We express our gratitude to Referee #1 for his comments and remarks

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Interactive comment on "Theoretical analysis of mixing in liquid clouds – Part 3: Inhomogeneous mixing" by M. Pinsky et al.

Anonymous Referee #1

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Review of "Theoretical analysis of mixing in liquid clouds", in three parts. Overall recommendation: reject and encourage rewriting and resubmission. General comments to all three parts (repeated in all three reviews). I read the papers with considerable interest mostly because this seemed to be a populartopic some time ago, in both observations and modeling. I was curious to see what new these manuscripts bring. Frankly, I was disappointed.

© First, the analysis concerns a highly idealized problem, with little applications to real clouds. Turbulent mixing in clouds is by far more complicated that situations depicted in Fig. 1 of part 1 (and then repeated in different shapes as Figs. 1 in Part 2 and 3). Second, I am aware of study in which the authors developed a fairly sophisticated model of microphysical evolution during turbulent stirring (Jarecka et al., JAS 2013)aiming at prediction of the homogeneity of mixing. They applied the model to LES simulations of shallow convective cloud field. The impact was surprisingly small and the authors of that paper argued why this might be so (the entrained air comes from the descending shell and is not far from saturation). So in a sense the subject is "old
news". Finally, the lengthy discussions, full of unnecessary caveats and references to details of small multi-panel figures, made the reading frustrating. All three parts read like a student dissertation, not a concise scientific paper highlighting key points and leaving the rest for the reader to follow. Thus, I read the manuscripts with decreasing interest, and my comments are more detailed for the part 1, and get more general forparts 2 and 3.

Overall, I do not believe that the subject matter deserves close to 100 pages and close to 50 figures. I feel that the material deserves a single, short and concise manuscript, with new material clearly separated from what I feel has been discussed in the past, perhaps not at such a level of detail. Reading introductions to all three parts made me mad, because all three say basically the same thing with different language and organization. Part 1 is mostly trivial in my view, with some parts speculative and other repeating already published material (see detailed comments). Parts 2 and 3 have some aspects that perhaps deserve to be published, but it is not clear to me how useful these are (not very much in my opinion). References to aircraft

observations are vague and missing the key aspect, which is the irrelevance of an idealized problem considered by the authors to low-spatial resolution observations of a complicated multiscale natural system.

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

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

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

b) This study addresses a “highly idealized problem” and uses simplified models in order to describe cloud mixing.

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

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

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

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

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

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

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

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

The authors regret that Referee 1 overlooked all these novelties.

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

©A small technical comment: I think the terminology the papers use is not correct. The limiting cases should be referred to as homogeneous and extremely inhomogeneous mixing. Everything between the two is the inhomogeneous mixing.

® Strictly speaking, as follows form the analysis in the paper, the limiting case referred to as extremely inhomogeneous (at which drop radii do not change) corresponds to the absence of mixing (actually some mixing remains that corresponds to the pure molecular mixing). That is why we introduced the concept of extremely inhomogeneous mixing as the case when the mixing process is very slow (very high Da). Mixing with smaller Da is referred to as inhomogeneous. We also reveal parameters at which homogeneous mixing dominates in some sense over inhomogeneous. Such cases we refer to as homogeneous mixing.

Specific comments to Part 3:

General comment:

© Part 3 discusses an idealized case of the 1D diffusion between initially monodisperse condensed water volume and subsaturated cloud-free volume. Such a problem is supposed to mimic the homogenization process in the inhomogeneous mixing scenario. The authors develop a nondimensional equations and solve them. I really run out of steam to read this part carefully. Thus, my comments are even less detailed than in the case of Part 2. That said, the diagram shown in Fig 16 is interesting and with proper exposition may become useful in the development of subgrid-scale schemes for LES.

® We agree that the delimitation introduced between regimes of mixing can be useful for

LES. Corresponding discussion about the perspectives of mixing investigation is added into the conclusion section.

© How the transitions between various mixing scenarios compare to the outcome of DNS simulations reported in Andrejczuk et al (2009)? That paper is not even mentioned, but I think it is relevant, like the Krueger's EMPM model mentioned in the conclusion section.

® The approaches are quite different. In our study time evolution of DSD and their moments as well as other parameters like RH in course on mixing are investigated. Andrejczuk et al (2009) present only final results of DNS simulations in the form resembling the standard mixing diagrams.
The references to the studies by Andrejczuk et al (2009) and Krueger's (EMPM model) are included.

©And what about the Jensen et al. (JAS, 1985) predictions (not mentioned either)?

® The study by Jensen et al. (1985) assumes that cloudy air has two sources: air coming from cloud base and air coming from cloud top. A method of calculation of cloud parameters in the case of multiple penetrations of dry volumes into the cloud air is proposed. The approach is tested using a model of ascending cloud parcel. The approach and subject of investigations in the study by Jensen et al. (1985) and in the present study are quite different.

Overall, I find Part 3 the most promising and I feel that focusing on results discussed in Part 3 should be the goal of the new paper.

® We also suppose that Pt 3 contains interesting results. As regards to the new paper, see our response above.

