad (\text{B11})$$

$$940 \quad \frac{dS}{dt} = -B(A_2 N)^{2/3} (A_2 q_{m0} + S_{m0} - S)^{1/3} S \quad (\text{B12})$$

941

942 5. Then one can obtain Eqs. (B10-B12) in a non-dimensional form. Let us define time scale

$$943 \quad \tau_{m0} = \left( BA_2 N^{2/3} q_{m0}^{1/3} \right)^{-1}, \text{ normalized liquid water mixing ratio } \tilde{q} = \frac{q}{q_{m0}}, \text{ normalized supersaturation}$$

$$944 \quad \tilde{S} = \frac{S}{A_2 q_{m0}}, \text{ and non-dimensional time } \tilde{t} = t / \tau_{m0} = BA_2 N^{2/3} q_{m0}^{1/3} t. \text{ The Eqs. (B10-B12) can be}$$

945 rewritten in a non-dimensional form as

$$946 \quad \tilde{S}(\tilde{t}) = -\tilde{q}(\tilde{t}) + \gamma \quad (\text{B13})$$

$$947 \quad \frac{d\tilde{q}}{d\tilde{t}} = \tilde{q}^{1/3} (\gamma - \tilde{q}) \quad (\text{B14})$$

$$948 \quad \frac{d\tilde{S}}{d\tilde{t}} = -(\gamma - \tilde{S})^{1/3} \tilde{S} \quad (\text{B15})$$



949 where non-dimensional parameter  $\gamma = 1 + \frac{S_{m0}}{A_2 q_{m0}}$  depends on initial supersaturation  $S_{m0}$  and  
 950 initial liquid water mixing ratio  $q_{m0}$ . Eq. (B14) should be solved with initial condition  $\tilde{q}(0) = 1$   
 951 and Eq. (B15) should be solved with initial condition  $\tilde{S}(0) = \frac{S_{m0}}{A_2 q_{m0}} < 0$ . Note that Eqs. (B14) and  
 952 (B15) are rigidly connected by Eq. (B13).

953

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1056 spectrum, *J. Atmos. Sci.*, **26**, 1049-1059, 1969.
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1060 **Table 1.** Linear scales of volumes experiencing homogeneous mixing at conditions typical of  
 1061 different cloud types.

Cloud type	$N, cm^{-3}$	LWC $gm^{-3}$	$r, \mu m$	Dissipation rate, $cm^2/s^3$	Phase relaxation time, s	Phase scale, m
Maritime Convective	100	2.0	16.8	300	2.01	0.49
Maritime Stratocumulus	100	0.5	10.6	10	3.19	0.18
Weak Stratocumulus	100	0.2	7.8	5	4.33	0.2
Continental Convective	500	2	8.0	500	0.75	0.6

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1065 **Table 2** Estimations of mixing results at different environmental conditions\*

T ( $^{\circ}C$ )	$A_2$	$q_1, g/kg$	$RH_2$ %	$\mu$ (cloud fraction)	$R \frac{1-\mu}{\mu}$	Result of mixing
$T = 0^{\circ}C$	400	1.0	95	0.5	-0.125	cloudy
$T = 0^{\circ}C$	400	1.0	50	0.5	-1.25	Non-cloudy
$-10^{\circ}C$	700	2.0	95	0.5	-0.036	cloudy
$-10^{\circ}C$	700	2.0	50	0.5	-0.36	cloudy
$-20^{\circ}C$	1500	2.0	95	0.5	-0.017	cloudy
$-20^{\circ}C$	1500	2.0	50	0.5	-0.17	cloudy
$-20^{\circ}C$	1500	2.0	95	0.1	-0.153	cloudy
$-20^{\circ}C$	1500	2.0	50	0.1	-1.53	Non-cloudy

1066 \* The values  $A_2$  are estimated as proposed by Pinsky et al., (2013).

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1069 **Tab.3** Parameters of the initial Gamma distributions

DSD	$N_{m0}$ , cm <sup>3</sup>	$\alpha$	$\beta$ , $\mu\text{m}$	Modal radius, $\mu\text{m}$	LWC, g/m <sup>3</sup>
Narrow	264.2	101.0	0.1	10.0	0.587
Wide	71.0	4.3	3.1	10.0	0.587

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1073 **Tab. A1. List of symbols**

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Symbol	Description	Units
$A_2$	$\frac{1}{q_v} + \frac{L^2}{c_p R_v T^2}$ , coefficient	nd
$B$	$\frac{3}{F} \left( \frac{4\pi\rho_w}{3\rho_a} \right)^{2/3}$ , coefficient	$\text{m}^2 \text{s}^{-1}$
$C$	constant of integration	nd
$c_p$	specific heat capacity of moist air at constant pressure	$\text{J kg}^{-1} \text{K}^{-1}$
$\mathcal{D}$	coefficient of water vapour diffusion in the air	$\text{m}^2 \text{s}^{-1}$
$e$	water vapor pressure	$\text{N m}^{-2}$
$e_s$	saturation vapour pressure above a flat water surface	$\text{N m}^{-2}$
$F$	$F = \frac{\rho_w L^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_s(T) \mathcal{D}}$ , coefficient	$\text{m}^{-2} \text{s}$
$f(r, t)$	droplet size distribution	$\text{m}^{-4}$
$f_0(r_0)$	droplet size distribution after the first stage of mixing	$\text{m}^{-4}$
$k_a$	coefficient of air heat conductivity	$\text{J m}^{-1} \text{s}^{-1} \text{K}^{-1}$
$L_{mix}$	characteristic spatial scale of mixing	m
$L_{pr}$	spatial scale of phase relaxation	m
$L$	latent heat for liquid water	$\text{J kg}^{-1}$
$N$	droplet concentration	$\text{m}^{-3}$
$N_1$	droplet concentration in a cloud volume	$\text{m}^{-3}$
$N_{m0}$	droplet concentration after the first stage of mixing	$\text{m}^{-3}$
$p$	pressure of moist air	$\text{N m}^{-2}$
$q$	liquid water mixing ratio	kg/kg
$q_1$	liquid water mixing ratio in a cloudy volume	kg/kg
$q_{m0}$	liquid water mixing ratio after the first stage of mixing	kg/kg
$q_w$	liquid water content (LWC)	$\text{gm}^{-3}$



