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Theoretical analysis of mixing in liquid clouds. Part 3: Inhomogeneous mixing

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27 **Abstract**

28 An idealized diffusion-evaporation model of time-dependent mixing between a cloud
29 volume and a droplet-free volume is analyzed. The initial droplet size distribution (DSD) in
30 the cloud volume is assumed to be monodisperse. It is shown that evolution of the
31 microphysical variables and the final equilibrium state are unambiguously determined by two
32 non-dimensional parameters. The first one is the potential evaporation parameter R ,
33 proportional to the ratio of the saturation deficit to the liquid water content in the cloud
34 volume, that determines whether the equilibrium state is reached at 100% relative humidity, or
35 is characterized by a complete evaporation of cloud droplets. The second parameter Da is the
36 *Damköhler* number equal to the ratio of the characteristic mixing time to the phase relaxation
37 time. Parameters R and Da determine the type of mixing.

38 The results are analyzed within a wide range of values of R and Da . It is shown that there
39 is no pure homogeneous mixing, since the first mixing stage is always inhomogeneous. The
40 mixing type can change during the mixing process. Any mixing type leads to formation of a tail
41 of small droplets in DSD and, therefore, to DSD broadening that depends on Da . At large Da ,
42 the final DSD dispersion can be as large as 0.2. The total duration of mixing varies from
43 several to one hundred phase relaxation time periods, depending on R and Da .

44 The definitions of homogeneous and inhomogeneous types of mixing are reconsidered and
45 clarified, enabling a more precise delimitation between them. The paper also compares the
46 results obtained with those based on the classic mixing concepts.

47 **Keywords:** homogeneous and inhomogeneous mixing, turbulent diffusion, droplet
48 evaporation

49

50 1. Introduction

51 Cloud physics typically investigates two types of turbulent mixing: homogeneous and
 52 extremely inhomogeneous (e.g. Burner and Brenguier, 2006; Andrejczuk et al., 2009; Devenish
 53 et al., 2012; Kumar et al., 2012). The concept of extremely inhomogeneous mixing in clouds
 54 was introduced by Latham and Reed (1977); Baker and Latham (1979), Baker et al. (1980) and
 55 Blyth et al. (1980). According to this concept, mixing of cloud air and sub-saturated air from
 56 cloud surrounding results in complete evaporation of a fraction of cloud droplets, whereas size
 57 of other droplets remain unchanged. The studies of extremely inhomogeneous mixing were
 58 closely related to investigation of different mechanisms underlying enhanced growth of cloud
 59 droplets and warm precipitation formation (Baker et al., 1980; Baker and Latham, 1982). The
 60 concept of homogeneous mixing suggests that all the droplets partially evaporate, so the liquid
 61 water content decreases while the droplet concentration remains unchanged (Lehmann et al.,
 62 2009; Pt1). The significance of the concepts of homogeneous and inhomogeneous mixing goes
 63 far beyond formation of large-sized droplets. In fact, these concepts are closely related to the
 64 mechanisms involved in formation of droplet size distributions (DSD) in clouds and to the
 65 description of this formation in numerical cloud models. A detailed analysis of the classical
 66 concepts of homogeneous and extremely inhomogeneous mixing is given by Korolev et al.
 67 (2016, hereafter Pt1).

68 Mixing in clouds includes two processes: mechanical mixing caused by turbulent diffusion
 69 and droplet evaporation accompanied by increasing relative humidity. The relative contribution
 70 of these processes can be evaluated by comparison of two characteristic time scales: the
 71 characteristic mixing time scale $\tau_{mix} \sim L^{2/3} \varepsilon^{-1/3}$ (where L is the characteristic linear scale of an
 72 entrained volume and ε is the dissipation rate of turbulent kinetic energy) and the time of
 73 phase relaxation $\tau_{pr} = (4\pi \mathcal{D} \bar{r} N)^{-1}$ (where N is droplet concentration in a cloud volume, \bar{r} is
 74 the mean droplet radius and \mathcal{D} is the diffusivity of water vapor) characterizing the response of
 75 the droplet population to changes in humidity (the list of notations is given in Appendix). The

76 choice of the phase relaxation time as the characteristic time scale of mixing is discussed by
77 Pinsky et al. (2016) (hereafter referred to as Pt. 2) and will be further elaborated below.

78 Mixing is considered homogeneous if $\tau_{mix} / \tau_{pr} \ll 1$. At the first stage of mixing, the initial
79 gradients of the microphysical and thermodynamic variables rapidly decrease to zero. By the
80 end of this stage, the fields of temperature, humidity (hence, the relative humidity, RH) and
81 droplet concentration are spatially homogenized and all the droplets within the mixing volume
82 experience the same saturation deficit. During the relatively lengthy second stage, droplets
83 evaporate and increase the relative humidity in the volume. It was shown that homogeneous
84 mixing takes place at scales below about 0.5 m (Pt. 2)

85 At spatial scales larger than ~ 0.5 m, $\tau_{mix} / \tau_{pr} > 1$ and the spatial gradients of RH remain for
86 a long time. Consequently, droplets within the mixing volume experience different
87 subsaturations, thus the mixing is considered inhomogeneous. At $\tau_{mix} / \tau_{pr} \gg 1$, the mixing is
88 considered extremely inhomogeneous.

89 According to the classical conceptual scheme, during the first stage of extremely
90 inhomogeneous mixing a fraction of droplets is transported into the droplet-free entrained
91 volume and evaporates completely. The evaporation continues until the evaporating droplets
92 saturate the initially droplet-free volume. At the second stage, turbulent mixing between the
93 cloud volume and the initially droplet-free (but already saturated) volume homogenizes the
94 gradients of droplet concentration and other quantities. Since both volumes are saturated,
95 mixing does not affect droplet sizes. As a result, the final (equilibrium) state is characterized by
96 the relative humidity $RH=100\%$ and the DSD shape similar to that before mixing, but with a
97 lower droplet concentration. The same result (a decrease in droplet concentration but
98 unchanged droplet size) is expected in cases of both monodisperse and polydisperse initial
99 DSD. Since the DSD shape does not change, the characteristic droplet sizes (i.e. the mean

100 square radius, the mean volume radius and the effective radius) do not change either in the
101 course of extremely inhomogeneous mixing.

102 Thus, according to the classical concepts, the final equilibrium state with RH=100% is
103 reached either by a partial evaporation of all droplets (homogeneous mixing) or a total
104 evaporation of a certain portion of droplets that does not affect the remaining droplets
105 (extremely inhomogeneous mixing) (Lehmann et al., 2009; Pt1).

106 In analyses of in-situ measurements, the observed data are usually compared with those
107 expected at the final state of mixing as assumed by the classical mixing concepts. If droplet
108 concentration decreases without a corresponding change in the characteristic droplet radius,
109 the mixing is considered “extremely inhomogeneous.” If the characteristic droplet radius
110 decreases with an increase of the dilution level while droplet concentration decreases
111 insignificantly, the mixing is identified as “homogeneous.” If both the characteristic droplet
112 radius and the droplet concentration change, the mixing is considered as "intermediate".
113 Quantitative evaluations of the microphysical processes specific for intermediate mixing
114 remain largely uncertain.

115 As was discussed in Pt 2, the final states of mixing suggested by the classical concepts are
116 only hypothetical. To understand the essence of the final equilibrium states of mixing and
117 evaluate the time needed to reach them, it is necessary to consider the time evolution of DSD in
118 the course of mixing process. Time-dependent process of homogeneous mixing was analyzed
119 in Pt. 2. It was shown that in important cases of wide polydisperse initial DSDs, the final state
120 substantially differs from that hypothesized by the classical concepts.

121 In this study, which is a Pt 3 of the set of studies, we analyze the time-dependent process of
122 inhomogeneous mixing. The structure of the paper is as follows. The main concept and the
123 basic equations for time-dependent inhomogeneous mixing are described in Section 2. Analysis
124 of non-dimensional diffusion-evaporation equations is presented in Section 3. The design and
125 the results of simulations of non-homogeneous mixing are outlined in Sections 4 and 5. A

126 discussion clarifying the concepts of homogeneous and inhomogeneous mixing is presented in
127 concluding Section 6.

128

129 **2. The main concept and the basic equations**

130 During mixing of cloud volume and entrained air volume, the following two processes
131 determine the change of the microphysical and thermodynamical variables: turbulent diffusion
132 resulting in mechanical smoothening of the gradients of temperature, water vapor and droplet
133 concentration, and droplet evaporation accompanied by phase transformation. In this study,
134 inhomogeneous mixing is investigated based on the analysis and solution of a 1D diffusion-
135 evaporation equation. To our knowledge, the idea of using a diffusive model of turbulent
136 mixing to describe the mixing process was first proposed by Baker and Latham (1982). A
137 diffusion-evaporation equation was also analyzed by Jeffery and Reisner (2006). In order to get
138 a more precise understanding of the physics of mixing process the analysis is performed under
139 the following main simplifying assumptions:

140 a) turbulent mixing is analyzed neglecting vertical motions of mixing volumes, droplet
141 collisions and droplet sedimentation.

142 b) the total mixing volume is assumed adiabatic.

143 c) mixing is assumed to take place only along the x -direction, i.e. a 1D task is considered;

144 d) the initial DSD in the cloud volume is assumed monodisperse.

145 Other assumptions and simplifications are discussed below.

146

147 A schematic illustration of the initial conditions used in the study is shown in **Figure 1**.
148 Two air volumes are assumed to mix: a cloud volume (left) and a droplet-free volume (right),
149 each having the linear size of $L/2$. The value of L is assumed within the range of several tens
150 to a few hundred meters. The mixing starts at $t = 0$. The cloud volume is initially saturated
151 $S_1 = 0$, the initial droplet concentration is N_1 and the initial liquid water mixing ratio is

152 $q_1 = \frac{4\pi\rho_w}{3\rho_a} N_1 r_0^3$. In the droplet-free volume the initial conditions are $RH_2 < 100\%$ (i.e.
 153 $S_2 < 0$), $N_2 = 0$ and $q_2 = 0$. Therefore, the initial profiles of these quantities along the x -axis
 154 are step functions

$$155 \quad N(x, 0) = \begin{cases} N_1 & \text{if } 0 \leq x < L/2 \\ 0 & \text{if } L/2 \leq x < L \end{cases} \quad (1a)$$

$$156 \quad S(x, 0) = \begin{cases} 0 & \text{if } 0 \leq x < L/2 \\ S_2 & \text{if } L/2 \leq x < L \end{cases} \quad (1b)$$

$$157 \quad q(x, 0) = \begin{cases} q_1 & \text{if } 0 \leq x < L/2 \\ 0 & \text{if } L/2 \leq x < L \end{cases} \quad (1c)$$

158 The initial profile of droplet concentration is shown in Fig. 1. In this study, averaged equations
 159 are used. We do not consider mixing at scales below several millimeters. At the scales of
 160 averaging, there exist clear definitions of droplet concentration, supersaturation and other "macro
 161 scale" quantities. The mixing is assumed to be driven by isotropic turbulence within the inertial
 162 sub-range where the Richardson's law is valid. Accordingly, turbulent diffusion (turbulent
 163 mixing) is described by a 1D equation of turbulent diffusion with a turbulent coefficient K . The
 164 turbulent coefficient is evaluated as proposed by Monin and Yaglom (1975)

165

$$166 \quad K(L) = C\varepsilon^{1/3}L^{4/3} \quad (2)$$

167 In Eq. (2), C is a constant. Eq. (2) is valid in case turbulent diffusion is considered, i.e. at scales
 168 where molecular diffusion can be neglected.

169 Since the total mixing volume is adiabatic, the fluxes of different quantities through the left
 170 and right boundaries of the volume are equal to zero at any time instance, i.e.

$$171 \quad \frac{\partial N(0,t)}{\partial x} = \frac{\partial N(L,t)}{\partial x} = 0; \quad \frac{\partial q(0,t)}{\partial x} = \frac{\partial q(L,t)}{\partial x} = 0; \quad \frac{\partial q_v(0,t)}{\partial x} = \frac{\partial q_v(L,t)}{\partial x} = 0 \quad (3)$$

172 where q_v is the water vapor mixing ratio.

173 During mixing, droplets in the mixing volume experience different subsaturations,
 174 therefore, the initially monodisperse DSD will become polydisperse. The droplets that were

175 transported into the initially droplet-free volume will undergo either partial or complete
 176 evaporation. The evaporation leads to a decrease in both droplet size and droplet concentration.

177 The basic system of equations that describes the processes of diffusion and of evaporation
 178 which occur simultaneously is to be derived. The first equation is written for value Γ defined
 179 as

$$180 \quad \Gamma = S + A_2 q \quad (4)$$

181 This value is conservative in a moist adiabatic process, i.e. it does not change during phase
 182 transitions (Pinsky et al., 2013, 2014). In Eq. (4), the coefficient $A_2 = \frac{1}{q_v} + \frac{L_w^2}{c_p R_v T^2}$ is a weak

183 function of temperature that changes by $\sim 10\%$ when temperatures change by $\sim 10^\circ\text{C}$ (Pinsky et
 184 al., 2013). In this study, it is assumed that $A_2 = \text{constant}$. In Eq. (4), $q = \frac{4\pi\rho_w}{3\rho_a} \int_0^\infty r^3 f(r) dr$ is the

185 liquid water mixing ratio and $f(r)$ is the DSD. The quantity Γ obeys the diffusion equation

$$186 \quad \frac{\partial\Gamma(x,t)}{\partial t} = K \frac{\partial^2\Gamma(x,t)}{\partial x^2} \quad (5)$$

187 with the boundary conditions $\frac{\partial\Gamma(0,t)}{\partial x} = \frac{\partial\Gamma(L,t)}{\partial x} = 0$ and the initial profile at $t = 0$

$$188 \quad \Gamma(x,0) = \begin{cases} A_2 q_1 & \text{if } 0 \leq x < L/2 \\ S_2 & \text{if } L/2 \leq x < L \end{cases} \quad (6)$$

189 Therefore, function $\Gamma(x,0)$ is positive in the left volume, and negative in the right volume.

190 Since Γ does not depend on phase transitions, Eq. (5) can be solved independently of other
 191 equations. The solution of Eq. (5) with initial conditions (6) is (Polyanin and Zaitsev, 2004)

$$192 \quad \Gamma(x,t) = \sum_{n=0}^{\infty} a_n \exp\left(-\frac{Kn^2\pi^2 t}{L^2}\right) \cos\left(\frac{n\pi x}{L}\right) = \quad (7)$$

$$\frac{1}{2}(S_2 + A_2 q_1) + (A_2 q_1 - S_2) \sum_{n=1}^{\infty} \frac{\sin(n\pi/2)}{n\pi/2} \exp\left(-\frac{Kn^2\pi^2 t}{L^2}\right) \cos\left(\frac{n\pi x}{L}\right)$$

193 where the Fourier coefficients of expanding the step function (6) are

$$194 \quad a_0 = \frac{1}{2}(A_2q_1 + S_2) \quad (8a)$$

$$195 \quad a_n = (A_2q_1 - S_2) \frac{\sin(n\pi/2)}{n\pi/2}, \quad n = 1, 2, \dots \quad (8b)$$

196 An example of spatial dependencies of $\Gamma(x, t)$ at different time instances during the mixing is
 197 shown in **Figure 2**. One can see a decrease in the initial gradients and a tendency to
 198 establishing a horizontally uniform value of Γ . Since the initial volume was divided into two
 199 equal parts, the diffusion leads to formation of a constant limit value of function Γ

$$200 \quad \Gamma(x, \infty) = \frac{1}{2}(\Gamma(0, 0) + \Gamma(L, 0)).$$

201 The second basic equation is the equation for diffusional droplet growth, taken in the
 202 following form (Pruppacher and Klett, 2007)

$$203 \quad \frac{d\sigma}{dt} = \frac{2S}{F} \quad (9)$$

204 where $\sigma = r^2$ is the square of droplet radius and $F = \frac{\rho_w L_w^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_s(T) \mathcal{D}}$. The value of

205 coefficient F is considered constant in this study. The solution of Eq. (9) is

$$206 \quad \sigma(t) = \frac{2}{F} \int_0^t S(t') dt' + \sigma_0 \quad (10)$$

207 The third main equation describes the evolution of DSD. In the following discussion, the
 208 DSD will be presented in the form $g(\sigma)$ which is the distribution of the square of the radius.
 209 This formulation directly utilizes the property of the diffusion growth equation (9) according to
 210 which the time changes of DSD are reduced to shifting the distributions in the space of square
 211 radii, while the shape of the distribution remains unchanged. The standard DSD $f(r)$ is related
 212 to $g(\sigma)$ as $f(r) = 2r \cdot g(r^2)$.

213 The normalized condition for $g(\sigma)$ is

$$214 \quad N = \int_0^{\infty} g(\sigma) d\sigma \quad (11)$$

215 where N is the droplet concentration. Using DSD $g(\sigma)$, the liquid water mixing ratio can be
216 presented as integral

$$217 \quad q = \frac{4\pi\rho_w}{3\rho_a} \int_0^{\infty} \sigma^{3/2} g(\sigma) d\sigma \quad (12)$$

218 The 1D diffusion-evaporation equation for the non-conservative function $g(\sigma)$ can be
219 written in the form (Rogers and Yau, 1989)

$$220 \quad \frac{\partial g(\sigma)}{\partial t} = K \frac{\partial^2 g(\sigma)}{\partial x^2} - \frac{\partial}{\partial \sigma} \left(\frac{d\sigma}{dt} g(\sigma) \right) \quad (13)$$

221 where the first term on the right-hand side of Eq. (13) describes changes in the DSD due to
222 spatial diffusion, while the second term on the right-hand side describes changes in the DSD
223 due to evaporation. Substitution of Eq. (9) into Eq. (13) leads to the following equation

$$224 \quad \frac{\partial g(x,t,\sigma)}{\partial t} = K \frac{\partial^2 g(x,t,\sigma)}{\partial x^2} - \frac{2S(x,t)}{F} \frac{\partial g(x,t,\sigma)}{\partial \sigma} \quad (14)$$

225 To close Eq. (14), Eq. (4) should be used in the form

$$226 \quad S(x,t) = \Gamma(x,t) - A_2 q(x,t) \quad (15)$$

227 where $q(x,t)$ is calculated according to Eq. (12). Eqs. (12, 14, 15) constitute a closed set of
228 equations allowing calculation of $g(x,t,\sigma)$.