Specific comments:

© 1. The time scale describing droplet evaporation is again taken as the phase relaxation time scale (and used to define the Domkoehler number applied in the investigation). Part 2 shows (not surprisingly) that this is the correct time scale for the homogeneous mixing. I am not convinced that the same applies to the inhomogeneous mixing.

® In our response to corresponding comment of Reviewer to Part 2 we showed that the phase relaxation time is a natural characteristic time of the mixing process, because this time determines both the time evolution of sub-saturation over ensemble of droplets and also time evolution of liquid water mass.

The fact that the drop relaxation time is the natural time scale at any type of mixing is clearly follows from the non-dimensional form of diffusion-evaporation equation showing that Da is the ratio of mixing time to the drop relaxation time.

Corresponding comments are included into the text and in the Conclusion section.

© In the limiting case of the extremely inhomogeneous mixing (which in the current setup corresponds to the mixing coefficient taken as the molecular diffusivity), the rate of the homogenization progress depends also on the initial humidity of the cloud-free volume, doesn't it? Thus, the time-scale of droplet evaporation should be some combination of the information provided by the phase relaxation time scale and the humidity of the cloud-free volume.

® The reviewer is correct. Yes, it is shown that the mixing -evaporation process depends on two parameters, Da and R. The parameter R is related to the initial humidity of the cloud-free volume. These two parameters determine process of mixing at any regime, not only in the case of the extremely inhomogeneous mixing.

© Note that the other time scale that can be used (calculated as the time required for the total evaporation of a single droplet as used in Jarecka et al. I think) excludes droplet concentration. However, droplet concentration clearly is a relevant parameter in the problem of the cloud interface propagation due to molecular diffusion in the 1D problem considered by the authors. Thus, I feel that 1D results applying molecular mixing can be used to calculate the proper time scale for the homogenization and to explore which time scale (the phase relaxation or the evaporation) is more appropriate.

® As we showed above, the characteristic time scale of the evaporation process is the phase relaxation time. This time is automatically follows from non-dimensional equations containing Da. In our 1-D problem we use averaged equations, so effects of turbulence are described using coefficient of turbulent diffusion. At the averaging scales there are clear definitions of concepts of droplet concentration, supersaturation, etc. The scales, where molecular diffusion dominates (Kolmogorov scales), are of the same order as the distance

between two droplets. At these scales the concepts of concentration, supersaturation over the droplet ensemble become uncertain. We do not consider mixing at such small scales, as it is mentioned in the paper.

We deal with turbulent mixing and with turbulent diffusion. The interface between initially cloud and droplet-free volumes in our problem is of turbulent nature. Corresponding comments are added into Section 2.

© 2. Sections 2 and 3 are in my view incomprehensible. Details of the mathematical derivations should be moved to the appendix and only key formulas should be left in the main text. Section 4 can be shortened to just a few sentences.

® We shortened Section 3. Expressions in Sections 2 should be kept in the main text because they show the physics of mixing process. Section 4 is very short. The further shortening would lead to loss of important information about method of solution.

© 3. I was not able to read through section 5. However, I noticed that the title of section 5.3 is practically the same as section 5.3.2. Shortening (!) and reorganizing is needed.

® The title of section 5.3.2 was changed. We do not see the ways to shorten the section 5 without loss of clarity.

© 4. Can the results be further synthesized? For instance, figures 6 to 9 show time evolutions of profiles across the simulation domain. Can just one such a figure be shown and outcome of other simulations be simply presented applying some measure(s) of the evolution? I think this is what Fig. 16 is showing, but honestly I was too tired reading the three parts to get the points clearly. Please simplify the discussion and streamline the presentation.

® As was shown in the study, the process of mixing depends on two non-dimansional parameters: Da and R. So, we showed time evolution of most important microphysical parameters at 3 values of Da and 3 values of R (to cover the range of changes of these parameters). Note that the time evolutions clearly show that times of reaching of final equilibrium state are quite different, so that in-situ observations hardly show only final states (as it is generally assumed in analysis of observed data using mixing diagrams).

So, we do not see the ways to shorten the Section.

We express our gratitude to Referee #2 for his valuable comments and remarks.

Comments on "Theoretical investigation of mixing in warm clouds – Part 3: Inhomogeneous mixing"

This paper, in my opinion, is the most insightful of the three parts. Analytical results are obtained for droplet response for an idealized one-dimensional mixing and growth model. The appearance of the second non-dimensional parameter R, in addition to the already known non-dimensional parameter Da. As illustrated in Figure 16, this opens up a whole new range of possible conditions for homogeneous versus inhomogeneous mixing. It may help explain why in some cases observations have seemed to favor one type of mixing limit even when it was not thought to be dominant. I consider the paper an important theoretical advance in this now old problem of homogeneous vs inhomogeneous mixing.

(R) Thank you.

General criticisms:

1. As stated already in the reviews of parts 1 and 2, the relationship between this paper and the previous two papers is weak. Especially this paper is more stand alone. The idealized case considered here is distinct, and in fact also different compared with the real atmosphere, albeit still instructive of what may be happening in the fully 3D environment with dynamical feedbacks, etc. To me, the simplicity is a strength because it allows the underlying physics to become clearer, but the assumptions and caveats still need to be stated along with those of the other two parts.