$q_{w0}$	LWC after the first stage of mixing	$\text{gm}^{-3}$
$q_v$	water vapor mixing ratio	$\text{kg/kg}$
$\tilde{q}$	normalised liquid water mixing ratio equal to normalized LWC	nd
$\tilde{q}_{\min}$	normalized equilibrium liquid water mixing ratio equal to normalized equilibrium LWC	nd
$Q$	change of square of droplet radius	$\text{m}^2$
$r$	droplet radius	$\text{m}$
$R$	$\frac{S_2}{A_2 q_1}$	nd
$R_a$	specific gas constant of moist air	$\text{J kg}^{-1} \text{K}^{-1}$
$R_v$	specific gas constant of water vapor	$\text{J kg}^{-1} \text{K}^{-1}$
$S$	$e/e_w - 1$ , supersaturation over water	nd
$S_{m0}$	supersaturation after the first stage of mixing	nd
$S_2$	supersaturation in a dry volume	nd
$\tilde{S}_{\max}$	maximal normalized supersaturation	nd
$\tilde{S}$	normalized supersaturation	nd
$T$	temperature	$\text{K}$
$T_1$	temperature in a cloud volume	$\text{K}$
$T_2$	temperature in a dry volume	$\text{K}$
$t_e$	normalized evaporation time	nd
$t$	time	$\text{s}$
$\tilde{t}$	non-dimensional time	nd
$x(\tilde{t})$	non-dimensional variable	nd
$\alpha$	parameter of the Gamma distribution	nd
$\beta$	parameter of the Gamma distribution	$\text{m}^{-1}$
$\chi$	$\gamma^{1/3}$ , non-dimensional parameter	nd
$\varepsilon$	turbulent dissipation rate	$\text{m}^2 \text{s}^{-3}$
$\gamma$	$1 + \frac{S_{m0}}{A_2 q_{m0}}$ , non-dimensional parameter	nd

$\delta q_m$	mixing ratio of liquid water required to saturate 1 kg of the cloud (or cloudy?) volume after instant mixing	nd
$\mu$	mass fraction of cloud air	nd
$\rho_a$	air density	kg m <sup>-3</sup>
$\rho_w$	density of liquid water	kg m <sup>-3</sup>
$\tau_{pr}$	phase relaxation time	s
$\tau_{mix}$	characteristic time of mixing	s
$\tau_{m0}$	time scale	s

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1076 "nd" denotes non-dimensional