229 To proceed to the equations for DSD moments, let us define a moment of DSD $g(\sigma)$ of
230 order α as

$$231 \quad m_\alpha = \overline{\sigma^\alpha} = \int_0^{\infty} \sigma^\alpha g(\sigma) d\sigma \quad (16)$$

232 Multiplying Eq. (14) by σ^α , integrating within limits $[0..\infty]$ and assuming that $\sigma^\alpha g(\sigma) \rightarrow 0$
233 when $\sigma \rightarrow \infty$, yield a recurrent formula for the DSD moments

234

$$235 \quad \frac{\partial m_\alpha(x,t)}{\partial t} = K \frac{\partial^2 m_\alpha(x,t)}{\partial x^2} + \alpha \frac{2S}{F} m_{\alpha-1}(x,t) \quad (17)$$

236 Eq. (17) provides a recurrent relationship between the DSD moments of different orders. This
 237 relationship was discussed by Pinsky et al.'s (2014) while analyzing diffusion growth in an
 238 ascending adiabatic parcel.

239 In particular, the equation for the liquid water mixing ratio that is a moment of the order of

240 $\alpha = \frac{3}{2}$ can be written as

$$241 \quad \frac{\partial q(x,t)}{\partial t} = K \frac{\partial^2 q(x,t)}{\partial x^2} + \frac{4\pi\rho_w N(x,t)\bar{r}(x,t)}{F\rho_a} S(x,t) \quad (18)$$

242 where the mean radius $\bar{r}(x,t) = \frac{m_{1/2}}{m_0}$.

243 In the general case, Eq. (18) is not closed, since concentration $N(x,t)$ and $\bar{r}(x,t)$ are unknown
 244 functions of time and spatial coordinates.

245 The characteristic time of evaporation and of supersaturation change is the phase relaxation
 246 time (Korolev and Mazin, 2003)

$$247 \quad \tau_{pr} = \frac{\rho_a F}{4\pi\rho_w A_2 N \bar{r}} \quad (19)$$

248 Using Eq. (19), Eq. (18) can be rewritten as

$$249 \quad \begin{aligned} \frac{\partial q(x,t)}{\partial t} &= K \frac{\partial^2 q(x,t)}{\partial x^2} + \frac{1}{\tau_{pr}(x,t)} \left[\frac{1}{A_2} \Gamma(x,t) - q(x,t) \right] = \\ &= K \frac{\partial^2 q(x,t)}{\partial x^2} + \frac{1}{A_2 \tau_{pr}(x,t)} S(x,t) \end{aligned} \quad (20)$$

250 From Eqs. (20) and (15), the equation for supersaturation can be written in the following simple
 251 form

$$252 \quad \frac{\partial S(x,t)}{\partial t} = K \frac{\partial^2 S(x,t)}{\partial x^2} - \frac{S(x,t)}{\tau_{pr}(x,t)} \quad (20a)$$

253 Eqs. (20) and (20a) show that changes in the microphysical variables are determined by the rate
 254 of spatial diffusion (the first term on the right-hand side of these equations) and of evaporation
 255 (the second term on the right-hand side).

256

257 **3. Analysis of non-dimensional equations**

258 Spatial diffusion and evaporation depend on many parameters. It is the best to start the
 259 analysis from the basic equation system presented in a non-dimensional form. A time scale
 260 corresponding to the initial phase relaxation time in a cloud volume can be defined as

$$261 \quad \tau_0 = \frac{\rho_a F}{4\pi\rho_w A_2 N_1 r_0} \quad (21)$$

262 and the non-dimensional time is $\tilde{t} = t / \tau_0$. Other non-dimensional parameters to be used are:

263 the non-dimensional phase relaxation time

$$264 \quad \tilde{\tau}_{pr} = \tau_{pr} / \tau_0 = \frac{N_1 r_0}{N(\tilde{x}, \tilde{t}) \bar{F}(\tilde{x}, \tilde{t})}, \quad (22a),$$

265 the normalized liquid water mixing ratio which is equal to the normalized liquid water content

$$266 \quad \tilde{q} = \frac{q}{q_1}, \quad (22b),$$

267 the normalized supersaturation

$$268 \quad \tilde{S} = \frac{S}{A_2 q_1} \quad (22c),$$

269 the non-dimensional conservative function

$$270 \quad \tilde{\Gamma} = \frac{\Gamma}{A_2 q_1}, \quad (22d),$$

271 the normalized square of droplet radius

$$272 \quad \tilde{\sigma} = \frac{\sigma}{r_0^2}, \quad (22e),$$

273 the normalized droplet concentration

$$274 \quad \tilde{N} = N / N_1 \quad (22f)$$

275 and the non-dimensional DSD

$$276 \quad \tilde{g}(\tilde{\sigma}) = \frac{r_0^2}{N_1} g(\sigma) \quad (22g).$$

277 with normalization $\tilde{N} = \int_0^1 \tilde{g}(\tilde{\sigma}) d\tilde{\sigma}$. The definition (22g) means that the integral of a non-

278 dimensional initial size distribution over the normalized square radius is equal to unity.

279 The non-dimensional distance and the non-dimensional time are defined as

$$280 \quad \tilde{x} = x/L; \quad \tilde{t} = t/\tau_0 \quad (22h)$$

281 A widely used non-dimensional parameter showing the comparative rates of diffusion and
282 evaporation is the Damköhler number:

$$283 \quad Da = \frac{\tau_{mix}}{\tau_0} = \frac{L^2}{K\tau_0} \quad (23)$$

284 where

$$285 \quad \tau_{mix} = \frac{L^2}{K} \quad (24)$$

286 is the characteristic time scale of mixing. Using the non-dimensional parameters listed above,

287 Eq. (20) can be rewritten in a non-dimensional form as

288

$$289 \quad \frac{\partial \tilde{q}(\tilde{x}, \tilde{t})}{\partial \tilde{t}} = \frac{1}{Da} \frac{\partial^2 \tilde{q}(\tilde{x}, \tilde{t})}{\partial \tilde{x}^2} + \frac{1}{\tilde{\tau}_{pr}(\tilde{x}, \tilde{t})} [\tilde{\Gamma}(\tilde{x}, \tilde{t}) - \tilde{q}(\tilde{x}, \tilde{t})] =$$

$$\frac{1}{Da} \frac{\partial^2 \tilde{q}(\tilde{x}, \tilde{t})}{\partial \tilde{x}^2} + \frac{1}{\tilde{\tau}_{pr}(\tilde{x}, \tilde{t})} \tilde{S}(\tilde{x}, \tilde{t}) \quad (25)$$

290 where

$$291 \quad \tilde{q}(\tilde{x}, \tilde{t}) = \frac{N(\tilde{x}, \tilde{t}) \overline{\sigma^{3/2}}}{N_1 r_0^3} = \int_0^\infty \tilde{\sigma}^{3/2} \tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma}) d\tilde{\sigma} \quad (26)$$

292 The initial conditions and the boundary conditions should be rewritten in a non-dimensional

293 form as well. For instance, the normalized initial condition for the non-dimensional function

294 $\tilde{q}(\tilde{x}, 0)$ can be derived from Eqs. (1c) and (22b)

$$295 \quad \tilde{q}(\tilde{x}, 0) = \begin{cases} 1 & \text{if } 0 \leq \tilde{x} < 1/2 \\ 0 & \text{if } 1/2 \leq \tilde{x} < 1 \end{cases} \quad (27)$$

296 The solution for $\tilde{\Gamma}(\tilde{x}, \tilde{t})$ obtained by a normalization of solution (7) is

$$297 \quad \tilde{\Gamma}(\tilde{x}, \tilde{t}) = \frac{1}{2}(1+R) + (1-R) \sum_{n=1}^{\infty} \frac{\sin(n\pi/2)}{n\pi/2} \exp\left(-\frac{n^2\pi^2\tilde{t}}{Da}\right) \cos(n\pi\tilde{x}), \quad (28)$$

298 where

$$299 \quad R = \frac{S_2}{A_2 q_1} \quad (29)$$

300 is a non-dimensional parameter referred to, hereafter, as a potential evaporation parameter
 301 (PEP). The PEP is proportional to the ratio of the amount of water vapour that should
 302 evaporate in order to saturate the initially droplet-free volume (that is determined by S_2) to the
 303 initial available liquid water q_1 in the cloud volume. The solution of Eq. (28) at $t \rightarrow \infty$ depends
 304 only on parameter R .

$$305 \quad \tilde{\Gamma}(\tilde{x}, \infty) = \frac{1}{2}(1+R) \quad (30)$$

306 The importance of PEP that determines a possible final state was illustrated in Pt. 1. PEP is
 307 also the sole parameter enabling calculation of the normalized mixing diagram for
 308 homogeneous mixing (Pt. 2). In this study, we consider cases when $R < 0$ since $S_2 < 0$, i.e.
 309 when droplets can only evaporate in the course of mixing.

310 The solution of Eq. (25) and the type of mixing depends on the values of two non-
 311 dimensional parameters, namely, Da and R . Thus, when $R = \frac{S_2}{A_2 q_1} < -1$, $\tilde{\Gamma}(\tilde{x}, \infty) < 0$. It means

312 that the initially droplet-free volume V_2 is too dry and all the droplets in the mixing volume
 313 evaporate completely. At the final equilibrium state $RH < 100\%$, i.e. $S(x, \infty) < 0$. If

314 $R = \frac{S_2}{A_2 q_1} > -1$, $\tilde{\Gamma}(\tilde{x}, \infty) > 0$. This means that the mixed volume in the final state contains

315 droplets, i.e. the mixing leads expands the volume with droplets, i.e. the cloud volume. At the

316 final equilibrium state, $RH = 100\%$ (i.e. $S(x, \infty) = 0$). The case when $|R| = \left| \frac{S_2}{A_2 q_1} \right| = |\tilde{S}_2| \ll 1$
 317 corresponds to either RH close to 100% (i.e. S_2 is close to zero) (this case corresponds to the
 318 degenerated case considered in Pt. 1), and/or to the case when the liquid water mixing ratio in
 319 the cloud volume is large. In case $|R| \ll 1$, the second term on the right-hand side of Eq. (25) is
 320 much smaller than the first term, and the mixing is driven by turbulent diffusion only.

321 In case $Da \rightarrow 0$ (often considered as homogeneous mixing), at the beginning of the mixing
 322 the diffusion term is much larger than the evaporation term, the second term on the right-hand
 323 side of Eq. (25). As mixing proceeds, within a short time period the total homogenization of all
 324 the variables in the mixing volume is established and all the spatial gradients become equal to
 325 zero. At this time instance, the first term on the right-hand side becomes equal to zero, and the
 326 second term on the right-hand side of Eq. (25), describing droplet evaporation, becomes
 327 dominant. Thus, the analysis of the Eq. (25) shows that mixing consists of two stages. The first
 328 mixing stage is a short stage of inhomogeneous mixing and the longer second stage of
 329 homogeneous mixing. The evolution of the microphysical variables during homogeneous
 330 mixing is described in detail in Pt. 2.

331 $Da \rightarrow \infty$ corresponds to extremely inhomogeneous mixing, according to the classic
 332 concept. In this case, the diffusion term is much smaller than the evaporation term, so
 333 evaporation takes place under significant spatial gradients of RH . At $Da = \infty$, the adjacent
 334 volumes do not mix at all and remain separated. This equivalent to existence of two
 335 independent adiabatic volumes. Another interpretation of the limiting case $Da = \infty$ is an
 336 infinite fast droplet evaporation. Both scenarios at $Da \rightarrow \infty$ indicate simplifications in the
 337 definition of the extremely inhomogeneous mixing. At intermediate values of Da , mixing is
 338 inhomogeneous, when both turbulent diffusion and evaporation contribute simultaneously to
 339 formation of the DSD.

340 Using Eq. (14) and normalization (22f), the equations for the non-dimensional size
 341 distribution can be written as

$$342 \frac{\partial \tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma})}{\partial \tilde{t}} = \frac{1}{Da} \frac{\partial^2 \tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma})}{\partial \tilde{x}^2} + \frac{2}{3} [\tilde{\Gamma}(\tilde{x}, \tilde{t}) - \tilde{q}(\tilde{x}, \tilde{t})] \frac{\partial \tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma})}{\partial \tilde{\sigma}} \quad (31)$$

343

344 Eq. (31) is solved with the following initial conditions

345

$$346 \tilde{g}(\tilde{x}, 0, \tilde{\sigma}) = \begin{cases} \delta(\tilde{\sigma} - 1) & \text{if } 0 \leq \tilde{x} < 1/2 \\ 0 & \text{if } 1/2 \leq \tilde{x} \leq 1 \end{cases} \quad (32)$$

347 where $\delta(\tilde{\sigma} - 1)$ is a delta function.

348

349 **Table 1** presents the list of all the non-dimensional variables used in this study and the
 350 ranges of their variation. It is shown that six parameters determining the geometrical and
 351 microphysical properties of mixing can be reduced to two non-dimensional parameters, which
 352 enables a more efficient analysis of mixing. The ranges of parameter variations in Tab. 1
 353 correspond to the simplifications used in the study (the initial DSD is monodisperse
 354 and $RH \leq 100\%$).

355

356 **4. Design of simulations**

357 ***Damköhler number Da in clouds***

358 The characteristic mixing time τ_{mix} can be evaluated using Eqs. (2) and (24)

$$359 \tau_{mix} = \frac{1}{C} \varepsilon^{-1/3} L^{2/3} \quad (33)$$

360 There is significant uncertainty regarding the evaluation of τ_{mix} and Da in clouds, which is
 361 largely related to the choice of coefficient C in expression (33). These values differ in different
 362 studies: $C = 10$ (Jeffery and Reisner, 2006); $C = 1$ (Lehmann et al., 2009) and $C \approx 0.2$ (
 363 Monin and Yaglom, 1975) and Boffetta and Sokolov (2002).

364 According to Lehmann et al. (2009), the values of Da in clouds of different types range
 365 from to 0.1 to several hundred. Thus, estimation of Da in clouds may vary within a wide range
 366 up to a few orders of magnitude. Da values of in stratocumulus clouds can be similar or even
 367 higher than those in cumulus clouds, since both τ_{mix} and τ_{pr} in stratiform clouds are larger than
 368 in cumulus clouds.

369 In our simulations, we compare the evolution of the microphysical parameters within a
 370 wide range of Da (from 1 up to 500) and of R (from -1.5 up to -0.1). $Da = 1$ represents the
 371 case closest to homogeneous mixing, while $Da = 500$ indicates extremely inhomogeneous
 372 mixing.

373

374 ***Numerical method***

375 Calculations were performed using MATLAB solver PDEPE. We solve the equation
 376 system (31) for normalized DSD $\tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma}_j)$ with the initial condition (32) and the Neumann
 377 boundary conditions

378

$$379 \quad \frac{\partial \tilde{g}(0, \tilde{t}, \tilde{\sigma}_j)}{\partial \tilde{x}} = \frac{\partial \tilde{g}(1, \tilde{t}, \tilde{\sigma}_j)}{\partial \tilde{x}} = 0 \quad (34)$$

380 where $j = 1 \dots 24$ are the bin numbers on a linear grid of square radii. The number of grid points
 381 along the \tilde{x} axis was set equal to 81.

382 In calculation of the last term on the right-hand side of Eq. (31), the normalized
 383 supersaturation \tilde{S} was calculated first using the normalized conservative equation

$$384 \quad \tilde{S}(\tilde{x}, \tilde{t}) = \tilde{\Gamma}(\tilde{x}, \tilde{t}) - \tilde{q}(\tilde{x}, \tilde{t}) \quad (35)$$

385 where $\tilde{\Gamma}(\tilde{x}, \tilde{t})$ is calculated using Eq. (28). Then, this term was formulated using Eq. (9) as

$$386 \quad \frac{2}{3} \tilde{S}(\tilde{x}, \tilde{t}) \frac{\partial \tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma}_j)}{\partial \tilde{\sigma}_j} \approx \frac{\tilde{g}\left(\tilde{x}, \tilde{t}, \tilde{\sigma}_j + \frac{2}{3} \tilde{S} \Delta \tilde{t}\right) - \tilde{g}(\tilde{x}, \tilde{t}, \tilde{\sigma}_j)}{\Delta \tilde{t}} \quad (36)$$

387 Therefore, at each time step, the DSD \tilde{g} first was shifted to the left to the value $\frac{2}{3}\tilde{S}\Delta\tilde{t}$, where
 388 $\Delta\tilde{t}$ is a small time increment chosen so that $\left|\frac{2}{3}\tilde{S}_{\max}\Delta\tilde{t}\right| \leq \frac{\Delta\tilde{\sigma}}{2}$. Next, the shifted DSD was
 389 remapped onto the fixed square radius grid $\tilde{\sigma}_j$. We used the remapping method proposed by
 390 Kovetz and Olund (1969), which conserves droplet concentration and LWC. After remapping,
 391 the differences between the new and old DSDs were recalculated. The new values of LWC then
 392 were determined using new values of DSD and Eq. (26). MATLAB utility PDEPE
 393 automatically chooses the time step needed to provide stability of calculations.

394

395 **5. Results of simulations**

396 **5.1 Full evaporation case**

397 First, we consider the case $R = -1.5$, when all the cloud water evaporates completely. This
 398 process corresponds to the cloud dissipation caused by mixing with the entrained dry air. At the
 399 final state, RH is expected to be uniform and negative over the entire mixing volume.

400 **Figure 3** shows spatial and time changes of \tilde{S} for $Da = 1, 50$ and 500 . At the final state
 401 for all the three cases $\tilde{S} = -0.25$, which is in agreement with the analytical solution of Eq. (30).
 402 The final negative value indicates that all the droplets completely evaporated during mixing. At
 403 $Da = 1$ (Fig.3ab), two stages of supersaturation evolution can be identified. The first short
 404 stage with $t < 0.4\tau_{pr}$ is the period of inhomogeneous mixing, when the gradients of RH
 405 persist. By end of the second stage of about $14\tau_{pr}$, the equilibrium state is reached. Thus, at
 406 small Da both types of mixing take place. In the cases of $Da = 50$ and $Da = 500$, the spatial
 407 gradients exit during the entire period of mixing until the equilibrium state is reached
 408 (approximately $50\tau_{pr}$ and $300\tau_{pr}$, respectively) (Fig.3cdef). Therefore, at these Da mixing is
 409 inhomogeneous during entire mixing.