(R) The list of main assumptions and simplifications is included to Section 2.

2. The results in this paper are very difficult to interpret in places. For example, there is no clear physical interpretation of the new dimensionless parameter R. In fact, it does not even have a name (in Table 1 it is simply referred to as "non-dimensional parameter). Please come up with a descriptive name and provide some interpretation. Can you explain what range of R is realistically achievable in the atmosphere, and how it might vary with typical cloud conditions?

(R) Done. This parameter is now referred to as potential evaporation parameter (PEP). Explanations concerning this parameter are presented in Section 3 and in Conclusion.

This parameter formally varies from $-\infty$, which corresponds to the cases when either LWC =0, to zero when RH=100%. When R=-1 means that total evaporation of all droplets leads to RH=100%. Evaluations show that typical range of R is between -2 to -0.1. To evaluate the value of R it is necessary to know LWC and RH in cloud surrounding.

3. This is not so much a criticism as a recommendation: can you, based on the findings of parts 2 and 3, suggest several specific measurements that could allow the type of mixing in a cloud to be identified with less ambiguity than is currently possible? Put another way, how can the central findings be experimentally verified? This kind of discussion will be very helpful in connecting the theoretical results to the experimental and observational parts of the field.

(R) Corresponding discussion is added into the conclusion section.

Specific comments

1. Figure 1 seems redundant with the previous sections, and is so simple that it can probably be deleted and instead described in words.

(R) fig 1 illustrate the design of the problem.

2. Check the sign of the second term in Equations 13, 14, 17, 18. It think they should be the opposite.

(R) Thank you. Corrected.

3. Same comment for q and LWC as in Parts 1 and 2.

(R) done

4. Same comment for S as in Part 2.

(R) done

© 5. Page 30343, line 10: “very” should be “vary”.

® The sentence is corrected

© 6. Page 30347, line 26-28: “at high R”, should be “at lower R” or “at higher |R|”? Also check other places in the text. For example: Page 30350, line 18: should be “lower |R|”? Page 30351, line 15: should be “small |R|”?

® We checked the values. We added the comment that since R<0, larger R is that closer to zero.

© 7. Add labels a) and b) in Figure 17.

® done

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

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Abstract

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

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

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

Keywords: homogeneous and inhomogeneous mixing, turbulent diffusion, droplet evaporation

1. Introduction

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

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

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

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

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

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

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

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

382 In analyses of in-situ measurements, the observed data are usually compared with those
383 expected at the final state of mixing as assumed by the classical mixing concepts. If droplet
384 concentration decreases without a corresponding change in the characteristic droplet radius,
385 the mixing is considered "extremely inhomogeneous." If the characteristic droplet radius
386 decreases with an increase of the dilution level while droplet concentration decreases
387 insignificantly, the mixing is identified as "homogeneous." If both the characteristic droplet
388 radius and the droplet concentration change, the mixing is considered as "intermediate".
389 Quantitative evaluations of the microphysical processes specific for intermediate mixing
390 remain largely uncertain.

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

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

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

2. The main concept and the basic equations

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

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

b) the total mixing volume is assumed adiabatic.

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

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

Other assumptions and simplifications are discussed below.

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

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

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

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

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

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

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

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

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

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

where q_v is the water vapor mixing ratio.

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

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

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

$$\Gamma = S + A_2 q \tag{4}$$

This value is conservative in a moist adiabatic process, i.e. it does not change during phase 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 function of temperature that changes by ~10 % when temperatures change by ~10 ^{o}C (Pinsky et al., 2013). In this study, it is assumed that $A_2 = constant$. In Eq. (4), $q = \frac{4\pi\rho_w}{3\rho_a}\int_0^\infty r^3 f(r)dr$ is the liquid water mixing ratio and $f(r)$ is the DSD. The quantity Γ obeys the diffusion equation

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

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

$$\Gamma(x,0) = \begin{cases} A_2 q_1 & \text{if} \quad 0 \le x < L/2 \\ \mathrm{S}_2 & \text{if} \quad L/2 \le x < L \end{cases} \tag{6}$$

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

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

$$\Gamma(x,t) = \sum_{n=0}^{\infty} a_n \exp\left(-\frac{Kn^2\pi^2 t}{L^2}\right)\cos\left(\frac{n\pi x}{L}\right) = \frac{1}{2}\left(S_2 + A_2 q_1\right) + (A_2 q_1 - S_2)\sum_{n=1}^{\infty}\frac{\sin(n\pi/2)}{n\pi/2}\exp\left(-\frac{Kn^2\pi^2 t}{L^2}\right)\cos\left(\frac{n\pi x}{L}\right) \tag{7}$$

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

$$a_0 = \frac{1}{2}\left(A_2 q_1 + S_2\right) \tag{8a}$$

$$a_n = (A_2 q_1 - S_2)\frac{\sin(n\pi/2)}{n\pi/2},\ n = 1,2,... \tag{8b}$$

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

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

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

$$\frac{d\sigma}{dt} = \frac{2S}{F} \tag{9}$$

where $\sigma = r^2$ is the square of droplet radius and $F = \frac{\rho_w L_w^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_S(T)\mathscr{D}}$. The value of coefficient F is considered constant in this study. The solution of Eq. (9) is