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1094 **Figures**

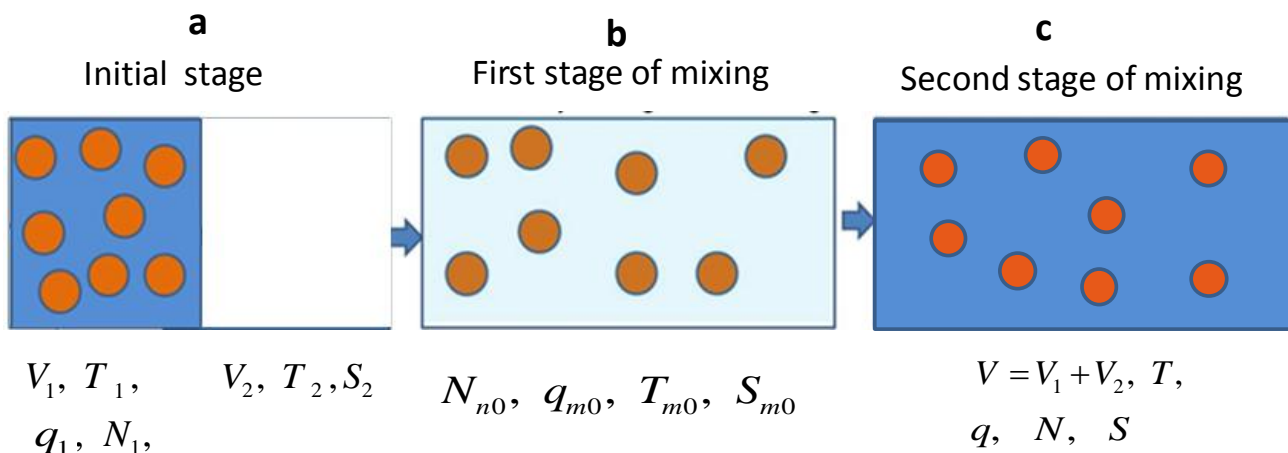
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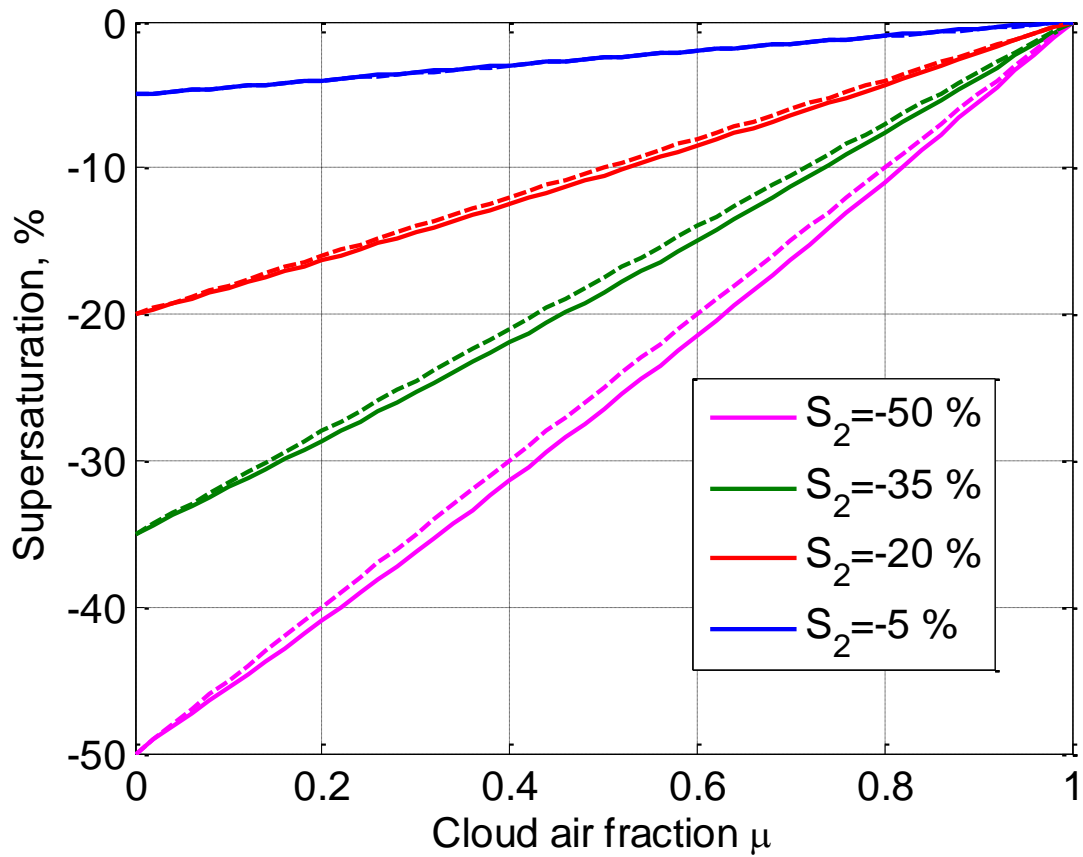
1102 **Fig. 1.** Conceptual scheme of homogeneous mixing in case of monodisperse DSD. The  
 1103 subsaturated volume of dry air is colored white, and the cloudy volumes with saturated air are  
 1104 colored dark blue. The volume forming as a result of mixing after total homogenization is  
 1105 colored light blue. Index 1 shows the initial values characterizing the initially cloudy volume.  
 1106 Index 2 denotes initial values in the droplet-free volume. Index "m0" denotes values in the  
 1107 mixing volume after the first stage of mixing.

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**Fig. 2.** Dependence of supersaturation on parameter  $\mu$ : simulation results (solid line) and an approximate linear dependence calculated using Eq. (6b) (dashed line). The initial temperatures of two volumes are  $T_1 = 8^\circ\text{C}$  and  $T_2 = 10^\circ\text{C}$ .

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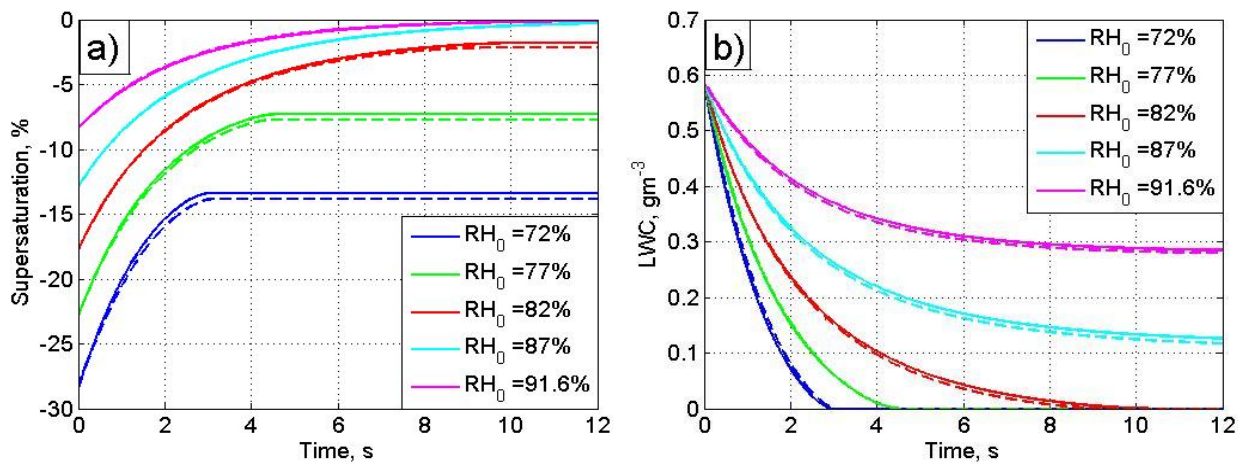
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1154 **Fig. 3.** Dependencies  $S(t)$  (a) and  $q_w(t)$  (b), calculated at different initial relative

1155 humidity  $RH_{m0}$  using closed differential equations (12,13) (solid lines) and using a parcel model

1156 (dashed lines). The calculation parameters are  $T_{m0} = 10^\circ C$ ,  $p_0 = 842$  mb,  $r_0 = 10 \mu m$ ,

1157  $N_{m0} = 140 \text{ cm}^{-3}$ ,  $q_{w0} = 0.58 \text{ gm}^{-3}$ .