410 **Figure 4** shows spatial changes (upper row) and changes in $\tilde{x}-\tilde{t}$ coordinates (lower row)
 411 of normalized LWC for the same case as in Fig.3. These diagrams demonstrate a significant
 412 difference in the evaporation rates at different Da values. Complete evaporation (LWC=0) is
 413 reached at $Da = 1, 50$ and 500 by about 12, 22 and 120 relaxation times periods , respectively.

414 Analysis of Figs. 3 and 4 allows to introduce two characteristic time periods: (1) period T_{mix}
 415 during which the spatial gradients of the microphysical parameters persist, and mixing is
 416 inhomogeneous, and (2) period T_{ev} during which droplet evaporation takes place. Both time
 417 periods are dimensionless and normalized using τ_0 . Time period T_{ev} is equal either to the time
 418 of complete droplet evaporation (when $R < -1.0$) or to the time period during which the
 419 saturation deficit in the mixing volume becomes equal to zero (or close to zero if $R > -1.0$),
 420 i.e. evaporation is actually terminated. Quantitative evaluations of T_{mix} and T_{ev} will be given in
 421 Section 5.3. At $\tilde{t} < T_{mix}$, droplets in the mixing volume experience different saturation deficits.
 422 Toward the end of time T_{mix} the saturation deficit becomes uniform over the entire mixing
 423 volume because of mechanic mixing. At $Da = 1$, the homogenization of the saturation deficit
 424 and all the microphysical variables takes place during a very short time of about $0.5\tau_{pr}$, and
 425 then the evaporation of droplets is assumed to take place under the same subsaturation
 426 conditions, so $T_{mix} \ll T_{ev}$.

427 Figs. 4a,b show that at $\tilde{t} \approx 0.35$, normalized LWC drops down from 1 to 0.4. Since the
 428 average value of the normalized LWC in the mixing volume is equal to 0.5 (see the initial
 429 condition in Eq. (27)), 20% of the droplet mass evaporates during this short inhomogeneous
 430 period. Thus, despite being quite short, inhomogeneous mixing stage plays an important role
 431 even at $Da = 1$.

432 Since at $t = 0$ the mixing volume is not spatially homogeneous by definition, there is
 433 always a period while spatial inhomogeneity exists. With increasing Da , the duration of the
 434 inhomogeneous stage increases and the duration of the homogeneous stage decreases. At

435 $Da = 500$, homogenization of the saturation deficit requires $250\tau_{pr}$, which is twice as long as
 436 the time of complete droplet evaporation, i.e. $T_{mix} \approx 2T_{ev}$. This means that at $Da = 500$, droplet
 437 evaporation takes place in the presence of the spatial gradients of supersaturation. After
 438 complete evaporation of droplets, spatial gradients of the water vapour mixing ratios remain.
 439 This kind of mixing is regarded as inhomogeneous.

440 At $Da = 50$, the time of complete evaporation is approximately equal to the time of
 441 supersaturation homogenization, i.e. $T_{mix} \approx T_{ev}$. In this case, as at $Da = 500$, the droplets
 442 experience different saturation deficit within the mixing volume, so mixing is inhomogeneous
 443 at $Da = 50$.

444 The differences in droplet evaporation at different Da can be seen in **Figure 5.**, showing
 445 the relationships between \tilde{N} and \tilde{q} plotted with a certain time increment, so that each symbol
 446 in the diagrams corresponds to a particular time instance. These symbols form curves. Each
 447 panel of Fig. 5 shows three curves corresponding to different \tilde{x} : the centre of the initially cloud
 448 volume ($\tilde{x} = 1/4$); the centre of the mixing volume ($\tilde{x} = 1/2$) and the centre of the initially
 449 droplet-free volume ($\tilde{x} = 3/4$). The directions of the time increase are shown by arrows along
 450 the corresponding curves. The initial points of the curves corresponding to $\tilde{t} = 0$ are
 451 characterized by values $\tilde{q} = 1$ and $\tilde{N} = 1$ at $\tilde{x} = 1/4$, and by values $\tilde{q} = 0$ and $\tilde{N} = 0$ at $\tilde{x} = 3/4$.

452 The behaviour of the $\tilde{N} - \tilde{q}$ relationship provides important information about mixing
 453 process. At $\tilde{t} < T_{mix}$, there are spatial gradients of \tilde{N} and \tilde{q} , i.e. \tilde{N} and \tilde{q} are different at
 454 different \tilde{x} . This means that the three curves at $\tilde{t} < T_{mix}$ do not coincide. At $\tilde{t} > T_{mix}$, the spatial
 455 gradients of \tilde{N} and \tilde{q} disappear and the three curves coincide. When the curves do not
 456 coincide, mixing is inhomogeneous, and the coincidence of the curves indicates that the mixing
 457 becomes homogeneous. In Fig. 5a and 5b ($Da = 1$ and $Da = 5$, respectively), the curves
 458 coincide at point A corresponding to time $\tilde{t} = T_{mix}$.

459 Figs. 5a,b show that at $Da = 1$ and $Da = 5$, mixing consists of two stages: inhomogeneous
 460 and homogeneous. The time instance $\tilde{t} = T_{mix}$ separates these two stages. In turn, the period of
 461 homogeneous mixing (when evaporation is spatially homogeneous) can be separated into two
 462 sub-periods. During the first sub-period, droplets evaporate only partially and \tilde{q} decreases at
 463 the same droplet concentration. This sub-period is very pronounced at $Da = 1$, when \tilde{q}
 464 decreases from about 0.4 to 0.1 at the unchanged droplet concentration. At the second sub-
 465 period, when $\tilde{q} < 0.1$, droplets evaporate completely, beginning with smaller ones, so both the
 466 droplet concentration and \tilde{q} rapidly drop to zero. At $Da = 5$ (Fig. 5b), at the stage of
 467 homogeneous evaporation (that begins at point “A”) the decrease in \tilde{q} is accompanied by a
 468 decrease in \tilde{N} .

469 At $Da = 50$ (Fig 5c), curves corresponding to different values of \tilde{x} do not coincide, except
 470 at the final point “F”, where $\tilde{N} = 0$ and $\tilde{q} = 0$. This means that horizontal gradients exist during
 471 the entire mixing process and mixing is inhomogeneous till the final equilibrium state is
 472 reached. Droplets penetrating into the initially droplet-free volume begin evaporating, so only a
 473 small fraction of droplets reaches the centre of the droplet-free volume, as seen in Fig. 5c,
 474 $\tilde{x} = 3/4$ (black curve). Accordingly, at $\tilde{x} = 3/4$ the droplet concentrations and \tilde{q} reach their
 475 maxima (of 0.1 and 0.05, respectively) and then decrease to zero. At $Da = 500$ (Fig 5d), all
 476 the droplets evaporate before reaching the centre of the dry volume, indicating an extremely
 477 high spatial inhomogeneity of droplet evaporation. Hence, only two curves for $\tilde{x} = 1/4$ and
 478 $\tilde{x} = 1/2$ are seen in Fig.5d.

479 Fig. 5 also shows that the slopes of the curves describing the $\tilde{N} - \tilde{q}$ relationships are
 480 different at different values of \tilde{x} and change over time. At large Da , the slopes of the curves
 481 describing the dependencies $\tilde{N} - \tilde{q}$ in the initially cloud volume are close to linear. However,
 482 the slope at a high value of \tilde{q} is still flatter than that at a low value of \tilde{q} . This can be attributed
 483 to the fact that when \tilde{q} is large, it decreases faster than the concentration \tilde{N} because some

484 fraction of droplets evaporate only partially. At the end of the mixing when \tilde{q} is small, \tilde{N}
 485 decreases faster than \tilde{q} , because the droplet concentration is determined by the smallest
 486 droplets, while \tilde{q} is determined by larger droplets.

487 As was discussed in Pt. 1, according to the classical concept of extremely inhomogeneous
 488 mixing, the ratio q/N remains constant. For dimensionless \tilde{N} and \tilde{q} , the scattering points
 489 should be aligned along the 1:1 line. Therefore, the closeness of particular cases to the classical
 490 extremely inhomogeneous mixing can be evaluated by the deviation of the $\tilde{N}-\tilde{q}$ curve from
 491 the 1:1 line. One can see that at $Da = 500$ the $\tilde{N}-\tilde{q}$ relationship is closer to linear.

492 Despite the fact that at $R < -1$ all the droplets within the mixing volume evaporate, it is
 493 interesting to follow the DSD evolution during this process. **Figure 6** shows the time evolution
 494 of a normalized DSD at $Da = 1$ and $Da = 50$. One can see a substantial difference in the DSD
 495 evolutions at different Da . At $Da = 1$, different DSDs are formed very rapidly at different
 496 values of \tilde{x} (panel a). The widest DSD occurs at $\tilde{x} = 1$, i.e. at the outer boundary of the initially
 497 droplet-free volume. This is natural, because the supersaturation deficit is the highest at $\tilde{x} = 1$.
 498 At $\tilde{t} > T_{mix} \approx 0.4$, DSD become similar at all values of \tilde{x} (Fig.6b). The DSD width continues to
 499 increase due to partial droplet evaporation. This time period corresponds to the horizontal
 500 segment of the $\tilde{N}-\tilde{q}$ relationship in Fig. 5a. Fig. 6c shows the DSD at the stage when a
 501 decrease in LWC is accompanied by a decrease in number droplet concentration. The
 502 corresponding point in the $\tilde{N}-\tilde{q}$ diagram at this time instance is quite close to the point “F” at
 503 which $\tilde{N} = 0$ and $\tilde{q} = 0$.

504 At $Da = 50$, DSD are different at different \tilde{x} during the entire period of mixing. While
 505 DSD at $\tilde{x} > 0.5$ are wide and droplet evaporation is accompanied by a shift of DSD maximum
 506 to smaller droplet radii (this feature is typically attributed to homogeneous mixing), the DSD
 507 maximum at $\tilde{x} < 0.5$ (the initially cloud volume) shifts toward smaller radii only slightly until
 508 $\tilde{t} = 3.17$ (Fig. 6e). Further droplet evaporation either leads to a complete evaporation (at

509 $\tilde{x} \geq 0.5$) or shifts the DSDs to smaller droplet sizes (panel f). The maximum droplet
 510 concentration takes place at $\tilde{x} = 0$. Fig. 6 shows that DSD shapes evolve substantially over
 511 time, although the final state is characterized by complete droplet evaporation.

512

513 **5.2 Partial evaporation case**

514 **5.2.1 Evolution of the microphysical parameters at different values of Da and R**

515 Here we consider the process of mixing at $R > -1$, i.e. when not all the droplets evaporate
 516 completely. **Figure 7** shows the horizontal profiles of a normalized supersaturation at different
 517 Da and R . One can see that in all cases, the final state occurs when the equilibrium
 518 supersaturation $\tilde{S} = 0$ (RH=100%). However, this final value is reached quite differently
 519 depending on Da . At $Da = 1$, rapid mixing leads to formation of spatially homogeneous
 520 humidity and supersaturation during a time period of a fraction of τ_{pr} . Then, supersaturation
 521 within the mixing volume grows by evaporation of droplets, which are uniformly distributed
 522 over the entire mixing volume. This process of homogeneous mixing was analyzed in detail in
 523 Pt. 2.

524 At $Da = 500$, changes in supersaturation take place largely within the initially droplet-free
 525 volume. RH in the initially cloud volume undergoes only small changes. This process agrees
 526 well with the classical concept of extremely inhomogeneous mixing. However, a strong
 527 gradient of supersaturation remains within the initially drop-free volume for a long time (tens
 528 of τ_{pr}). At $Da = 50$, the situation is intermediate. Mixing is intensive enough to decrease RH
 529 in the initially cloud volume, but spatially uniform RH is established within about $5-10\tau_{pr}$,
 530 increasing with an increase in $|R|$. After this time instance, mixing takes place according to the
 531 homogeneous scenario.

532 **Figure 8** shows the horizontal profiles of normalized LWC at different Da and R . At the
 533 same R , the final equilibrium values of LWC are identical, as follows from Eq. (30); LWC

534 decreases with an increase in $|R|$. At any Da , the decrease in the LWC in the cloud volume is
 535 caused largely by diffusion of droplets from the cloud volume into the initially droplet-free
 536 volume .

537 At $Da = 500$, evaporation in the cloud volume is small because \tilde{S} in these volumes is high
 538 in cloud volumes during mixing (Fig. 7). At $Da = 1$, the process of spatial homogenization
 539 takes place during fractions of τ_{pr} , i.e. $T_{mix} < 1$. Then, during a relatively lengthy period of
 540 $10\tau_{pr}$, evaporation decreases LWC over the entire mixing volume, which is characteristic of
 541 homogeneous mixing. At $Da = 50$, spatial homogenization takes place during about $T_{mix} \approx 15$.
 542 This is a slightly shorter time than it takes to establish the final equilibrium stage T_{tot} . Different
 543 Da 's cases reach equilibrium at different times. The process of reaching a final uniform LWC
 544 lasts for $100\tau_{pr}$ at $Da = 500$ and for about τ_{pr} at $Da = 1$.

545 **Figure 9** shows the profiles of the normalized droplet concentrations at different Da and
 546 R . In contrast to LWC, the final concentration depends both on Da and R . Hence, profiles at
 547 different Da can have different shapes at the same value of R . At $R = -0.1$ (which corresponds
 548 to high RH in the initially dry volume) none of the droplets evaporate, so the final normalized
 549 droplet concentration is equal to $\tilde{N} = 1/2$. This means that all the droplets in the initially cloud
 550 volume are now uniformly distributed between both mixing volumes. At larger $|R|$, i.e., at
 551 lower RH in an initially droplet-free volume, some droplets evaporate completely. The final
 552 concentration decreases with an increase in Da .

553 The physical interpretation of this dependence is clear. At low Da , fast mixing leads to
 554 formation of a uniform RH throughout the entire mixing volume, and this affects all the
 555 droplets. At high Da , RH in the initially droplet-free volume remains low for a long time,
 556 and droplets that penetrate can evaporate. Therefore, the fraction of completely evaporated
 557 droplets increases with Da : at $R = -0.1$ there are no completely evaporated droplets at any Da .

558 At $R = -0.3$ a decrease in the droplet concentration takes place only at $Da = 500$, and at
 559 $R = -0.5$ the droplet concentration decreases already at $Da \geq 50$.

560 The comparative contributions of different factors in establishing the final states of mixing
 561 are well seen in **Figure 10** presenting the relationships between normalized concentration and
 562 normalized LWC at three values of \tilde{x} : $1/4$ (centre of the cloudy volume), $1/2$ and $3/4$ (centre of
 563 the initially dry volume) at $R = -0.5$ and different values of Da . Fig. 10 is analogous to Fig. 5,
 564 but plotted for $R > -1$.

565 At $Da = 1$ the mixing is very fast, which leads to a rapid decrease in LWC and in the
 566 droplet concentration in the initially cloud volume and to an increase of these quantities in the
 567 initially droplet-free volume. As a result of the rapid mixing and homogenization, all the curves
 568 coincide at point “A” (left panel). After this time instance, spatial homogeneous evaporation
 569 takes place. Since at $Da = 1$ only partial, but not total, droplet evaporation occurs, the droplet
 570 concentration remains unchanged even while LWC decreases. At $Da = 50$ and $Da = 500$, the
 571 three curves coincide at the final point “F” only. At $Da = 500$, the relationship between the
 572 droplet concentration and the mass becomes more linear (blue curve). The linear dependence is
 573 consistent with the concept of extremely inhomogeneous mixing (see Pt1). Considerations
 574 regarding the closeness of the $\tilde{N} - \tilde{q}$ relationship to the line 1:1 as a measure of
 575 inhomogeneity of mixing made at $R < -1$ are also valid for $R > -1$.

576

577 **5.2.2 Evolution of DSDs and the DSD parameters**

578 **Figure 11** presents examples of the DSD evolution at the center of the initially cloud
 579 volume ($\tilde{x} = 1/4$) (upper row) and of the initially droplet-free volume ($\tilde{x} = 3/4$) at $R = -0.5$
 580 and different values of Da . Several specific features of the DSD are notable. As a result of the
 581 rapid mixing at $Da = 1$ (left column), DSD become similar in both volumes already at
 582 $t = 0.317\tau_{pr}$ (black lines). Further evolution is similar in both volumes and is characterized by
 583 broadening of the DSD and its shifting and of the DSD toward smaller droplet sizes. This shift

584 means a decrease in the mass at constant droplet concentration, which is typical of
 585 homogeneous mixing.

586 The initially monodisperse DSDs become polydisperse. The mechanism of the DSD
 587 broadening at $Da=1$ is illustrated in **Figure 12**, showing the DSD at the earlier,
 588 inhomogeneous stage at different \tilde{x} . One can see that within very short periods when the
 589 spatial gradient of saturation deficit exists, droplets entering the initially droplet-free volume
 590 partially evaporate, reaching their minimal size at $\tilde{x}=1$. In this way, a polydisperse DSD
 591 forms. As the mixing proceeds, DSD become spatially homogenized, as seen in the right panel
 592 of Fig. 12.

593 At $Da=50$ and $Da=500$, the DSD shapes substantially differ from those at $Da=1$.
 594 There are two main differences: the peak of the distribution shifts only slightly (at $Da=50$) or
 595 does not shift at all (at $Da=500$). At the same time, the DSD develops a long tail of small
 596 droplets. Since the mixing rate at these values of Da is slow, droplets penetrating deeper into
 597 the initially dry volume remain there for long time and get smaller. As a result, at moderate
 598 and large Da , a polydisperse DSDs form with droplet sizes ranging from zero to 1. Formation
 599 of a long tail of small droplets in case of inhomogeneous mixing was simulated in direct
 600 numerical simulation (DNS) by Kumar et al. (2012), as well as by means of “the explicit-
 601 mixing parcel model” (EMPM) (Krueger et al., 1997; Su et al., 1998; Schlüter, 2006).