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

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

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

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

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

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

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

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

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

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

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

$$S(x,t) = \Gamma(x,t) - A_2 q(x,t) \tag{15}$$

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

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

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

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

511 $$\frac{\partial m_\alpha(x,t)}{\partial t} = K\frac{\partial^2 m_\alpha(x,t)}{\partial x^2} + \alpha\frac{2S}{F}m_{\alpha-1}(x,t) \quad (17)$$

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

515 In particular, the equation for the liquid water mixing ratio that is a moment of the order of
516 $\alpha = \frac{3}{2}$ can be written as

517 $$\frac{\partial q(x,t)}{\partial t} = K\frac{\partial^2 q(x,t)}{\partial x^2} + \frac{4\pi\rho_w N(x,t)\overline{r}(x,t)}{F\rho_a}S(x,t) \quad (18)$$

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

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

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

523 $$\tau_{pr} = \frac{\rho_a F}{4\pi\rho_w A_2 N\overline{r}} \quad (19)$$

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

525 $$\begin{aligned}\frac{\partial q(x,t)}{\partial t} &= K\frac{\partial^2 q(x,t)}{\partial x^2} + \frac{1}{\tau_{pr}(x,t)}\left[\frac{1}{A_2}\Gamma(x,t) - q(x,t)\right] = \\ &= K\frac{\partial^2 q(x,t)}{\partial x^2} + \frac{1}{A_2\tau_{pr}(x,t)}S(x,t)\end{aligned} \quad (20)$$

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

528 $$\frac{\partial S(x,t)}{\partial t} = K\frac{\partial^2 S(x,t)}{\partial x^2} - \frac{S(x,t)}{\tau_{pr}(x,t)} \quad (20a)$$

529 Eqs. (20) and (20a) show that changes in the microphysical variables are determined by the rate
530 of spatial diffusion (the first term on the right-hand side of these equations) and of evaporation
531 (the second term on the right-hand side).
532
533 **3. Analysis of non-dimensional equations**
534 Spatial diffusion and evaporation depend on many parameters. It is the best to start the
535 analysis from the basic equation system presented in a non-dimensional form. A time scale
536 corresponding to the initial phase relaxation time in a cloud volume can be defined as

537 $$\tau_0 = \frac{\rho_a F}{4\pi\rho_w A_2 N_1 r_0} \qquad (21)$$

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

540 $$\tilde{\tau}_{pr} = \tau_{pr}/\tau_0 = \frac{N_1 r_0}{N(\tilde{x},\tilde{t})\bar{r}(\tilde{x},\tilde{t})}, \qquad (22a),$$

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

542 $$\tilde{q} = \frac{q}{q_1}, \qquad (22b),$$

543 the normalized supersaturation

544 $$\tilde{S} = \frac{S}{A_2 q_1} \qquad (22c),$$

545 the non-dimensional conservative function

546 $$\tilde{\Gamma} = \frac{\Gamma}{A_2 q_1}, \qquad (22d),$$

547 the normalized square of droplet radius

548 $$\tilde{\sigma} = \frac{\sigma}{r_0^2}, \qquad (22e),$$

549 the normalized droplet concentration

550 $$\tilde{N} = N/N_1 \qquad (22f)$$

551 and the non-dimensional DSD

552 $$\tilde{g}(\tilde{\sigma}) = \frac{r_0^2}{N_1} g(\sigma) \qquad (22g).$$

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

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

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

556 $$\tilde{x} = x / L; \quad \tilde{t} = t / \tau_0 \qquad (22h)$$

557 A widely used non-dimensional parameter showing the comparative rates of diffusion and

558 evaporation is the Damkölher number:

559 $$Da = \frac{\tau_{mix}}{\tau_0} = \frac{L^2}{K\tau_0} \qquad (23)$$

560 where

561 $$\tau_{mix} = \frac{L^2}{K} \qquad (24)$$

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

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

564

565 $$\frac{\partial \tilde{q}(\tilde{x},\tilde{t})}{\partial \tilde{t}} = \frac{1}{Da}\frac{\partial^2 \tilde{q}(\tilde{x},\tilde{t})}{\partial \tilde{x}^2} + \frac{1}{\tilde{\tau}_{pr}(\tilde{x},\tilde{t})}\left[\tilde{\Gamma}(\tilde{x},\tilde{t}) - \tilde{q}(\tilde{x},\tilde{t})\right] = \frac{1}{Da}\frac{\partial^2 \tilde{q}(\tilde{x},\tilde{t})}{\partial \tilde{x}^2} + \frac{1}{\tilde{\tau}_{pr}(\tilde{x},\tilde{t})}\tilde{S}(\tilde{x},\tilde{t}) \qquad (25)$$

566 where

567 $$\tilde{q}(\tilde{x},\tilde{t}) = \frac{N(\tilde{x},\tilde{t})\overline{\sigma^{3/2}}}{N_1 r_0^3} = \int_0^\infty \tilde{\sigma}^{3/2} \tilde{g}(\tilde{x},\tilde{t},\tilde{\sigma}) d\tilde{\sigma} \qquad (26)$$