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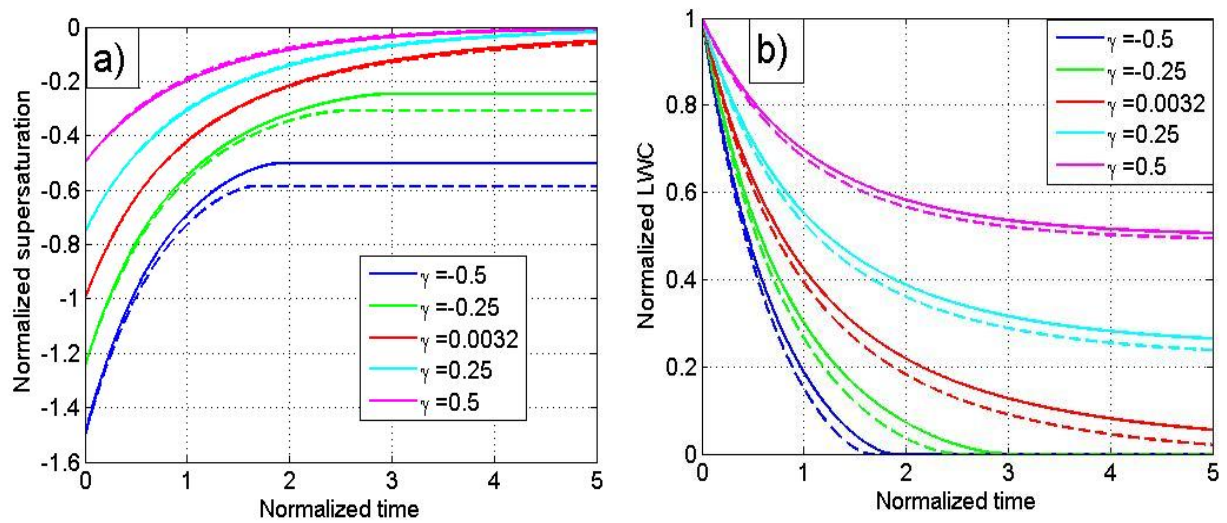
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**Fig. 4.** Universal dependencies  $\tilde{S}(\tilde{t})$  (a) and  $\tilde{q}(\tilde{t})$  (b), calculated at different values of parameter  $\gamma$  using Eqs. (17-18) (solid lines) and using a parcel model (dashed lines).

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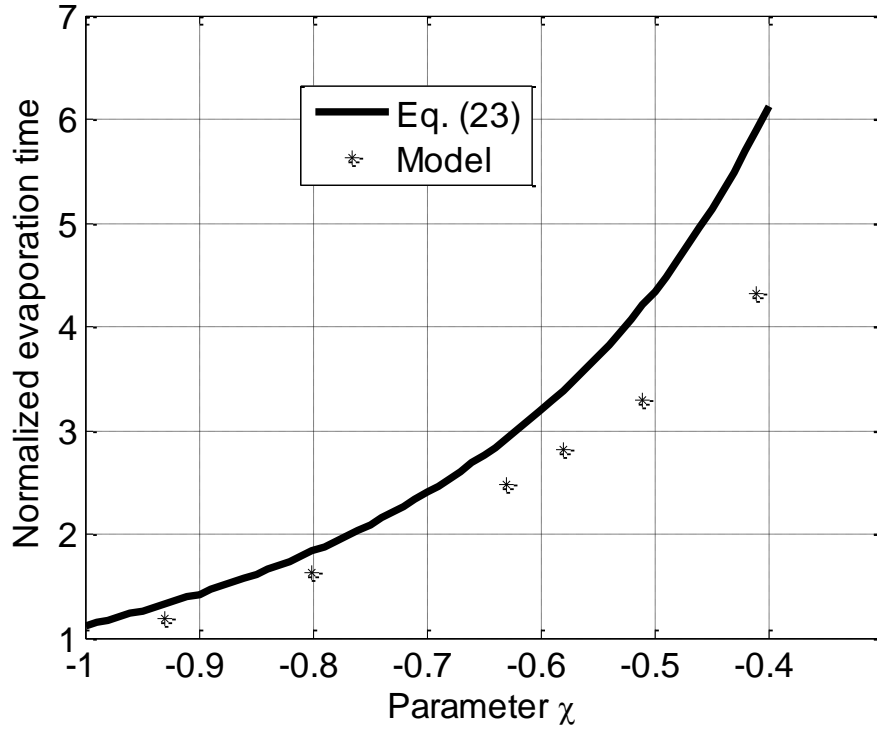
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**Fig. 5.** Dependence of the evaporation time on parameter  $\chi$ ,  $t_e(\chi)$ . Time is measured at the relaxation time scales. The values obtained using a parcel model are shown by asterisks.