602 **Figure 13** shows the spatial dependencies of the DSD dispersion (ratio of DSD r.m.s. width
 603 and the mean radius) at different time instances and different values of Da and R . One can see
 604 that the dispersion increases with an increase in Da and in $|R|$. This behavior can be accounted
 605 for by the fact that the DSD broadening toward smallest droplet size increases with the increase
 606 in Da and in $|R|$. The DSD dispersion increases with time and with an increase in \tilde{x} , i.e.
 607 further into the initially droplet free volume. At the same time, spatial homogenization takes
 608 place, so at the final state at $R=-0.5$ the DSD dispersion reaches 0.11 at $Da=1$ and about 0.2
 609 at $Da=50$ and $Da=500$.

610 Observed DSD dispersion in different clouds typically ranges from 0.1 to 0.4 (Khain et al.,
 611 2000; Martin et al., 2004; Prabha et al., 2012) and can be caused the following factors: in-
 612 cloud nucleation (e.g. Khain et al., 2000; Pinsky and Khain, 2002), spatial averaging along
 613 aircraft traverses (Korolev, 1995) and non-symmetry in droplet nucleation/denucleation
 614 (Korolev, 1995). As seen in Fig. 13, this dispersion may be also caused by mixing at cloud
 615 edges at moderate and large Da . Hence, inhomogeneous mixing leads to DSD broadening.

616 The effective radius, r_{eff} , is an important DSD characteristic. According to the classical
 617 concept, r_{eff} remains unchanged during extremely inhomogeneous mixing, whereas decreases
 618 during homogeneous mixing. **Figure 14** shows spatial dependencies of r_{eff} at different time
 619 instances and different values of Da and R . At $R = -0.1$ (high RH in the surrounding volume)
 620 r_{eff} is similar for all values of Da . So, at high R (i.e., close to zero), the behaviour of r_{eff} does
 621 not allow to distinguish between mixing types.

622 At a given R , the final r_{eff} increases with increasing Da . For instance, at $R = -0.5$, r_{eff} at
 623 the final state differs from the initial r_{eff} value by less than 6% at $Da = 500$, while at $Da = 1$
 624 r_{eff} decreases by 20%. At moderate and high Da , large gradients of r_{eff} exist during the
 625 mixing process. However, the gradient is high only in the initially droplet-free volume where
 626 r_{eff} decreases significantly due to the intense evaporation of droplets. Besides, r_{eff} growth very
 627 rapidly in the initially droplet free volume, so at high Da during most of the mixing time r_{eff}
 628 within the mixing volume becomes close to the initial r_{eff} value in the cloudy volume.

629

630 **5.3 Delimitation between mixing types**

631 Typically, the Da value is used as a criterion for delimitation between mixing types.
 632 $Da = 1$ is usually used as a boundary value separating homogeneous and inhomogeneous
 633 mixing. As shown in Section 4, mixing always starts as inhomogeneous. In the course of

634 mixing, the initial spatial gradients decrease and the air volumes either become identical or
 635 remain different. In the former case, the second mixing stage is homogeneous. If
 636 inhomogeneity persists until the equilibrium state is established, mixing remains
 637 inhomogeneous during the entire period. Both mixing stages can be characterized by duration,
 638 change in the droplet concentrations or LWCs, and other quantitative characteristics. These
 639 characteristics are functions of two non-dimensional parameters R and Da , which can be
 640 calculated and used for delimitation between mixing types. Since mixing between volumes
 641 may turn from inhomogeneous into homogeneous before reaching the equilibrium state, it is
 642 necessary to use some quantitative criteria to delimit mixing types. Below, delimitation is
 643 performed for $R > -1$ which corresponds to partial evaporation of droplets by the end of
 644 mixing.

645

646 **5.3.1. Characteristic time periods of mixing**

647 Three characteristic time periods of mixing are distinguished: a) mixing period T_{mix} , during
 648 which spatial gradients are smoothening (may be also called the homogenization period); b)
 649 period T_{ev} during which $S < 0$ and droplets evaporate until saturation is reached and c) the total
 650 mixing period T_{tot} that lasts until the final equilibrium stage is reached. In our analysis, all the
 651 three periods are assumed dimensionless quantities.

652 We use solution (28) for conservative function $\tilde{\Gamma}(\tilde{x}, \tilde{t})$ to define quantitatively time period
 653 T_{mix} . The deviation of the solution from its final value $\Delta\tilde{\Gamma} = \tilde{\Gamma}(\tilde{x}, \tilde{t}) - \tilde{\Gamma}(\tilde{x}, \infty)$ at $\tilde{t} \rightarrow \infty$ can be
 654 approximately estimated using the first term of the series expansion as

$$\begin{aligned}
 655 \quad |\Delta\tilde{\Gamma}|_{\max} &\approx \left| (1-R) \frac{\sin(\pi/2)}{\pi/2} \exp\left(-\frac{\pi^2 \tilde{t}}{Da}\right) \cos(\pi \tilde{x}) \right|_{\max} = \\
 &(1-R) \frac{2}{\pi} \exp\left(-\frac{\pi^2 \tilde{t}}{Da}\right)
 \end{aligned} \tag{37}$$

656 From Eq. (37) the estimation of T_{mix} can be written as

657

$$658 \quad T_{mix} = -\frac{Da}{\pi^2} \ln \left[\frac{\pi}{2(1-R)} \left| \Delta \tilde{\Gamma} \right|_{\max} \right] \quad (38a)$$

659 Suppose the value of the maximum deviation is $\left| \Delta \tilde{\Gamma} \right|_{\max} = 0.02$. This is a small value
 660 compared to the initial leap of function $\tilde{\Gamma}$, which is equal to $1-R$. At $\left| \Delta \tilde{\Gamma} \right|_{\max} = 0.02$ the
 661 duration of the non-homogeneous stage is evaluated as

$$662 \quad T_{mix} = -\frac{Da}{\pi^2} \ln \left[\frac{0.01\pi}{1-R} \right] \quad (38b)$$

663

664 Several studies evaluate the evaporation time for droplets of a particular size using the
 665 equation for diffusion growth (e.g. Lehmann et al., 2009). In our study, the evaporation time
 666 duration T_{ev} is defined as the period during which the maximum deviation of supersaturation
 667 from zero exceeds the small value chosen as $\left| \Delta \tilde{S} \right|_{\max} = 0.02$:

$$668 \quad \left| \tilde{S}(\tilde{x}, T_{ev}) \right| \leq \left| \Delta \tilde{S} \right|_{\max} = 0.02 \quad (39)$$

669

670 Although criterion (39) is rather subjective, it has an advantage over the criterion used by
 671 Lehmann et al. (2009), as Eq. (32) characterizes evaporation of the droplet population taking
 672 into account the simultaneous increase in supersaturation, but not of individual droplets of
 673 particular size at constant S as in Lehmann et al. (2009).

674 At the end of the mixing, both the thermodynamic equilibrium and the diffusion
 675 equilibrium are reached. Accordingly, the total time of mixing T_{tot} is evaluated as the
 676 maximum of the two time periods needed to achieve equilibrium $T_{tot} = \max \{ T_{mix}, T_{ev} \}$. All the
 677 three characteristic time periods are normalized on the phase relaxation time, and, therefore,
 678 depend on the two non-dimensional parameters R and Da . The contours of the characteristic
 679 time durations T_{mix} , T_{ev} and T_{tot} in the $Da-R$ diagrams are shown in **Figure 15**.

680 As follows from Eq. (38b), T_{mix} is proportional to Da . The dependence of T_{mix} on R is not
 681 very strong, so T_{mix} slightly decreases with increasing R . This can be attributed to the fact that
 682 the lower R , the smaller the initial inhomogeneity of function $\tilde{\Gamma}$ and the shorter the time to
 683 align this inhomogeneity is. At small Da (high rate of homogenization of the volume), T_{ev}
 684 depends largely on R . At large Da , T_{ev} depends substantially on Da , since the evaporation
 685 rate depends on the number of droplets that diffuse to drier parts of the mixing volume. A
 686 comparison of Fig. 15c with Figs. 15a and 15b shows that at small Da , time T_{tot} is determined
 687 by T_{ev} , while at large Da , T_{tot} is determined by T_{mix} .

688

689 5.3.2. Determination of boundaries between the mixing types on the $R-Da$ plane

690 Several criteria can be proposed for delimitation between mixing types. We consider these
 691 criteria for $R > -1$. As discussed above, mixing always starts as inhomogeneous and late either
 692 become homogeneous or remains inhomogeneous till the final equilibrium state is established.
 693 At small Da , the homogenization takes place during $T_{mix} < T_{tot}$. The value of time fraction λ_1 of
 694 the inhomogeneous stage can serve as a criterion for definition of homogeneous mixing. This
 695 formula for the fraction can be written as

$$696 \lambda_1 = \frac{T_{mix}}{T_{tot}} \quad (40)$$

697 The case $\lambda_1 \leq 0.5$, most time the mixing takes place according the homogeneous scenario and
 698 such regime is reasonable to regard as homogeneous mixing. If $\lambda_1(R, Da)$ changes within the
 699 range of $0.5 < \lambda_1 \leq 1$, mixing appears to be intermediate. The criterion (40) depends on the non-
 700 dimensional parameters R and Da . **Figure 16a** shows the boundaries separating mixing types
 701 on the $Da-R$ plane. These boundaries separate all plane into several zones. At very small R ,
 702 the duration of the phase transition is negligibly small. According to criterion (40), in this case
 703 mixing should be considered inhomogeneous, irrespective of the Da value.

704 Another criterion of delimitation between mixing types can be determined from a
 705 comparison of LWC variation rates due to different mechanisms. The mean normalized LWC
 706 (which is equal to the mean normalized liquid water mixing ratio) can be written as integral

707 $\langle \tilde{q}(\tilde{t}) \rangle = \int_0^1 \tilde{q}(\tilde{x}, \tilde{t}) d\tilde{x}$. The initial mean LWC is equal to $\langle \tilde{q}(t=0) \rangle = \frac{1}{2}$. The final equilibrium

708 LWC is equal to $\langle \tilde{q}(t=\infty) \rangle = \frac{1}{2}(1+R)$ (Eq. (30)). The total amount of liquid water that

709 evaporates in the course of mixing can be quantified by the difference between these two

710 values $\langle \tilde{q}(t=0) \rangle - \langle \tilde{q}(t=\infty) \rangle = -\frac{1}{2}R$. The amount of liquid water evaporated in the course of the

711 first inhomogeneous mixing stage is calculated by the equation

712 $\langle \tilde{q}(t=0) \rangle - \langle \tilde{q}(T_{mix}) \rangle = \frac{1}{2} - \langle \tilde{q}(T_{mix}) \rangle$. Hence, parameter λ_2 which is a ratio of

713

$$714 \lambda_2 = \frac{\langle \tilde{q}(t=0) \rangle - \langle \tilde{q}(T_{mix}) \rangle}{\langle \tilde{q}(t=0) \rangle - \langle \tilde{q}(t=\infty) \rangle} = \frac{2\langle \tilde{q}(T_{mix}) \rangle - 1}{R} \quad (41)$$

715 can serve as another possible criterion for delimitation between mixing types. This ratio

716 characterizes the fraction of liquid water that evaporates at the initial inhomogeneous stage.

717 Condition $\lambda_2 < 0.5$ in this case corresponds to homogeneous mixing, while condition

718 $0.5 \leq \lambda_2 < 1$ corresponds to intermediate mixing. We regard the case $\lambda_2 = 1$ as inhomogeneous

719 mixing. Certainly, criterion λ_2 depends on the non-dimensional parameters R and Da . Fig.

720 16b illustrates delimitation between mixing types on the $Da-R$ plane according to criterion

721 λ_2 .

722 Comparison of Figs. 16a and 16b shows that both criteria lead to nearly similar separation

723 of the $Da-R$ plane into three zones corresponding to homogeneous, intermediate and

724 inhomogeneous mixing. At the same time, the boundaries separating these zones are different

725 depending on the delimitation criterion used. Nevertheless, it can be concluded that mixing can

726 be considered homogeneous at Da below 4-10 and $R < -0.1$ and inhomogeneous at Da
 727 exceeding several tens.

728 Terms "inhomogeneous mixing" (Burner and Brenguier, 2007) and "extremely
 729 inhomogeneous mixing" (Lehmann et al., 2009; Gerber et al., 2008; Pt1) are used to denote the
 730 mixing regime when the relationship between the normalized values \tilde{N} and \tilde{q} is represented
 731 by a straight 1:1 line, which is equivalent to the constant mean volume radius (in some studies,
 732 the effective radius is used instead of the mean volume radius. According to the definition used
 733 in the present study, extremely inhomogeneous mixing is the limiting case of inhomogeneous
 734 mixing when $Da \rightarrow \infty$. Despite the fact that the extremely inhomogeneous mixing is only an
 735 idealization our approach allows to determine to what extent mixing can be considered to be
 736 close to this limiting case. The measure of inhomogeneity of mixing is the closeness of the
 737 $\tilde{N} - \tilde{q}$ relationship to the 1:1 straight line (see discussion above related to Figs. 5 and 10).

738 **Figure 17a** shows r.m.s. distance between the $\tilde{N} - \tilde{q}$ relationship and the 1:1 straight line,
 739 depending on Da and R . These dependences were calculated using the set of points \tilde{N}_i, \tilde{q}_i
 740 uniformly distributed over spatial interval $[0 \ 1]$ and time interval $[0 \ T_{tot}]$. The equation for

741 estimation is $\delta = \sqrt{\frac{1}{2M} \sum_{i=1}^M (\tilde{N}_i - \tilde{q}_i)^2}$, where M is the total number of points. This distance

742 corresponds to r.m.s. deviation of the normalized mean volume radius from 1. The dependences
 743 of the last deviation on Da and R and estimated as $\delta/3$ are shown in Fig. 17b. This estimation
 744 is based on the fact that the total mass of droplets is proportional to the cube of the mean
 745 volume radius. As expected, the distance decreases with increasing in Da . At large R , all the
 746 curves coincide indicating a degenerative case when type of mixing becomes indistinguishable.

747 We choose the value $\delta/3$ equal to 0.02 to determine the boundary of the extremely
 748 inhomogeneous mixing zone. The value of 0.02 corresponds to droplet radii deviation of a few
 749 tenths of a micron, which is so low that in in-situ measurements this case would always be
 750 attributed to extremely inhomogeneous mixing. In Fig.16 this boundary is marked by broken

751 line. The boundary shows that the mixing at Da exceeding several hundred can be attributed
 752 to the extremely inhomogeneous. Between the boundary separating inhomogeneous mixing
 753 from the intermediate one and the boundary separated inhomogeneous mixing from extremely
 754 inhomogeneous there exists a wide zone of inhomogeneous mixing where the mean volume (or
 755 the effective) radius may drop by 10% and more (Fig. 14), and where the DSD dispersion is
 756 substantial and the tail of small droplets is long enough (Fig. 11). Mixing diagrams currently
 757 used for analysis of observed data ($N - q$ dependences in the final equilibrium state of mixing)
 758 do not contain this zone which, therefore, has remained unrecognized and uninvestigated.

759

760 **6. Summary and conclusions**

761 In this study, inhomogeneous turbulent mixing is investigated using a simple a 1D model of
 762 mixing between a saturated cloud volume and an undersaturated droplet-free volume. The
 763 mixing is simulated by solving a diffusion-evaporation equation written in the non-dimensional
 764 form. For simplicity, the initial volumes of cloudy and droplet-free air were assumed to be
 765 equal, and the initial DSD in the cloudy volume was assumed monodisperse.

766 Analysis of the diffusion-evaporation equation shows that the time-dependent process of
 767 mixing and the final equilibrium state depend on two non-dimensional parameters. The first
 768 parameter R , referred in this paper as potential evaporation parameter (PEP) is proportional to
 769 the ratio between the saturation deficit in the initially droplet-free volume and the initial liquid
 770 water content in the cloudy volume. At $R < -1$, the final state is characterized by complete
 771 droplet evaporation and a spatially homogeneous saturation deficit, which indicates dissipation
 772 of the cloudy volume. At $R > -1$, the final state is characterized by existence of droplets and
 773 zero saturation deficit (RH=100%). In this case, the cloud volume expands after mixing with the
 774 entrained air. At small values of $|R|$ (e.g., when RH in the entrained volume is close to 100%),
 775 the effect of droplet evaporation on microphysics is small, and, formally, this kind of mixing
 776 should be regarded as extremely inhomogeneous. Strictly speaking, this is a degenerate case,

777 when homogeneous and inhomogeneous mixing cannot be distinguished (see also Pt. 1). At
 778 $R = 0$, the droplet population turns into a passive admixture and its turbulent diffusion will be
 779 the same as different thermodynamic parameters.

780 The second parameter is the *Damköhler* number (Da) which is the ratio between the
 781 characteristic mixing time and the phase relaxation time. This parameter compares the rates of
 782 spatial diffusion and evaporation. Parameter Da (Eq. (23)) is logically appears in the non-
 783 dimensional form of the diffusion-evaporation equation showing that Da is the ratio of the
 784 mixing time defined as $\tau_{mix} = \frac{L^2}{K}$, to the initial drop relaxation time. The expression for this non-
 785 dimensional parameter clearly shows that since we consider an ensemble of evaporating droplets,
 786 the drop relaxation time evaluated just before the mixing is the characteristic time scale of
 787 inhomogeneous mixing process. In several studies (e.g., Baker and Latham, 1979; Burnet and
 788 Brenguier, 2007; Andejchuk et al., 2009) a question was raised as to which time scale should be
 789 used in formulation of the *Damköhler* number: the time of an individual droplet evaporation at
 790 constant saturation deficit, or the phase relaxation time. This study, as well Pt. 2 show that the
 791 phase relaxation time is the answer. The mixing time is introduced via the turbulent diffusion
 792 coefficient which is a natural measure characterizing the diffusion rate and, in particular,
 793 determines the propagation rate of the fronts in the fields of droplet concentration and other
 794 microphysical parameters. The turbulent diffusion coefficient is widely used to describe mixing
 795 in cloud models at resolved scales.