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

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

570 $\tilde{q}(\tilde{x},0)$ can be derived from Eqs. (1c) and (22b)

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

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

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

where

$$R=\frac{S_2}{A_2q_1} \tag{29}$$

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

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

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

The solution of Eq. (25) and the type of mixing depends on the values of two non-dimensional parameters, namely, Da and R. Thus, when $R=\frac{S_2}{A_2q_1}<-1$, $\tilde{\Gamma}(\tilde{x},\infty)<0$. It means that the initially droplet-free volume V_2 is too dry and all the droplets in the mixing volume evaporate completely. At the final equilibrium state $RH<100\%$, i.e. $S(x,\infty)<0$. If $R=\frac{S_2}{A_2q_1}>-1$, $\tilde{\Gamma}(\tilde{x},\infty)>0$. This means that the mixed volume in the final state contains droplets, i.e. the mixing leads expands the volume with droplets, i.e. the cloud volume. At the

592 final equilibrium state, $RH = 100\%$ (i.e. $S(x,\infty) = 0$). The case when $|R| = \left|\frac{S_2}{A_2 q_1}\right| = |\tilde{S}_2| << 1$

593 corresponds to either RH close to 100% (i.e. S_2 is close to zero) (this case corresponds to the
594 degenerated case considered in Pt. 1), and/or to the case when the liquid water mixing ratio in
595 the cloud volume is large. In case $|R| << 1$, the second term on the right-hand side of Eq. (25) is
596 much smaller than the first term, and the mixing is driven by turbulent diffusion only.

597 In case $Da \to 0$ (often considered as homogeneous mixing), at the beginning of the mixing
598 the diffusion term is much larger than the evaporation term, the second term on the right-hand
599 side of Eq. (25). As mixing proceeds, within a short time period the total homogenization of all
600 the variables in the mixing volume is established and all the spatial gradients become equal to
601 zero. At this time instance, the first term on the right-hand side becomes equal to zero, and the
602 second term on the right-hand side of Eq. (25), describing droplet evaporation, becomes
603 dominant. Thus, the analysis of the Eq. (25) shows that mixing consists of two stages. The first
604 mixing stage is a short stage of inhomogeneous mixing and the longer second stage of
605 homogeneous mixing. The evolution of the microphysical variables during homogeneous
606 mixing is described in detail in Pt. 2.

607 $Da \to \infty$ corresponds to extremely inhomogeneous mixing, according to the classic
608 concept. In this case, the diffusion term is much smaller than the evaporation term, so
609 evaporation takes place under significant spatial gradients of RH. At $Da = \infty$, the adjacent
610 volumes do not mix at all and remain separated. This equivalent to existence of two
611 independent adiabatic volumes. Another interpretation of the limiting case $Da = \infty$ is an
612 infinite fast droplet evaporation. Both scenarios at $Da \to \infty$ indicate simplifications in the
613 definition of the extremely inhomogeneous mixing. At intermediate values of Da, mixing is
614 inhomogeneous, when both turbulent diffusion and evaporation contribute simultaneously to
615 formation of the DSD.

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

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

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

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

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

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

4. Design of simulations

Damköller number Da ***in clouds***

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

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

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

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

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

649

650 ***Numerical method***

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

654

655 $$\frac{\partial \tilde{g}(0,\tilde{t},\tilde{\sigma}_j)}{\partial \tilde{x}} = \frac{\partial \tilde{g}(1,\tilde{t},\tilde{\sigma}_j)}{\partial \tilde{x}} = 0 \qquad (34)$$

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

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

660 $$\tilde{S}(\tilde{x},\tilde{t}) = \tilde{\Gamma}(\tilde{x},\tilde{t}) - \tilde{q}(\tilde{x},\tilde{t}) \qquad (35)$$

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

662 $$\frac{2}{3}\tilde{S}(\tilde{x},\tilde{t})\frac{\partial \tilde{g}(\tilde{x},\tilde{t},\tilde{\sigma}_j)}{\partial \tilde{\sigma}_j} \approx \frac{\tilde{g}\left(\tilde{x},\tilde{t},\tilde{\sigma}_j + \frac{2}{3}\tilde{S}\Delta\tilde{t}\right) - \tilde{g}\left(\tilde{x},\tilde{t},\tilde{\sigma}_j\right)}{\Delta\tilde{t}} \qquad (36)$$

663 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

664 $\Delta\tilde{t}$ is a small time increment chosen so that $\left|\frac{2}{3}\tilde{S}_{\max}\Delta\tilde{t}\right| \le \frac{\Delta\tilde{\sigma}}{2}$. Next, the shifted DSD was

665 remapped onto the fixed square radius grid $\tilde{\sigma}_j$. We used the remapping method proposed by

666 Kovetz and Olund (1969), which conserves droplet concentration and LWC. After remapping,

667 the differences between the new and old DSDs were recalculated. The new values of LWC then

668 were determined using new values of DSD and Eq. (26). MATLAB utility PDEPE

669 automatically chooses the time step needed to provide stability of calculations.

670

671 **5. Results of simulations**

672 **5.1 Full evaporation case**

673 First, we consider the case $R = -1.5$, when all the cloud water evaporates completely. This

674 process corresponds to the cloud dissipation caused by mixing with the entrained dry air. At the

675 final state, RH is expected to be uniform and negative over the entire mixing volume.