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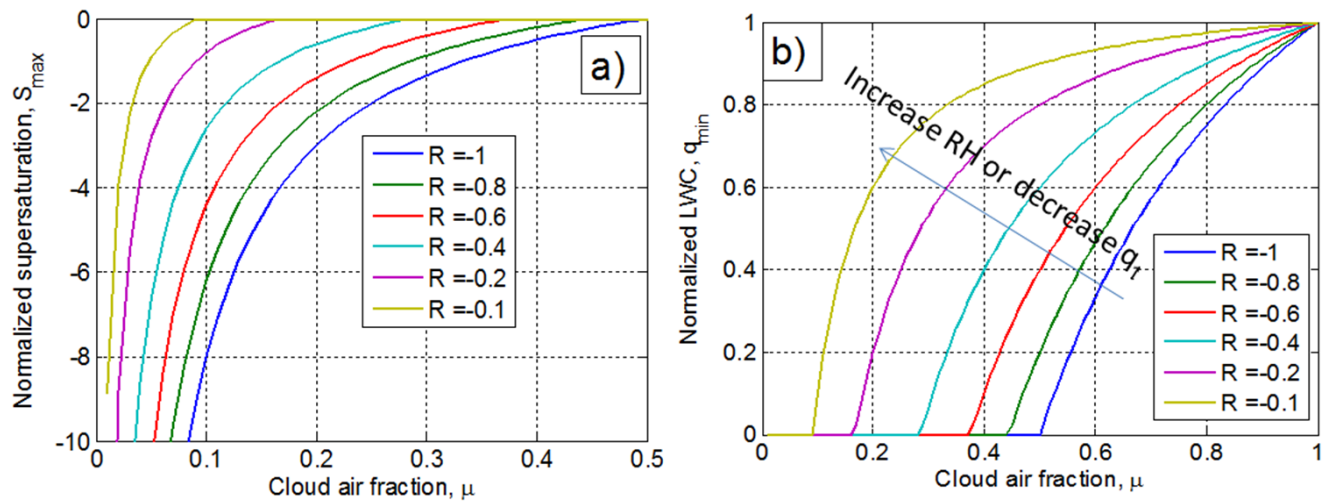
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1224 **Fig. 6.** Dependencies of normalized equilibrium supersaturation  $\tilde{S}_{\max}$  (a) and normalized

1225 equilibrium LWC (which is equal to normalized equilibrium liquid water mixing ratio)  $\tilde{S}_{\max}$  (b)

1226 on cloudy air fraction at the final stage of homogeneous mixing. Curves of different colors

1227 correspond to different values of non-dimensional parameter  $R = \frac{S_2}{A_2 q_1}$ .

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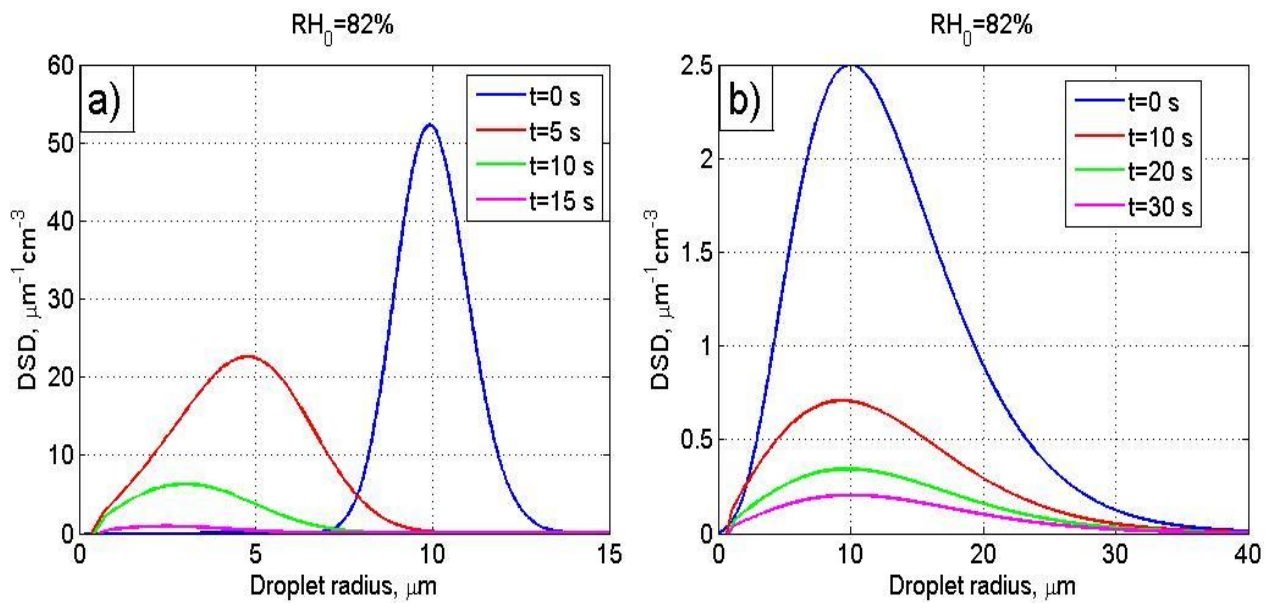
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**Fig. 7.** Time evolution of an initially narrow DSD (a) and an initially wide DSD (b). The

initial calculation parameters are the same in both examples:  $T_{m0} = 10 \text{ }^\circ\text{C}$ ,  $p = 829 \text{ mb}$ ,

$RH_{m0} = 82 \%$  and  $q_{w0} = 0.587 \text{ g/m}^3$ . The parameters of the initial Gamma distributions are

given in Tab. 3.