796 The analysis was performed within a wide range of Da (from 1 to 500) and of R (from -
 797 1.5 to -0.1). The final LWC and the humidity in the mixing volume are determined by the mass
 798 conservation and do not depend on Da (see also Pt. 1 and Pt. 2). At the same time, the droplet
 799 concentration, as well as the shape of DSD and their parameters strongly depend on Da .

800 It is shown that the mixing of air volumes with initially different thermodynamical and
 801 microphysical parameters consists of two stages characterized by two time periods: the time

802 during which microphysical characteristics become uniform over the total mixing volume T_{mix} ,
 803 and the time during which zero saturation deficit is reached (at $R > -1$), T_{ev} . At $\tilde{t} < T_{mix}$, the
 804 spatial gradients of the microphysical values remain and the mixing regime can be regarded as
 805 inhomogeneous. At $\tilde{t} > T_{mix}$, droplet evaporation, if it occurs at all, takes place within a
 806 spatially homogeneous medium, so all the droplets in the mixing volume experience equal
 807 saturation deficit. This regime can be regarded as homogeneous. It is shown, therefore, that at
 808 small Da mixing between two volumes that starts as inhomogeneous can become
 809 homogeneous towards the end of mixing.

810 This finding allows to delimit between mixing types. We presented two quantitative criteria
 811 on the $Da-R$ plane that allow to delimit three mixing regimes: homogeneous, intermediate
 812 and inhomogeneous. These criteria are based on comparison of the characteristic duration
 813 mixing and the evaporation rates. According to the criteria, at Da below about 5, mixing can
 814 be regarded as homogeneous, i.e. the main microphysical changes take place during the
 815 homogeneous stage. At $5 < Da < 50$, the changes in the microphysical parameters are more
 816 significant at the inhomogeneous stage than at the homogeneous stage. In this case, the mixing
 817 can be regarded as intermediate. Finally, at Da exceeding several tens, the spatial
 818 microphysical gradients remain until the final equilibrium stage is reached. In this case, the
 819 mixing can be regarded as inhomogeneous. At Da exceeding a few hundred the deviations
 820 from predictions based of the classical concept of extremely inhomogeneous become relatively
 821 small, which justifies attribute regarding this mixing as extremely inhomogeneous.

822 On the whole, the results of the present study are in line with the classic concepts defining
 823 homogeneous and inhomogeneous mixing types. However, several important points emerge
 824 from our work show serious limitations of classical concepts. A comparison of the classical
 825 concepts and the present study is presented in **Table 2**. Analysis of Tab. 2 shows the following.

826 a) In contrast to many studies that analyze only the hypothetical final (equilibrium) state of
 827 mixing (Barnet and Brenguier, 2007; Gerber et al., 2008; Morrison and Grabowski, 2008; Hill

828 et al., 2009), we consider the entire time-dependent processes of mixing and evaporation. At
 829 moderate and high Da , the mixing can last several minutes. In in-situ observations, we see
 830 mostly non-equilibrium stages which may account for a rather wide scattering of mixing
 831 diagrams even at the same values of Da (e.g., Lehmann et al., 2009).

832 Note that time dependent mixing was also considered in several studies (e.g. Baker et al.,
 833 1980; Baker and Latham, 1982; Jeffery and Reisner, 2006; Krueger et al., 1997; Kumar et al.,
 834 2012) using different approaches and numerical models. These studies, however, do not contain
 835 analysis on non-dimensional diffusion-evaporation equation.

836 b) It is also shown in the study that the slopes of the $\tilde{N} - \tilde{q}$ relationship (between the
 837 normalized droplet concentration and LWC) tends to the 1:1 line with increasing Da . The
 838 closeness can be considered as a measure of extremely inhomogeneous mixing in terms of the
 839 classical concept (see Pt. 1). It has been found that the slope of the $\tilde{N} - \tilde{q}$ relationship depends
 840 on the LWC and, accordingly, on time. At large LWC, \tilde{q} changes with time faster than \tilde{N} ,
 841 while at low LWC the concentration changes faster. Although mixing types are usually
 842 separated into homogeneous and extremely inhomogeneous, we have shown that there are wide
 843 ranges of Da and R at which mixing should be considered intermediate or inhomogeneous,
 844 but not extremely inhomogeneous. Within these ranges the effective radius can change by more
 845 than 10-15%. Standard mixing diagrams do not include this range that, to our knowledge, has
 846 never been investigated despite the fact that multiple in-situ measurements indicate its
 847 existence (e.g. Lu et al. 2014)

848 c) Many studies assume the existence of pure homogeneous mixing during which the
 849 initially monodisperse DSD remains monodisperse. Our study shows that at the very beginning,
 850 mixing is always inhomogeneous. This inhomogeneous stage leads to formation of a
 851 polydisperse DSD that broadens in the course of droplet evaporation. Hence, even at $Da = 1$
 852 the initially monodisperse spectrum becomes polydisperse.

853 d) It is shown that at small Da , mixing includes both inhomogeneous and homogeneous
854 stages, which means that type of mixing can change during the mixing process.

855 e) The classical concept assumes that the effective radius always decreases during
856 homogeneous mixing. Assuming an initially monodisperse DSD, we have found this
857 conclusion largely valid, with the exception small R . At the same time, it was shown in Pt. 2
858 that during homogeneous mixing, the effective radius can decrease, remain constant or increase
859 depending of the initial DSD shape. Thus, a decrease in the effective radius during mixing
860 cannot always be considered an indication of homogeneous mixing. Similarly, the invariability
861 the effective radius during mixing in the process cannot always be considered an indication of
862 extremely inhomogeneous mixing.

863 f) It is generally assumed that during homogeneous mixing droplet concentration remains
864 unchanged. In the present study, as well as in Pt. 2, it is shown that since mixing leads to a
865 polydisperse DSD, the smallest droplets may completely evaporate. At $R < -1$, the DSD
866 becomes very wide and all the droplets, the smallest ones first, evaporate.

867 g) It is generally assumed that inhomogeneous mixing does not alter DSD shape, but only
868 decreases droplet concentration. The present study showed that inhomogeneous mixing
869 significantly changes the DSD shape. DSD were found to be quite different in different regions
870 of mixing volumes. The main feature is the DSD broadening toward small droplet size, so the
871 relative dispersion grows up to 0.2-0.3. These values are quite close to those observed in
872 atmospheric clouds (Khain et al., 2000). Elongated tails of small droplets during mixing were
873 simulated by Schlüter (2006) who described turbulent diffusion following to Kruger et al.,
874 (1997) and Su et al., (1998) as well as by Kumar et al. (2012) using DNS. We see that
875 formation of a polydisperse DSD is a natural result of inhomogeneous mixing and, therefore,
876 inhomogeneous mixing is an important mechanism of DSD broadening. A significant impact
877 of mixing on DSD shape was found identified in multiple studies, beginning with Warner
878 (1973).

879 h) The effective radius has been assumed to remain constant during extremely
880 inhomogeneous mixing. Our results indicate that, indeed, at the final equilibrium stage at
881 comparatively high RH the effective radius is close to that in the initially cloudy volume
882 (especially at high Da). At the same time, we found that the effective radius varies in size and
883 is smaller in the initially droplet-free volumes.

884 The results obtained in parts Pt1 and Pt 2, and especially in the current study (Pt 3)
885 dedicated to analysis of turbulent mixing mechanisms in clouds determine the directions for
886 future work. Since the widely used mixing diagrams show only a hypothetical equilibrium
887 state, but not the instantaneous state of mixing that likely correspond to transition periods, the
888 efficiency of the standard mixing diagrams is questionable. Moreover, the standard diagrams
889 miss a very important mixing regime, namely, inhomogeneous mixing that occurs between two
890 limiting cases of homogeneous and extremely inhomogeneous mixing (Fig. 16).

891 We believe that the results obtained will help to improve understanding and interpretation
892 of mixing process both in in-situ measurements and modeling. The approach allows to
893 investigate the relationship between the main microphysical parameters typical of
894 inhomogeneous mixing, that differ from those in the limiting cases of extremely
895 inhomogeneous mixing. In addition, utilization of polydisperse DSD when solving diffusion-
896 evaporation equation allows to investigate the role of the initial DSD shape in mixing. In-situ
897 measurements (e.g., Burnet and Brenguier, 2007; Gerber et al., 2008; Lehmann et al., 2009)
898 and numerical models (Magaritz-Ronen et al., 2016) show a wide scattering of data on the
899 scattering diagrams. We expect location of various points on the diagrams (e.g. r_v^3 vs. dilution
900 rates) depends on the shape of the initial DSDs and characterizes the stage of mixing. The
901 method applied in the study allows investigation of evolution of DSD moments over space and
902 time .

903 Recently, there has been vigorous discussions concerning the possible existence of high
904 humidity layer near cloud edges that might affect mixing of cloud with its surrounding (Gerber

905 et al., 2008; Lehmann et al., 2009). In our opinion, this layer does exist and forms as a result of
906 turbulent mixing of cloud with surrounding dry air, accompanied by complete droplet
907 evaporation. The approach developed in the present paper allows to analyze formation of such
908 humid layers.

909 We believe that the results obtained in this study will foster the development of physically
910 grounded parameterization of mixing in cloud models.

911

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919 **Appendix. List of symbols**

920

921 **Table A here**

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1031 **Table 1. Main parameters of the problem and their non-dimensional forms***

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Quantity	Symbol	Non-dimensional form	Range of normalized values
Time	t	$\tilde{t} = \frac{t}{\tau_0}$	$[0 \dots \infty]$
Distance	x	$\tilde{x} = \frac{x}{L}$	$[0 \dots 1]$
Square of drop radius	σ	$\tilde{\sigma} = \frac{\sigma}{r_0^2}$	$[0 \dots 1]$
Droplet concentration	N	$\tilde{N} = \frac{N}{N_1}$	$[0 \dots 1]$
Liquid water mixing ratio	q	$\tilde{q} = \frac{q}{q_1}$	$[0 \dots 1]$
Distribution of square of drop radius	$g(\sigma)$	$\tilde{g}(\tilde{\sigma}) = \frac{r_0^2}{N_1} g(\sigma)$	
Conservative function	Γ	$\tilde{\Gamma} = \frac{\Gamma}{A_2 q_1}$	$[-\infty \dots 1]$
Supersaturation	S	$\tilde{S} = \frac{S}{A_2 q_1}$	$[-\infty \dots 0]$
Relaxation time	τ_{pr}	$\tilde{\tau}_{pr} = \frac{\tau_{pr}}{\tau_0}$	$[1 \dots \infty]$
Damköhler number	Da	$Da = \frac{\tau_{mix}}{\tau_0} = \frac{L^2}{K \tau_0}$	$[0 \dots \infty]$
Potential evaporation parameter (PEP)	R	$R = \frac{S_2}{A_2 q_1}$	$[-\infty \dots 0]$

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1034 *All normalized values depend on the initially given values of L , N_1 , r_0 , A_2 , S_2 and K

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1037 **Table 2.** Comparison of analysis based on the classic concepts of mixing and the results of
 1038 the present study
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Classical concept	The present study
Only the final equilibrium state is typically analyzed; results of in-situ observations are interpreted assuming the equilibrium state.	The mixing period can last several minutes and more. The microphysical structure of the mixing volumes during this period can differ substantially from that at the final state
Types of mixing are separated into homogeneous and extremely inhomogeneous.	There are the wide ranges of Da and R values, at which mixing can be regarded as intermediate or inhomogeneous (but not extremely inhomogeneous).
Mixing can start as purely homogeneous	Any mixing starts with the inhomogeneous stage
Homogeneous mixing leads to a DSD shift to small droplet sizes	Homogeneous mixing does not always lead to the DSD shift to small droplet sizes (Pt. 2). The shift depends on the DSD shape.
Mixing can be analyzed within the framework of a monodisperse DSD	Mixing always leads to formation of polydisperse DSD
In the course of homogeneous mixing, droplet concentration remains constant	In the course of homogeneous mixing, droplet concentration does not always remain constant (Pt. 2)
Extremely inhomogeneous mixing does not change the DSD shape	Inhomogeneous mixing (including extremely inhomogeneous) leads to broadening of the DSD towards small sizes
In the course of inhomogeneous mixing, the	The effective radius varies only slightly (5-20

effective radius remains constant	%) in the initially cloud volume. The effective radius rapidly increases in the initially droplet-free volume, approaching the value of effective radius in the cloud volume. With increasing Da , the difference between the values of the effective radius in the initially cloud volume and that at the final state decreases in agreement with the classic concept.
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Tab. A. List of symbols

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("nd" means non-dimensional)

Symbol	Description	Units
A_2	$\frac{1}{q_v} + \frac{L_w^2}{c_p R_v T^2}$, coefficient	nd
a_0, a_n	the Fourier series coefficients	nd
C	the Richardson's law constant	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	$\text{m}^2 \text{s}^{-1}$
Da	the <i>Damköhler</i> number	nd
e	water vapor pressure	N m^{-2}
e_s	saturation vapour pressure above a flat water surface	N m^{-2}
F	$F = \frac{\rho_w L_w^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_s(T) \mathcal{D}}$, coefficient	$\text{m}^{-2} \text{s}$
$f(r)$	droplet size distribution	m^{-4}
$g(\sigma)$	distribution of square radius	m^{-5}
$\tilde{g}(\tilde{\sigma})$	normalized distribution of square radius	nd
k_a	coefficient of air heat conductivity	$\text{J m}^{-1} \text{s}^{-1} \text{K}^{-1}$
K	turbulent diffusion coefficient	$\text{m}^2 \text{s}^{-1}$
L	characteristic spatial scale of mixing	m
L_w	latent heat for liquid water	J kg^{-1}
m_α	moment of DSD of order α	m^{-3}
N	droplet concentration	
\tilde{N}	normalized droplet concentration	nd
N_1	Initial droplet concentration in a cloud volume	m^{-3}
p	pressure of moist air	N m^{-2}
q	liquid water mixing ratio	kg/kg
q_1	Initial liquid water mixing ratio in a cloudy volume	kg/kg
q_v	water vapor mixing ratio	kg/kg
\tilde{q}	normalised liquid water mixing ratio equal to	nd

	normalized LWC	
r	droplet radius	m
r_0	initial droplet radius	m
r_0	mean droplet radius	m
r_v	mean volume radius	m
R	$\frac{S_2}{A_2 q_1}$, potential evaporation parameter (PEP)	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
\tilde{S}	normalized supersaturation	nd
S_2	Initial supersaturation in a dry volume	nd
\tilde{S}_{\max}	maximal normalized supersaturation	nd
T	temperature	K
T_{mix}	normalized duration of inhomogeneous stage	nd
T_{ev}	normalized duration of evaporation	nd
T_{tot}	normalized duration of mixing	nd
t	time	s
\tilde{t}	non-dimensional time	nd
x	distance	m
\tilde{x}	non-dimensional distance	nd
λ_1, λ_2	criteria of delimitation between the types of mixing	nd
ε	turbulent dissipation rate	$\text{m}^2 \text{s}^{-3}$
$\Gamma(x, t)$	conservative function	nd
$\tilde{\Gamma}$	normalized conservative function	nd
ρ_a	air density	kg m^{-3}
ρ_w	density of liquid water	kg m^{-3}
σ	square of droplet radius	m^2
$\tilde{\sigma}$	normalized square of droplet radius	nd
τ_{pr}	phase relaxation time	s

$\tilde{\tau}_{pr}$	normalized phase relaxation time	nd
τ_{mix}	characteristic time of mixing	s
τ_0	Initial time scale	s

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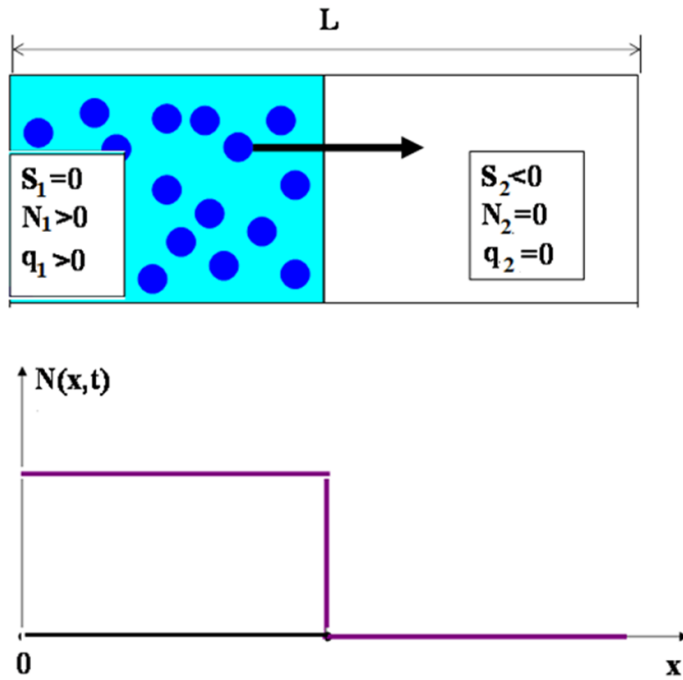
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1083 **Figures**

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1089 **Fig.1.** The schematic illustration of the 1D mixing problem considered in the study. The

1090 initial state at $t = 0$ is illustrated. The left volume of length $L/2$ is a saturated cloudy volume;

1091 the right volume is a non-saturated air volume from the cloud environment.