676 **Figure 3** shows spatial and time changes of $\tilde{S}$ for $Da = 1$, 50 and 500. At the final state

677 for all the three cases $\tilde{S}$ =-0.25, which is in agreement with the analytical solution of Eq. (30).

678 The final negative value indicates that all the droplets completely evaporated during mixing. At

679 $Da = 1$ (Fig.3ab), two stages of supersaturation evolution can be identified. The first short

680 stage with $t < 0.4\tau_{pr}$ is the period of inhomogeneous mixing, when the gradients of RH

681 persist. By end of the second stage of about $14\tau_{pr}$, the equilibrium state is reached. Thus, at

682 small Da both types of mixing take place. In the cases of $Da = 50$ and $Da = 500$, the spatial

683 gradients exit during the entire period of mixing until the equilibrium state is reached

684 (approximately $50\tau_{pr}$ and $300\tau_{pr}$, respectively) (Fig.3cdef). Therefore, at these Da mixing is

685 inhomogeneous during entire mixing.

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

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

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

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

711 $Da = 500$, homogenization of the saturation deficit requires $250\tau_{pr}$, which is twice as long as
712 the time of complete droplet evaporation, i.e. $T_{mix} \approx 2T_{ev}$. This means that at $Da = 500$, droplet
713 evaporation takes place in the presence of the spatial gradients of supersaturation. After
714 complete evaporation of droplets, spatial gradients of the water vapour mixing ratios remain.
715 This kind of mixing is regarded as inhomogeneous.

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

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

728 The behaviour of the $\tilde{N} - \tilde{q}$ relationship provides important information about mixing
729 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
730 different $\tilde{x}$. This means that the three curves at $\tilde{t} < T_{mix}$ do not coincide. At $\tilde{t} > T_{mix}$, the spatial
731 gradients of $\tilde{N}$ and $\tilde{q}$ disappear and the three curves coincide. When the curves do not
732 coincide, mixing is inhomogeneous, and the coincidence of the curves indicates that the mixing
733 becomes homogeneous. In Fig. 5a and 5b ($Da = 1$ and $Da = 5$, respectively), the curves
734 coincide at point A corresponding to time $\tilde{t} = T_{mix}$.

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

745 At $Da = 50$ (Fig 5c), curves corresponding to different values of $\tilde{x}$ do not coincide, except
746 at the final point "F", where $\tilde{N} = 0$ and $\tilde{q} = 0$. This means that horizontal gradients exist during
747 the entire mixing process and mixing is inhomogeneous till the final equilibrium state is
748 reached. Droplets penetrating into the initially droplet-free volume begin evaporating, so only a
749 small fraction of droplets reaches the centre of the droplet-free volume, as seen in Fig. 5c,
750 $\tilde{x} = 3/4$ (black curve). Accordingly, at $\tilde{x} = 3/4$ the droplet concentrations and $\tilde{q}$ reach their
751 maxima (of 0.1 and 0.05, respectively) and then decrease to zero. At $Da = 500$ (Fig 5d), all
752 the droplets evaporate before reaching the centre of the dry volume, indicating an extremely
753 high spatial inhomogeneity of droplet evaporation. Hence, only two curves for $\tilde{x} = 1/4$ and
754 $\tilde{x} = 1/2$ are seen in Fig.5d.

755 Fig. 5 also shows that the slopes of the curves describing the $\tilde{N} - \tilde{q}$ relationships are
756 different at different values of $\tilde{x}$ and change over time. At large Da, the slopes of the curves
757 describing the dependencies $\tilde{N} - \tilde{q}$ in the initially cloud volume are close to linear. However,
758 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
759 to the fact that when $\tilde{q}$ is large, it decreases faster than the concentration $\tilde{N}$ because some

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

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

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

780 At $Da=50$, DSD are different at different $\tilde{x}$ during the entire period of mixing. While
781 DSD at $\tilde{x}>0.5$ are wide and droplet evaporation is accompanied by a shift of DSD maximum
782 to smaller droplet radii (this feature is typically attributed to homogeneous mixing), the DSD
783 maximum at $\tilde{x}<0.5$ (the initially cloud volume) shifts toward smaller radii only slightly until
784 $\tilde{t}=3.17$ (Fig. 6e). Further droplet evaporation either leads to a complete evaporation (at