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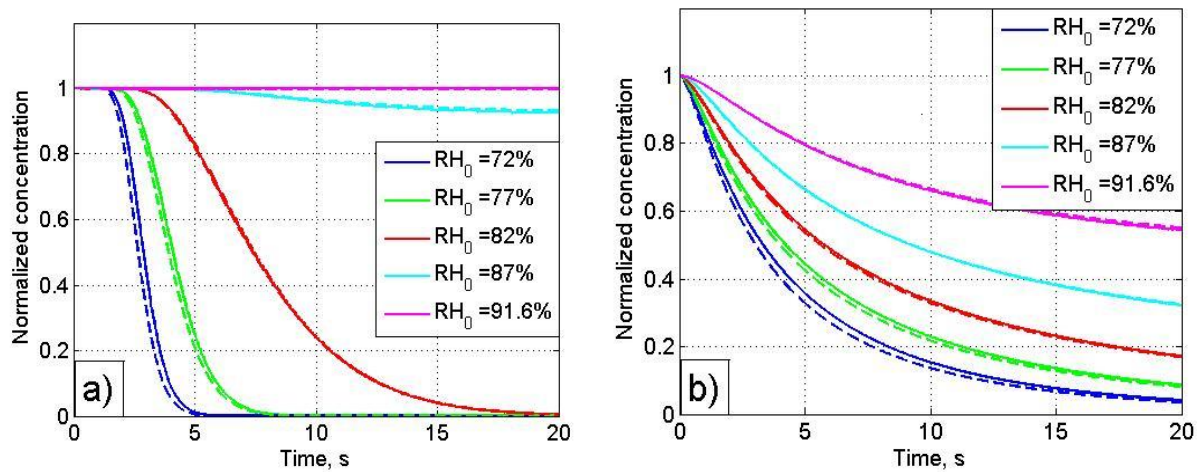
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**Fig.8.** Time dependencies of normalized droplet concentration for an initially narrow DSD

(a) and an initially wide DSD (b) at initially different values of  $RH_{m0}$  in the resulting volume.

The dependencies are calculated directly using a parcel model (solid lines) and using Eq. (33)

(dashed lines). The thermodynamic parameters are the same as in Fig.7. Parameters of the initial

DSDs are given in Tab. 3.

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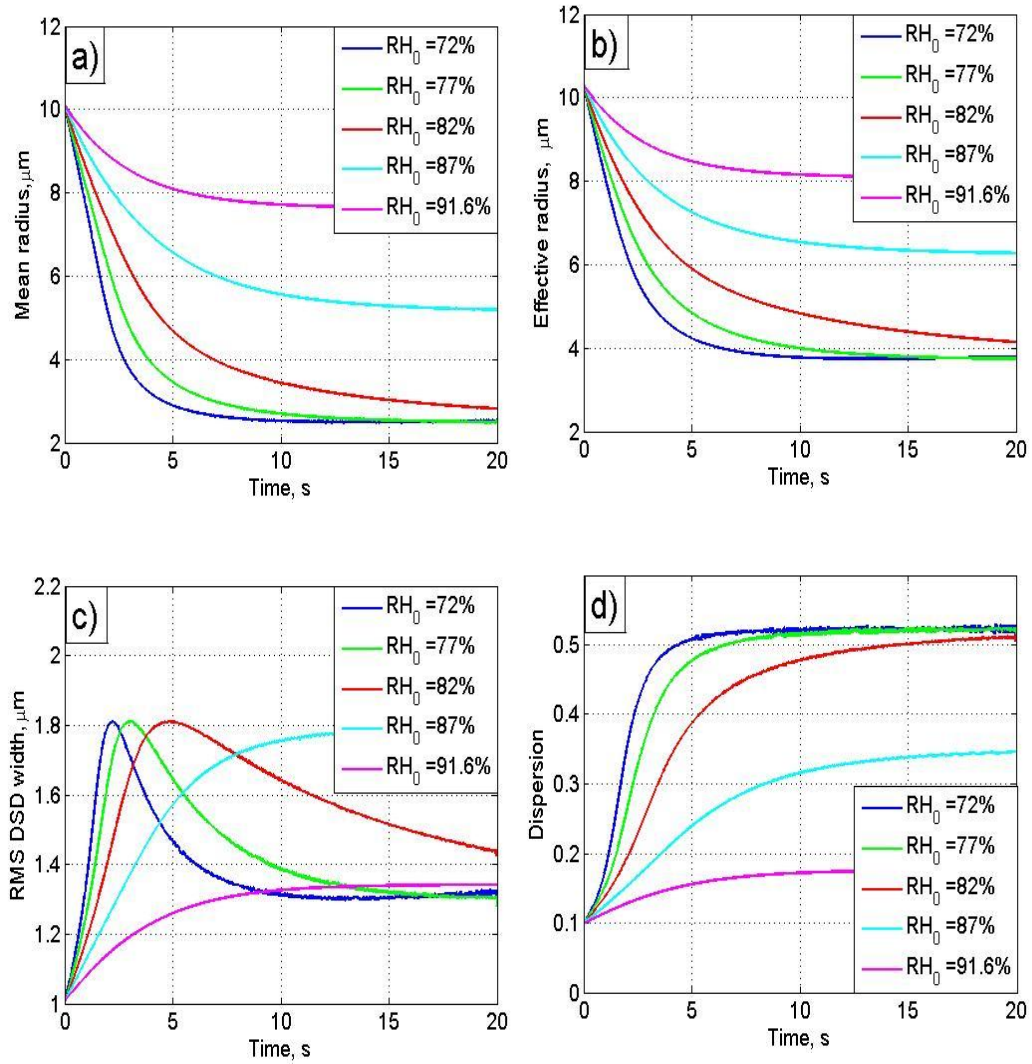
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**Fig. 9.** Dependencies of moment functions typically used for characterizing DSD shape at different values of the initial relative humidity  $RH_{m0}$  in the resulting volume. The dependencies are calculated using a parcel model for an initially narrow DSD (Tab. 3). The thermodynamic parameters are the same as in Fig. 7.