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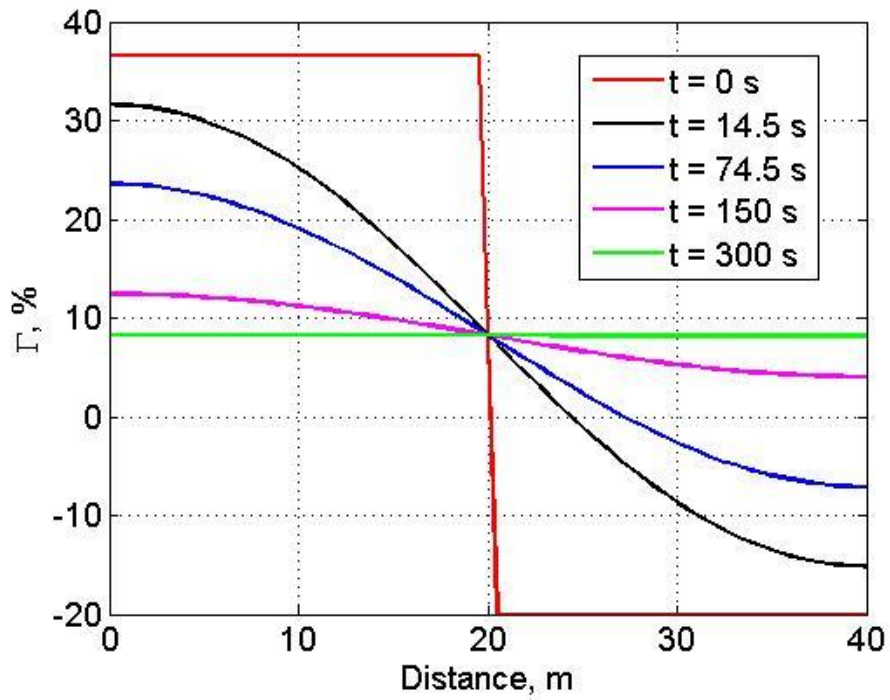


Fig. 2. An example of $\Gamma(x,t)$ evolution during mixing.

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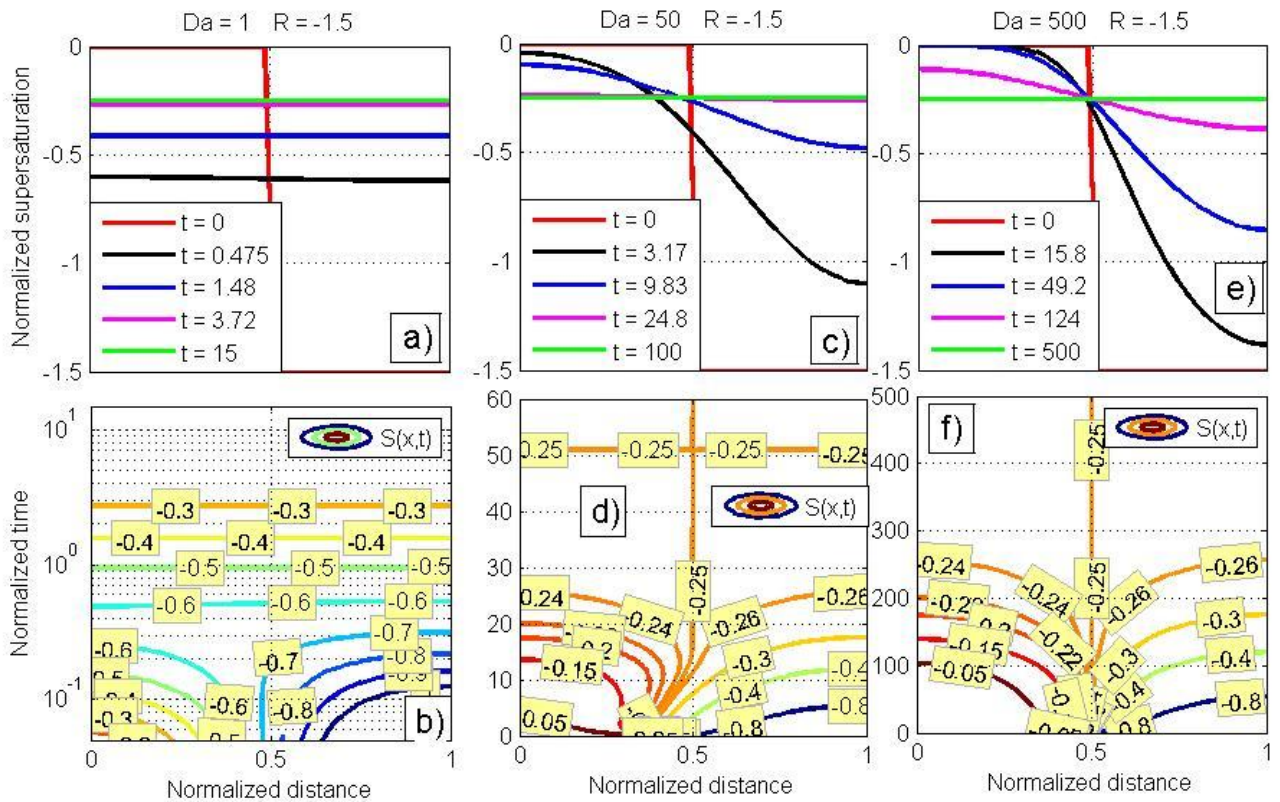


Fig. 3. Horizontal dependencies (upper row) and $\tilde{x}-\tilde{t}$ dependencies (lower row) of normalized supersaturation at $Da=1$, $Da=50$ and $Da=500$ and at $R=-1.5$. Panel b is plotted in semi-log scale.

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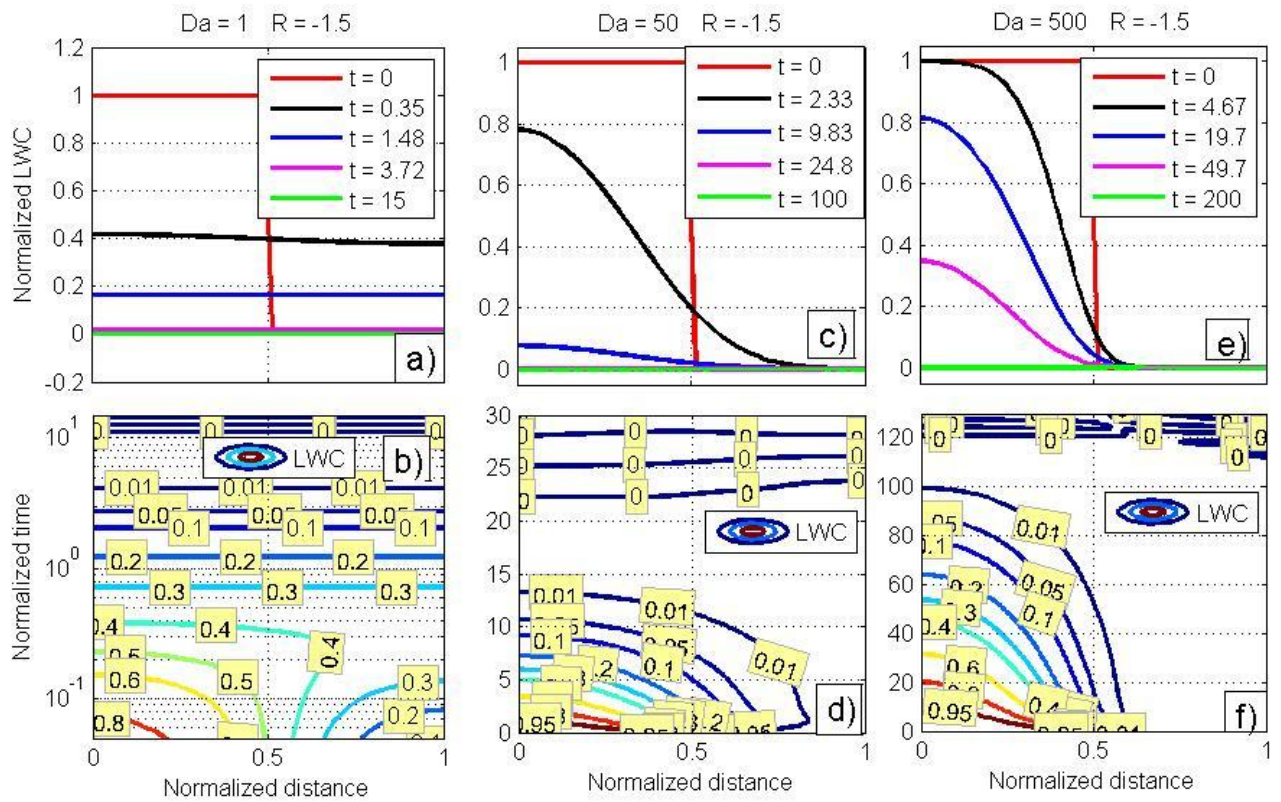
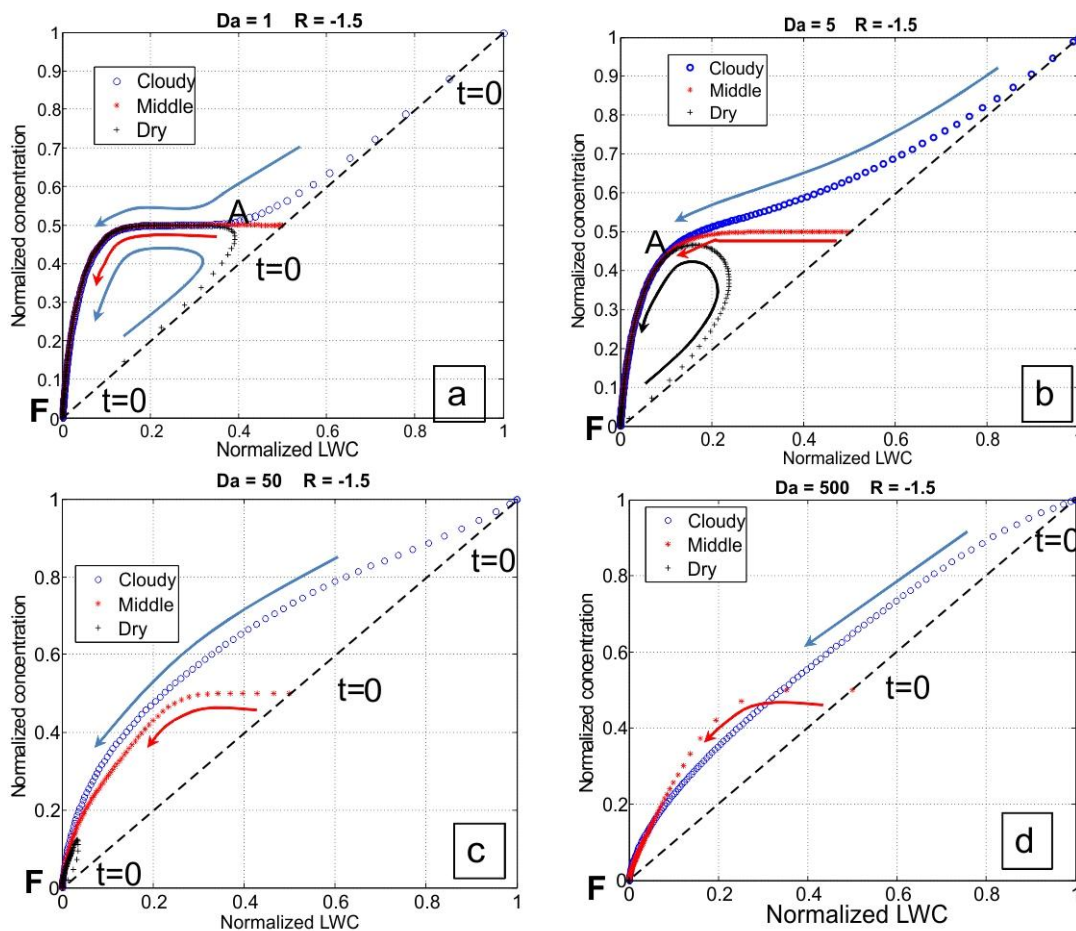


Fig. 4. The same as in Fig. 3, but for normalized LWC. Left bottom panel is plotted in semi-log scale.



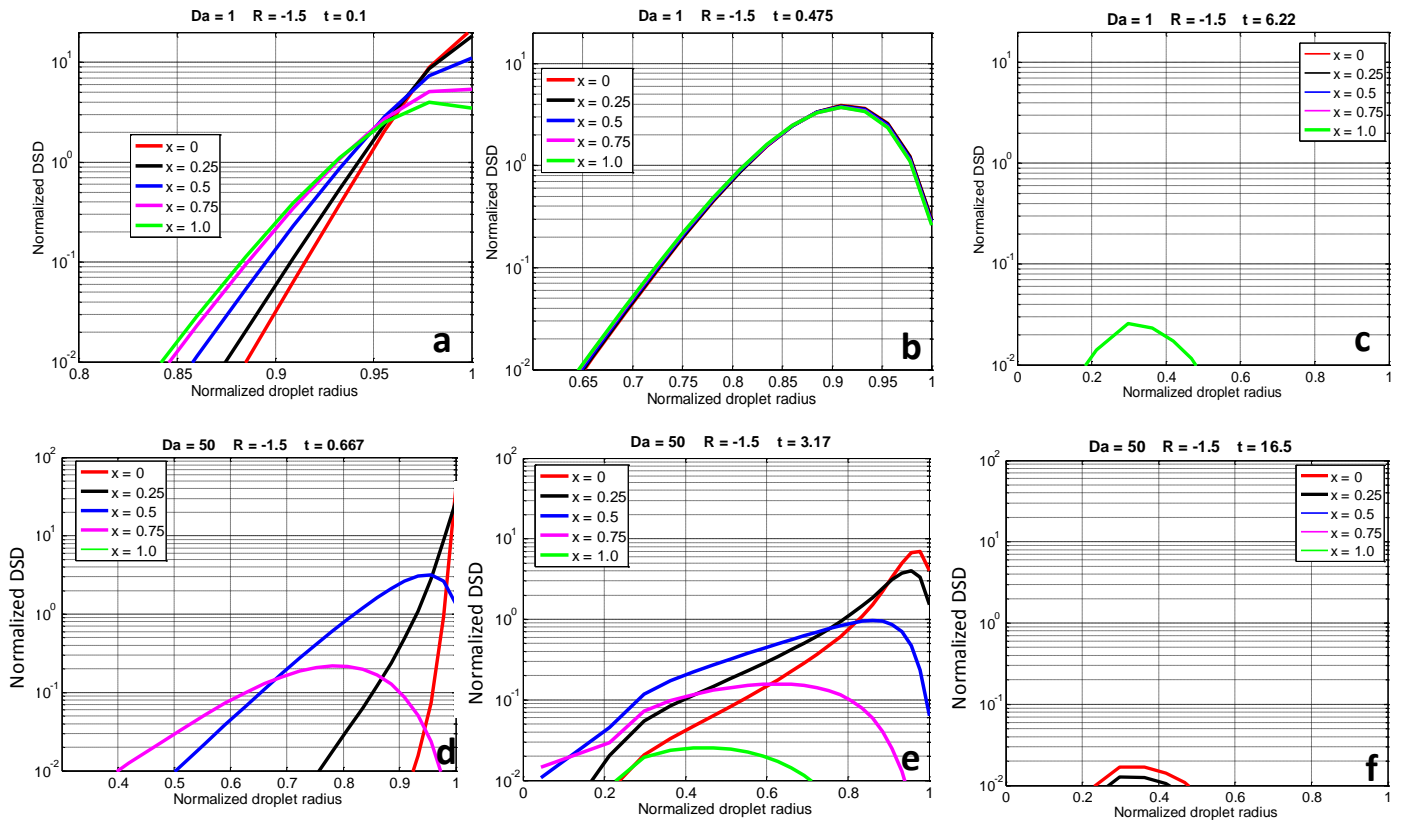
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1165 **Fig. 5.** Dependencies of normalized values of droplet concentration on normalized LWC
 1166 at different Da and $R = -1.5$. Blue symbols mark the centre of the cloudy volume ($\tilde{x} = 1/4$),
 1167 red symbols mark the interface between the cloudy volume and the dry volume ($\tilde{x} = 1/2$), and
 1168 black crosses mark the centre of the initially droplet-free volume ($\tilde{x} = 3/4$). Symbols are
 1169 plotted at different time instances. Symbols at $t=0$ show initial values of droplet concentration
 1170 and LWC at the three values of \tilde{x} . Arrows show the direction of movement of the points at the
 1171 diagram with time. Point “A” marks the beginning of the spatially homogeneous stage, $\tilde{t} = T_{mix}$.
 1172 Point “F” marks the final state. The dashed line indicates the relationship between \tilde{N} and \tilde{q} in
 1173 extremely inhomogeneous mixing (according to the classical concept).

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1179 **Fig. 6** Time evolution of DSD during droplet evaporation at $Da=1$ (upper row) and

1180 $Da = 50$ (bottom row). In each panel, the normalized DSD are shown at different values of

1181 horizontal coordinate \tilde{x} . Different panels show DSD at different time instances.