785 $\tilde{x} \geq 0.5$) or shifts the DSDs to smaller droplet sizes (panel f). The maximum droplet
786 concentration takes place at $\tilde{x} = 0$. Fig. 6 shows that DSD shapes evolve substantially over
787 time, although the final state is characterized by complete droplet evaporation.
788
789 **5.2 Partial evaporation case**
790 **5.2.1 Evolution of the microphysical parameters at different values of Da and R**
791 Here we consider the process of mixing at $R > -1$, i.e. when not all the droplets evaporate
792 completely. **Figure 7** shows the horizontal profiles of a normalized supersaturation at different
793 Da and R. One can see that in all cases, the final state occurs when the equilibrium
794 supersaturation $\tilde{S} = 0$ (RH=100%). However, this final value is reached quite differently
795 depending on Da. At $Da = 1$, rapid mixing leads to formation of spatially homogeneous
796 humidity and supersaturation during a time period of a fraction of τ_{pr}. Then, supersaturation
797 within the mixing volume grows by evaporation of droplets, which are uniformly distributed
798 over the entire mixing volume. This process of homogeneous mixing was analyzed in detail in
799 Pt. 2.
800 At $Da = 500$, changes in supersaturation take place largely within the initially droplet-free
801 volume. RH in the initially cloud volume undergoes only small changes. This process agrees
802 well with the classical concept of extremely inhomogeneous mixing. However, a strong
803 gradient of supersaturation remains within the initially drop-free volume for a long time (tens
804 of τ_{pr}). At $Da = 50$, the situation is intermediate. Mixing is intensive enough to decrease RH
805 in the initially cloud volume, but spatially uniform RH is established within about 5-10 τ_{pr},
806 increasing with an increase in $|R|$. After this time instance, mixing takes place according to the
807 homogeneous scenario.
808 **Figure 8** shows the horizontal profiles of normalized LWC at different Da and R. At the
809 same R, the final equilibrium values of LWC are identical, as follows from Eq. (30); LWC

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

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

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

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

834 At $R=-0.3$ a decrease in the droplet concentration takes place only at $Da=500$, and at
835 $R=-0.5$ the droplet concentration decreases already at $Da\geq 50$.

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

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

852

853 **5.2.2 Evolution of DSDs and the DSD parameters**

854 **Figure 11** presents examples of the DSD evolution at the center of the initially cloud
855 volume ($\tilde{x}=1/4$) (upper row) and of the initially droplet-free volume ($\tilde{x}=3/4$) at $R=-0.5$
856 and different values of Da. Several specific features of the DSD are notable. As a result of the
857 rapid mixing at $Da=1$ (left column), DSD become similar in both volumes already at
858 $t=0.317\tau_{pr}$ (black lines). Further evolution is similar in both volumes and is characterized by
859 broadening of the DSD and its shifting and of the DSD toward smaller droplet sizes. This shift

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

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

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

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

886 Observed DSD dispersion in different clouds typically ranges from 0.1 to 0.4 (Khain et al.,
887 2000; Martin et al., 2004; Prabha et al., 2012) and can be caused the following factors: in-
888 cloud nucleation (e.g. Khain et al., 2000; Pinsky and Khain, 2002), spatial averaging along
889 aircraft traverses (Korolev, 1995) and non-symmetry in droplet nucleation/denucleation
890 (Korolev, 1995). As seen in Fig. 13, this dispersion may be also caused by mixing at cloud
891 edges at moderate and large Da. Hence, inhomogeneous mixing leads to DSD broadening.

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

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

905

906 **5.3 Delimitation between mixing types**

907 Typically, the Da value is used as a criterion for delimitation between mixing types.
908 $Da = 1$ is usually used as a boundary value separating homogeneous and inhomogeneous
909 mixing. As shown in Section 4, mixing always starts as inhomogeneous. In the course of

910 mixing, the initial spatial gradients decrease and the air volumes either become identical or
911 remain different. In the former case, the second mixing stage is homogeneous. If
912 inhomogeneity persists until the equilibrium state is established, mixing remains
913 inhomogeneous during the entire period. Both mixing stages can be characterized by duration,
914 change in the droplet concentrations or LWCs, and other quantitative characteristics. These
915 characteristics are functions of two non-dimensional parameters R and Da, which can be
916 calculated and used for delimitation between mixing types. Since mixing between volumes
917 may turn from inhomogeneous into homogeneous before reaching the equilibrium state, it is
918 necessary to use some quantitative criteria to delimit mixing types. Below, delimitation is
919 performed for $R > -1$ which corresponds to partial evaporation of droplets by the end of
920 mixing.
921

922 **5.3.1. Characteristic time periods of mixing**

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

928 We use solution (28) for conservative function $\tilde{\Gamma}(\tilde{x},\tilde{t})$ to define quantitatively time period
929 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 $\tilde{t} \to \infty$ can be
930 approximately estimated using the first term of the series expansion as

931
$$\left|\Delta\tilde{\Gamma}\right|_{\max} \approx \left|(1-R)\frac{\sin(\pi/2)}{\pi/2}\exp\left(-\frac{\pi^2\tilde{t}}{Da}\right)\cos(\pi\tilde{x})\right|_{\max} = (1-R)\frac{2}{\pi}\exp\left(-\frac{\pi^2\tilde{t}}{Da}\right) \quad (37)$$

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

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

Suppose the value of the maximum deviation is $\left|\Delta\tilde{\Gamma}\right|_{\max} = 0.02$. This is a small value 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 duration of the non-homogeneous stage is evaluated as