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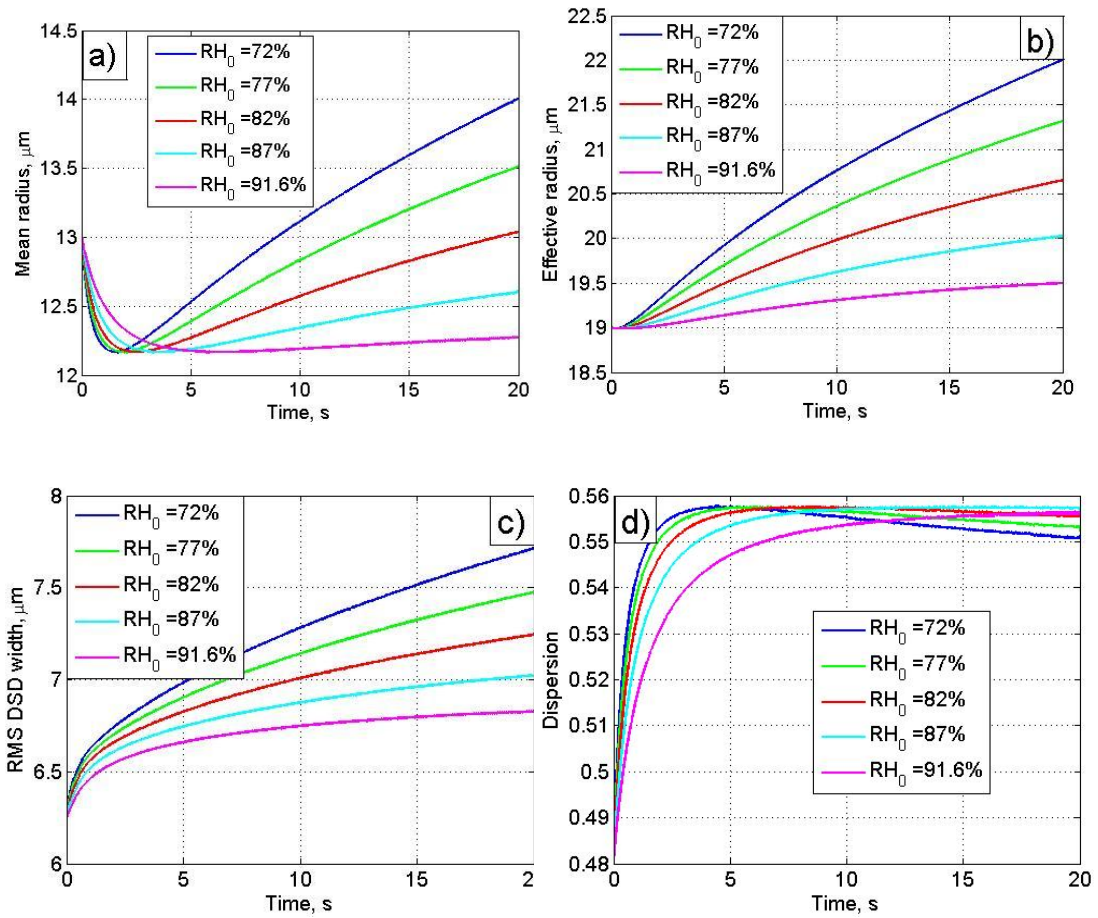
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**Fig. 10.** The same as in Fig. 9 but for an initially wide DSD.

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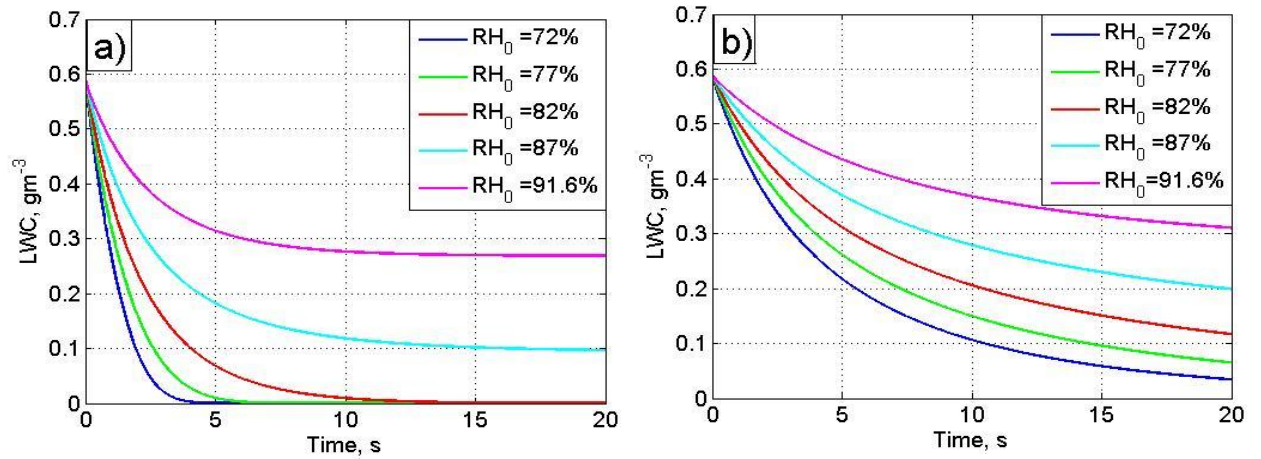
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**Fig. 11.** Time dependencies of LWC calculated using a parcel model at different values of

$RH_{m0}$  in the resulting volume, for an initially narrow DSD (a) and an initially wide DSD (b).