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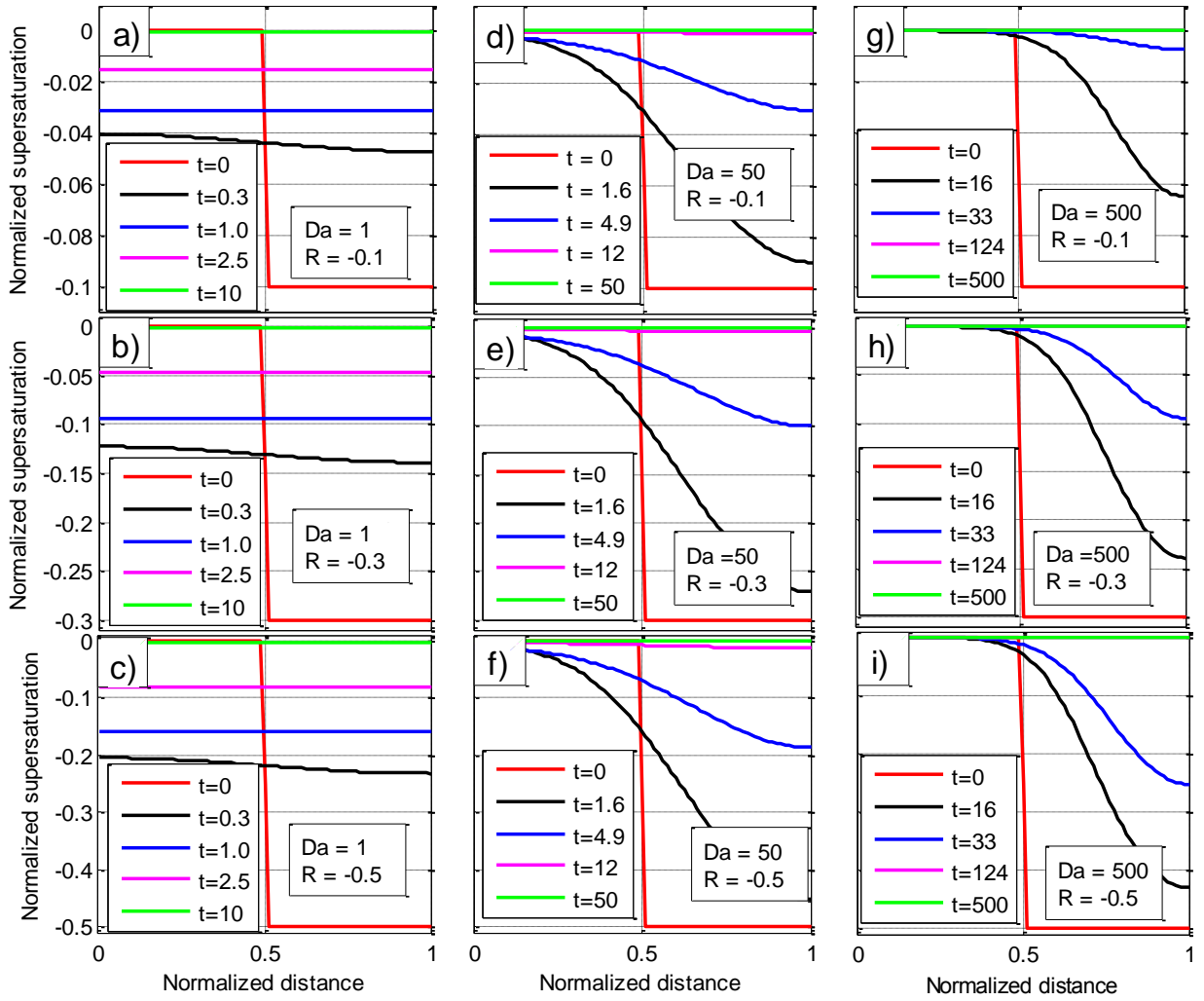


Fig. 7. Profiles of normalized supersaturation at different Da and different $R > -1$.

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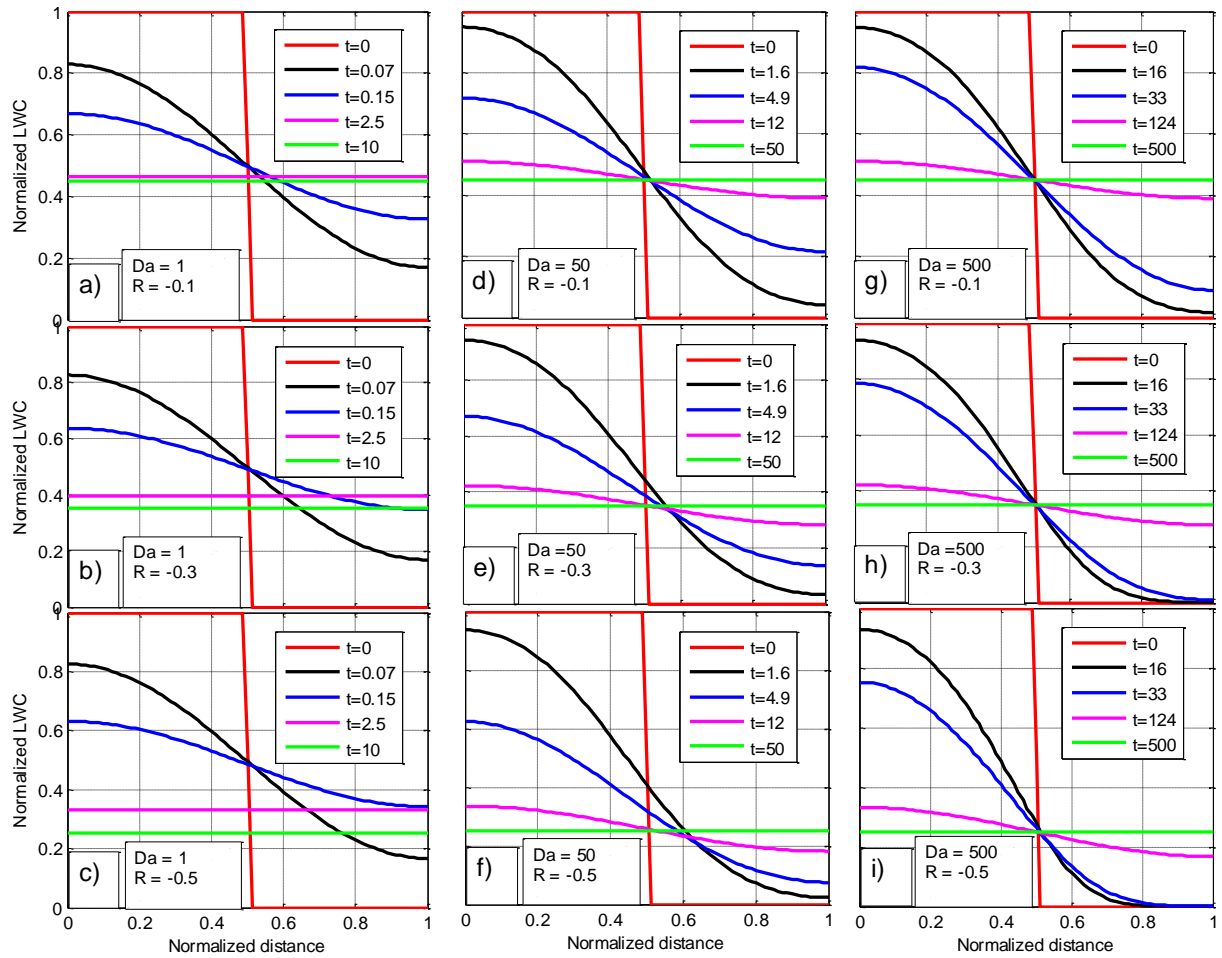


Fig. 8. Profiles of normalized LWC at different Da and at different $R > -1$.

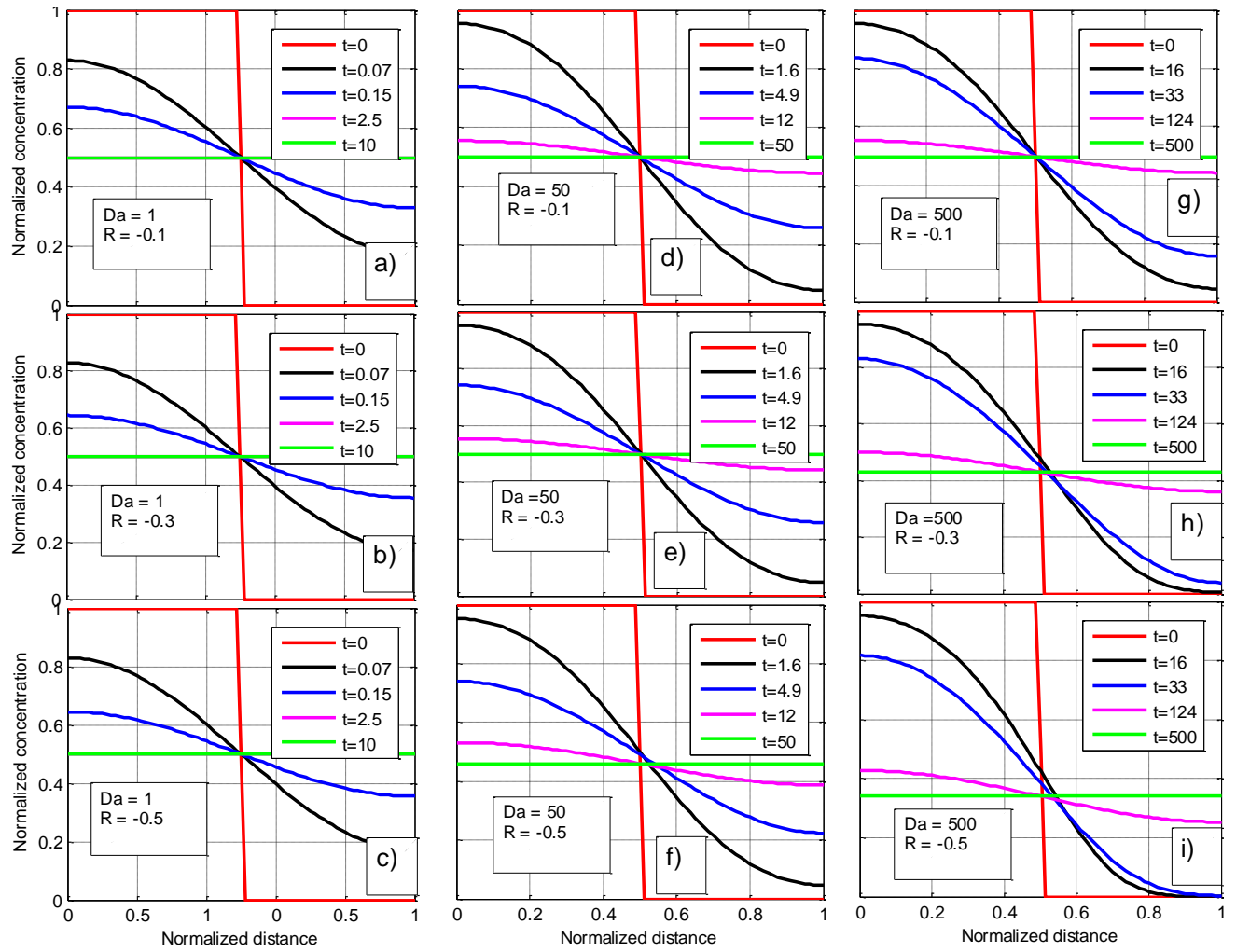
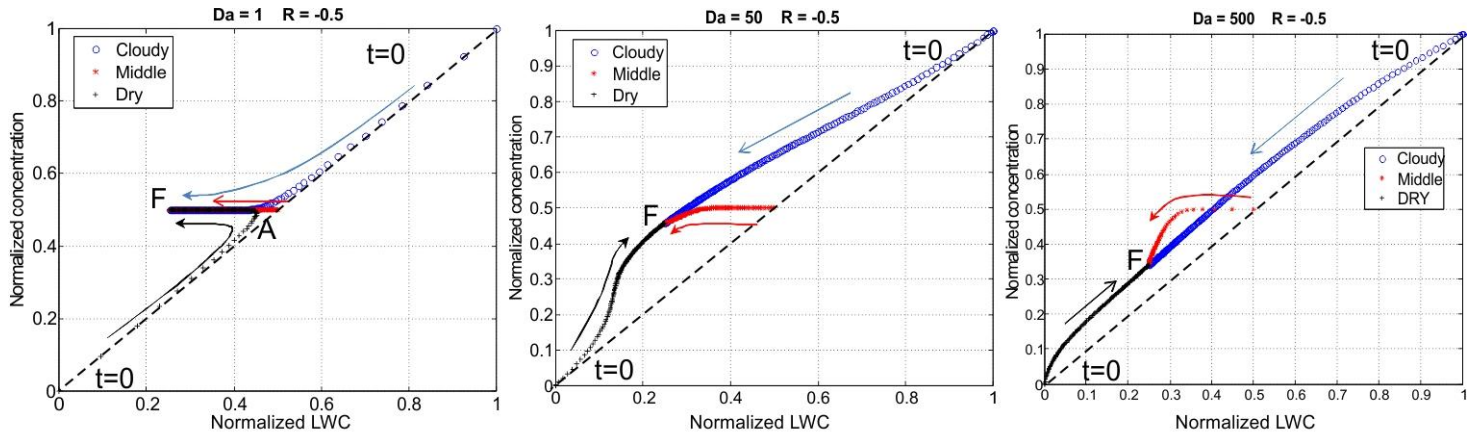


Fig. 9. Profiles of normalized droplet concentration at different Da and at different $R > -1$.

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1259 **Fig. 10.** Dependencies of normalized values of droplet concentration on normalized LWC
 1260 at different Da and at $R = -0.5$. Blue circles mark the centre of the cloudy volume ($\tilde{x} = 1/4$),
 1261 red symbols mark the initial interface ($\tilde{x} = 1/2$) and black crosses mark the centre of the
 1262 initially dry volume ($\tilde{x} = 3/4$). Arrows show the direction of movement of the points with
 1263 time. Point “F” marks the final stationary state of the system. The dashed line indicates the
 1264 relationship between \tilde{N} and \tilde{q} in extremely inhomogeneous mixing (according to the classical
 1265 concept).

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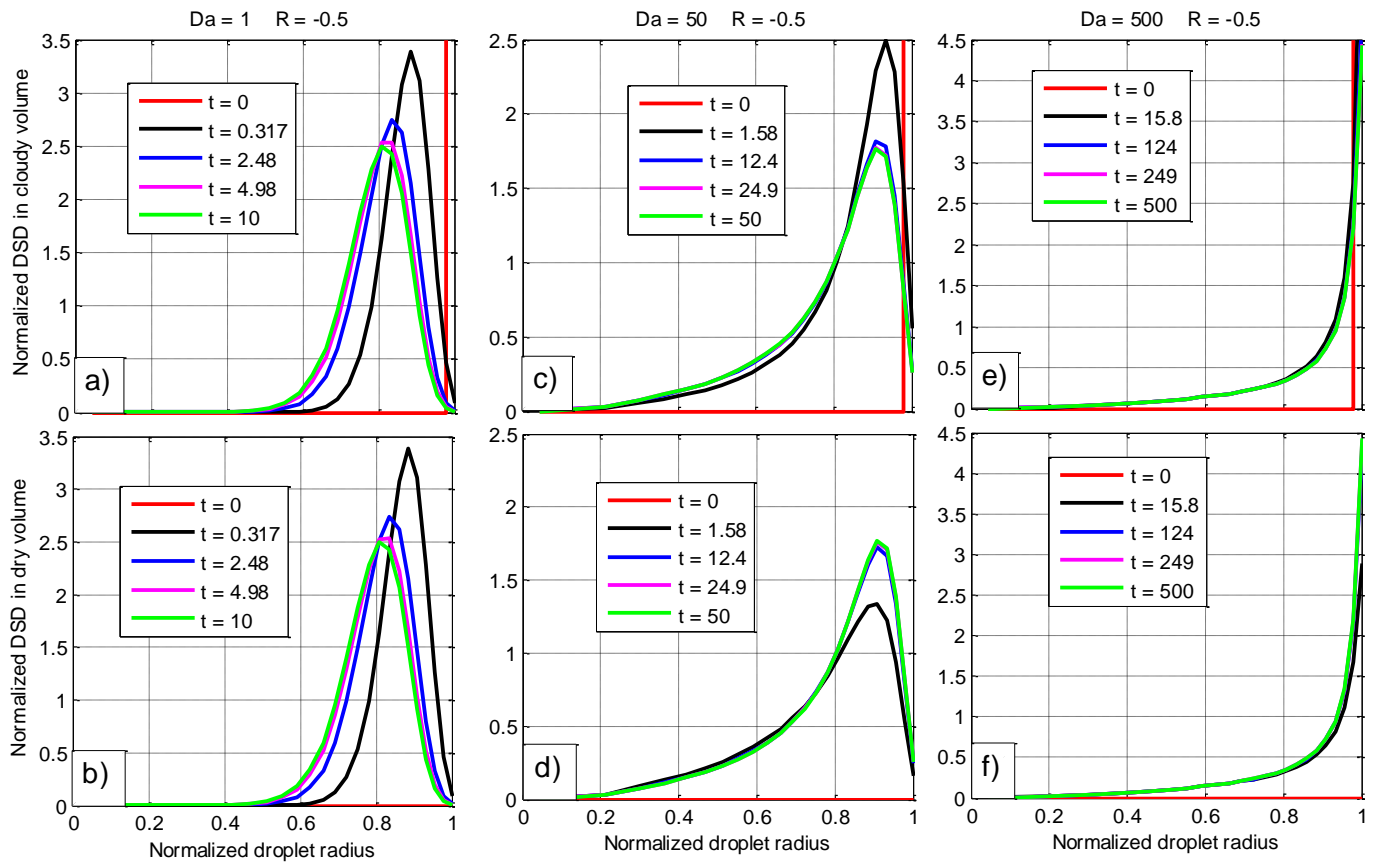
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1275 **Fig. 11** Examples of DSD evolution in the initially cloudy volume ($\tilde{x} = 1/4$) (upper row)

1276 and in the initially dry volume ($\tilde{x} = 3/4$) (lower row) at $R = -0.5$ and at different values of

1277 Da .

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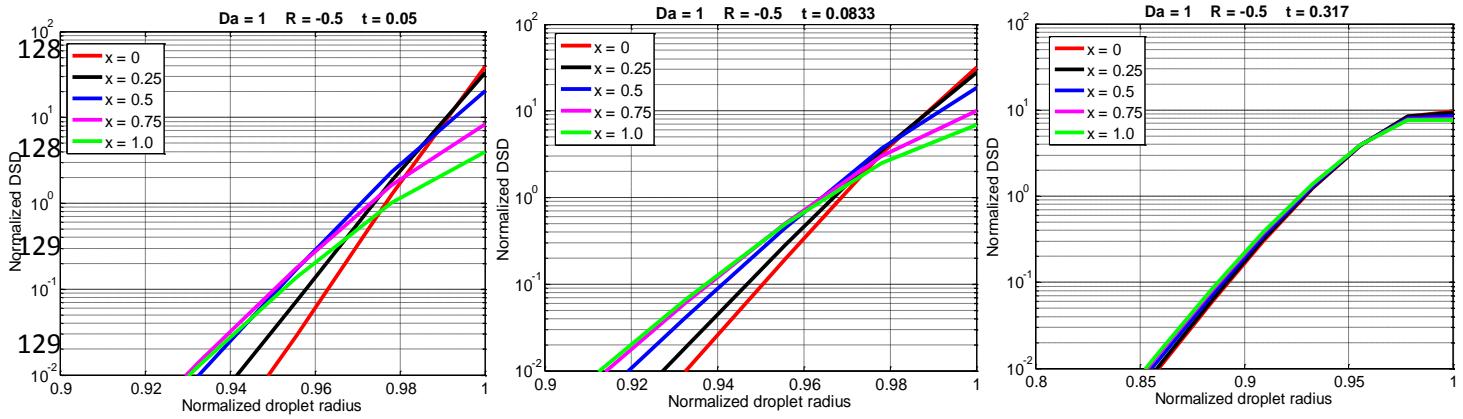
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1296 **Fig. 12.** DSD at different \tilde{x} at the beginning of the mixing process for $Da=1$ and

1297 $R = -0.5$.