The thermodynamic parameters are the same as in Fig.7. The parameters of the initial DSDs are

given in Tab. 3.

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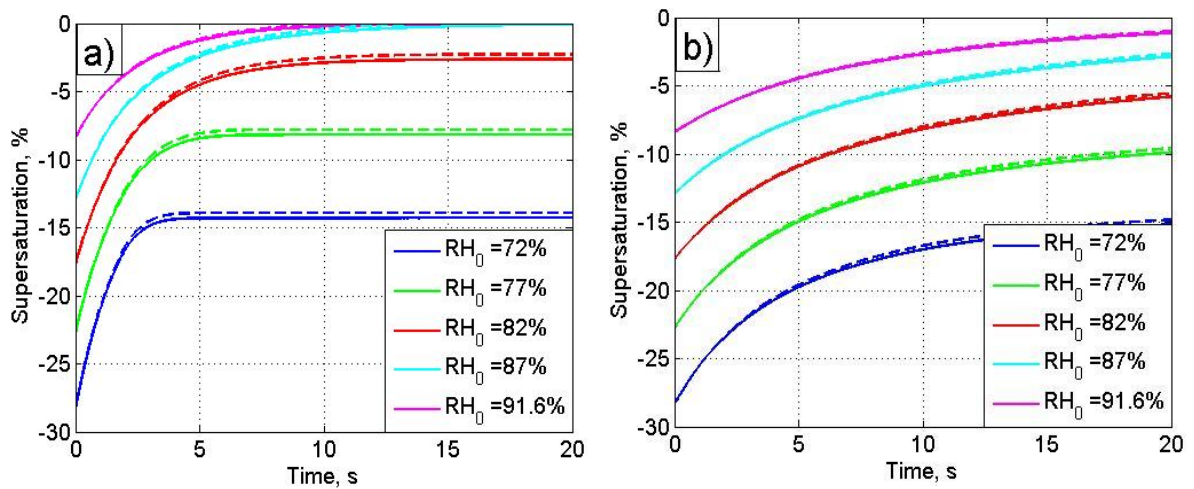
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**Fig. 12.** Time dependencies of supersaturation calculated at different values of the initial relative humidity  $RH_{m0}$  in the resulting volume, using Eq. (8) (solid lines) and Eq. (40) (dashed lines), for an initially narrow DSD (a) and an initially wide DSD (b). The thermodynamic parameters are the same as in Fig.7. The parameters of the initial DSDs are given in Tab.3.

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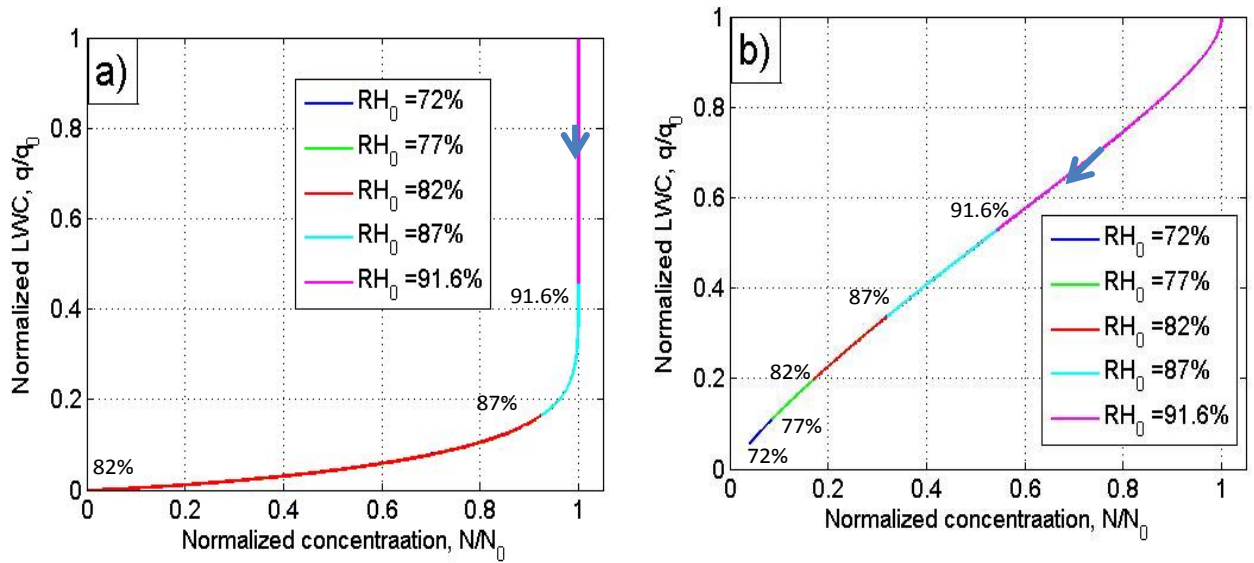
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**Fig. 13.** Dependencies of normalized LWC on the normalized number concentration of droplets calculated at different values of the initial relative humidity  $RH_{m0}$  in the resulting volume, for an initially narrow DSD (a) and an initially wide DSD (b). The thermodynamic parameters are the same as in Fig.7. The parameters of the initial DSDs are given in Tab. 3. Arrows denote the direction of increasing time.