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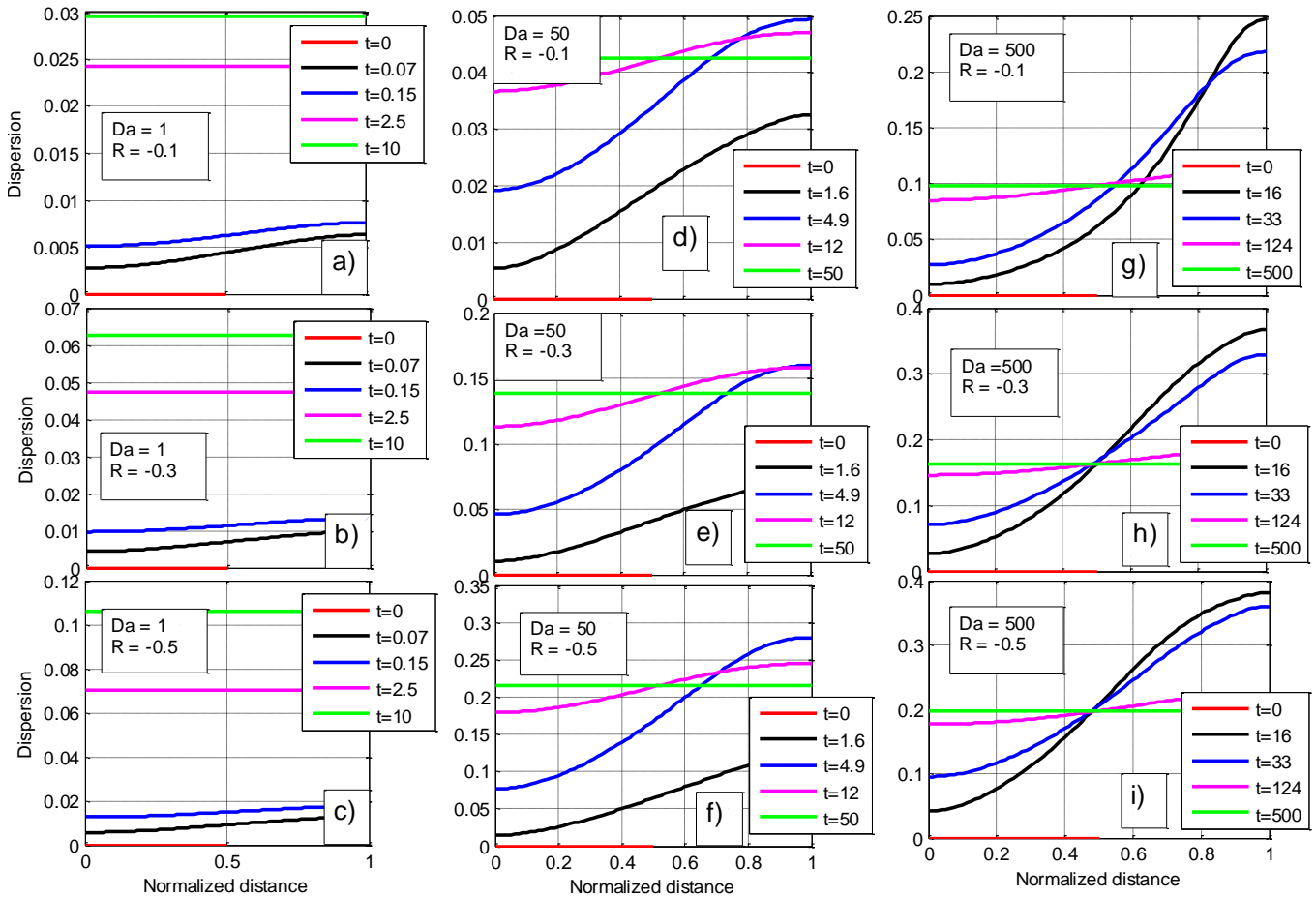
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1310 **Fig. 13.** Spatial dependencies of the relative DSD dispersion at different time instances and1311 at different values of Da and different $R > -1$

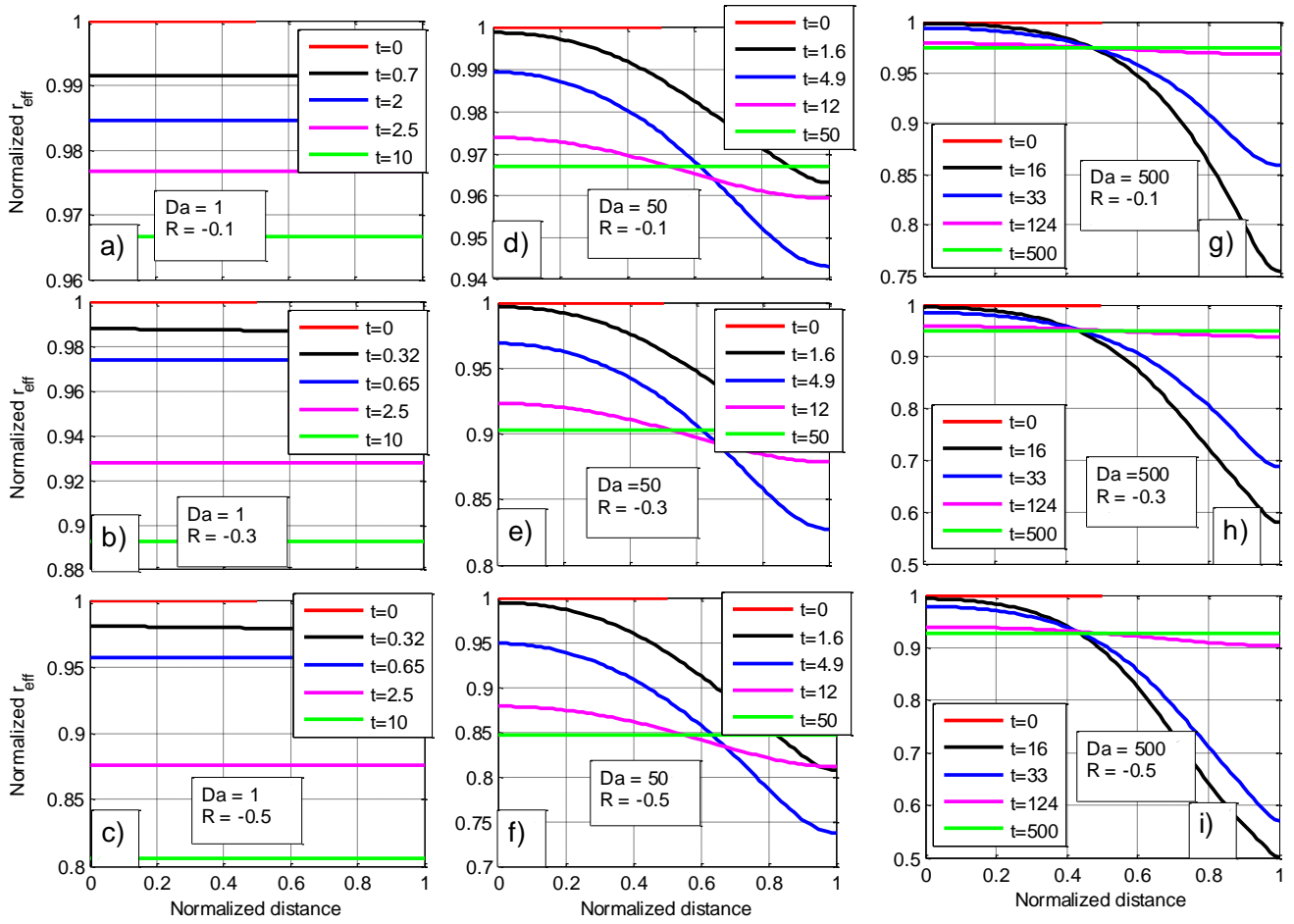
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1319 **Fig. 14.** Spatial dependencies of the effective radius at different time instances and at

1320 different values of Da and different $R > -1$

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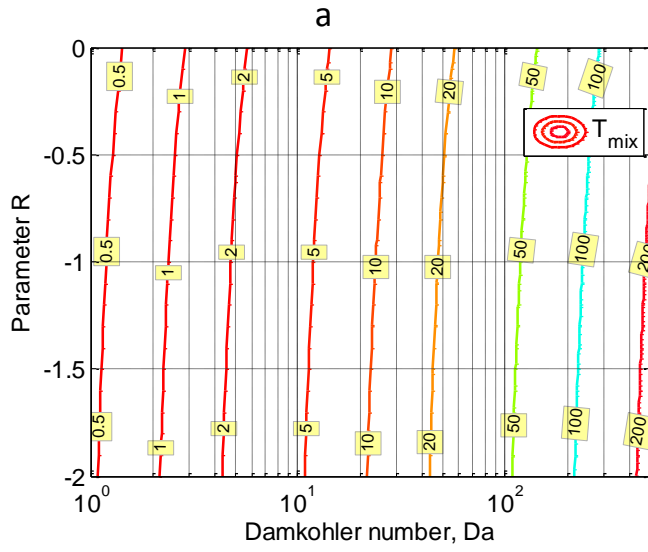
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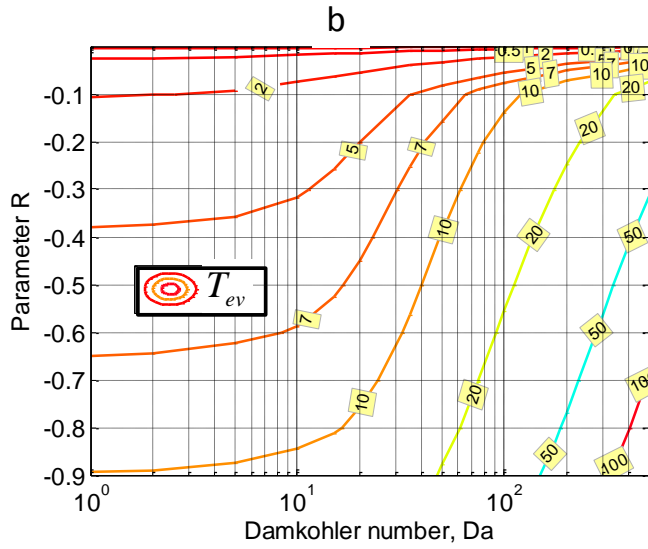
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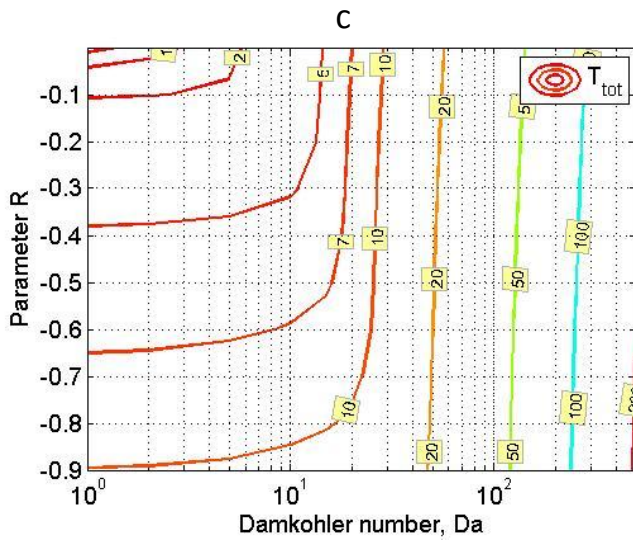
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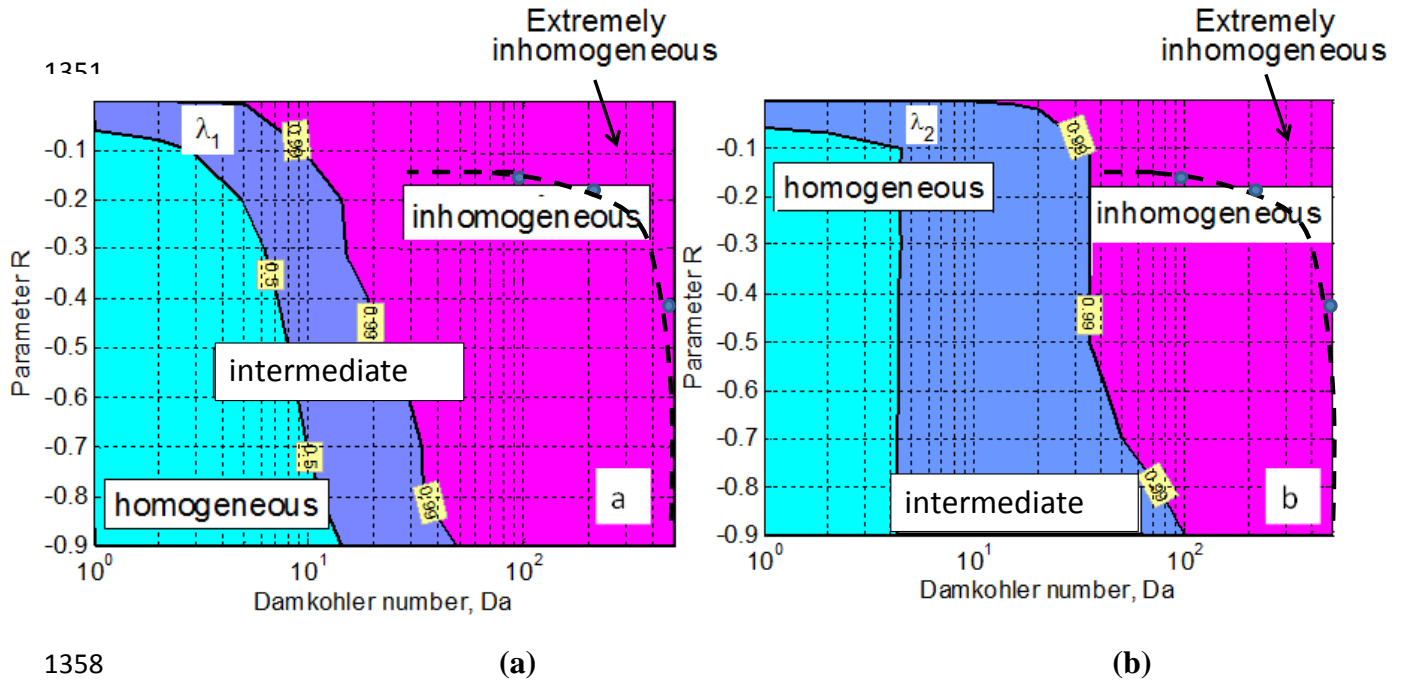


1346 **Fig. 15.** Contours of normalized mixing duration times on $Da-R$ plane. (a) mixing time
 1347 T_{mix} , (b) evaporation time T_{ev} , and (c) the total duration mixing time T_{tot} .

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1360 **Fig. 16.** (a) The boundaries between mixing types on the $Da-R$ plane designed according

1361 to criteria $\lambda_1 = \frac{T_{mix}}{T_{tot}}$; (b) The boundaries between mixing types on the $Da-R$ plane designed

1362 according to criterion $\lambda_2 = \frac{2\langle \tilde{q}(T_{mix}) \rangle - 1}{R}$ (Eq. 41). Dashed lines indicate the line corresponding

1363 to 2% deviation from the initial mean volume radius.

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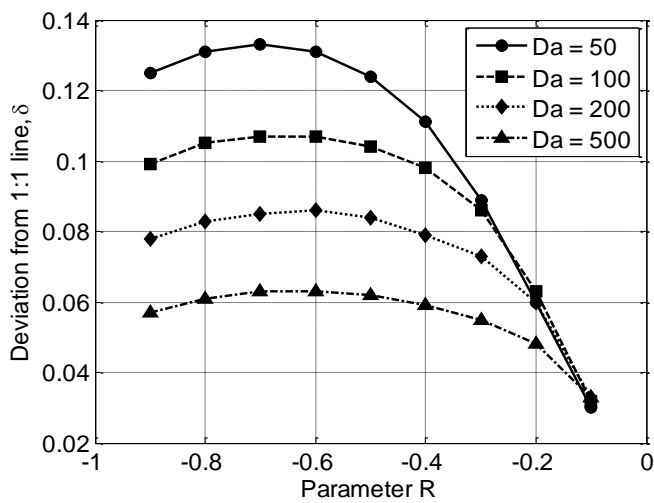
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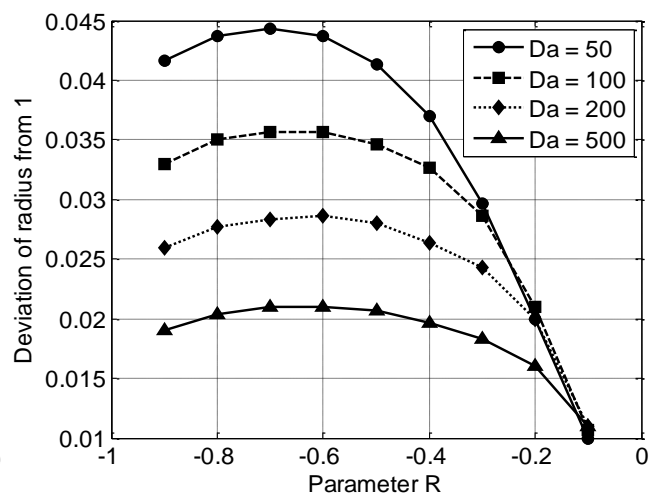
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Fig 17. (a) Dependencies of the r.m.s. distance of the $\tilde{N} - \tilde{q}$ relationship curve from straight line 1:1 suggested by classical concept of extremely inhomogeneous mixing. The dependencies are plotted for different values of Da and R . (b) The same as to the left panel but for r.m.s. deviations of the mean volume radius curve from that initial constant value assumed in the classical concept.