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

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

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

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

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

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

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

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

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

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

Another criterion of delimitation between mixing types can be determined from a comparison of LWC variation rates due to different mechanisms. The mean normalized LWC (which is equal to the mean normalized liquid water mixing ratio) can be written as integral $\langle \tilde{q}(\tilde{t}) \rangle = \int_0^1 \tilde{q}(\tilde{x},\tilde{t}) d\tilde{x}$. The initial mean LWC is equal to $\langle \tilde{q}(t=0) \rangle = \frac{1}{2}$. The final equilibrium LWC is equal to $\langle \tilde{q}(t=\infty) \rangle = \frac{1}{2}(1+R)$ (Eq. (30)). The total amount of liquid water that evaporates in the course of mixing can be quantified by the difference between these two 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 first inhomogeneous mixing stage is calculated by the equation $\langle \tilde{q}(t=0) \rangle - \langle \tilde{q}(T_{mix}) \rangle = \frac{1}{2} - \langle \tilde{q}(T_{mix}) \rangle$. Hence, parameter λ_2 which is a ratio of

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

can serve as another possible criterion for delimitation between mixing types. This ratio characterizes the fraction of liquid water that evaporates at the initial inhomogeneous stage. Condition $\lambda_2 < 0.5$ in this case corresponds to homogeneous mixing, while condition $0.5 \le \lambda_2 < 1$ corresponds to intermediate mixing. We regard the case $\lambda_2 = 1$ as inhomogeneous mixing. Certainly, criterion λ_2 depends on the non-dimensional parameters R and Da. Fig. 16b illustrates delimitation between mixing types on the $Da - R$ plane according to criterion λ_2.

Comparison of Figs. 16a and 16b shows that both criteria lead to nearly similar separation of the $Da - R$ plane into three zones corresponding to homogeneous, intermediate and inhomogeneous mixing. At the same time, the boundaries separating these zones are different depending on the delimitation criterion used. Nevertheless, it can be concluded that mixing can

1002 be considered homogeneous at Da below 4-10 and $R < -0.1$ and inhomogeneous at Da
1003 exceeding several tens.

1004 Terms "inhomogeneous mixing" (Burner and Brenguier, 2007) and "extremely
1005 inhomogeneous mixing" (Lehmann et al., 2009; Gerber et al., 2008; Pt1) are used to denote the
1006 mixing regime when the relationship between the normalized values $\tilde{N}$ and $\tilde{q}$ is represented
1007 by a straight 1:1 line, which is equivalent to the constant mean volume radius (in some studies,
1008 the effective radius is used instead of the mean volume radius. According to the definition used
1009 in the present study, extremely inhomogeneous mixing is the limiting case of inhomogeneous
1010 mixing when $Da \to \infty$. Despite the fact that the extremely inhomogeneous mixing is only a an
1011 idealization our approach allows to determine to what extent mixing can be considered to be
1012 close to this limiting case. The measure of inhomogenity of mixing is the closeness of the
1013 $\tilde{N} - \tilde{q}$ relationship to the 1:1 straight line (see discussion above related to Figs. 5 and 10).

1014 **Figure 17a** shows r.m.s. distance between the $\tilde{N} - \tilde{q}$ relationship and the 1:1 straight line,
1015 depending on Da and R. These dependences were calculated using the set of points $\tilde{N}_i, \tilde{q}_i$
1016 uniformly distributed over spatial interval $0 \div 1$ and time interval $0 \div T_{tot}$. The equation for
1017 estimation is $\delta = \sqrt{\frac{1}{2M}\sum_{i=1}^{M}\left(\tilde{N}_i - \tilde{q}_i\right)^2}$, where M is the total number of points. This distance
1018 corresponds to r.m.s. deviation of the normalized mean volume radius from 1. The dependences
1019 of the last deviation on Da and R and estimated as $\delta/3$ are shown in Fig. 17b. This estimation
1020 is based on the fact that the total mass of droplets is proportional to the cube of the mean
1021 volume radius. As expected, the distance decreases with increasing in Da. At large R, all the
1022 curves coincide indicating a degenerative case when type of mixing becomes indistinguishable.

1023 We choose the value $\delta/3$ equal to 0.02 to determine the boundary of the extremely
1024 inhomogeneous mixing zone. The value of 0.02 corresponds to droplet radii deviation of a few
1025 tenths of a micron, which is so low that in in-situ measurements this case would always be
1026 attributed to extremely inhomogeneous mixing. In Fig.16 this boundary is marked by broken

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

6. Summary and conclusions

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

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

1053 when homogeneous and inhomogeneous mixing cannot be distinguished (see also Pt. 1). At
1054 $R = 0$, the droplet population turns into a passive admixture and its turbulent diffusion will be
1055 the same as different thermodynamic parameters.

1056 The second parameter is the *Damkölher* number (Da) which is the ratio between the
1057 characteristic mixing time and the phase relaxation time. This parameter compares the rates of
1058 spatial diffusion and evaporation. Parameter Da (Eq. (23)) is logically appears in the non-
1059 dimensional form of the diffusion-evaporation equation showing that Da is the ratio of the
1060 mixing time defined as $\tau_{mix} = \frac{L^2}{K}$, to the initial drop relaxation time. The expression for this non-
1061 dimensional parameter clearly shows that since we consider an ensemble of evaporating droplets,
1062 the drop relaxation time evaluated just before the mixing is the characteristic time scale of
1063 inhomogeneous mixing process. In several studies (e.g., Baker and Latham, 1979; Burnet and
1064 Brenguier, 2007; Andejchuk et al., 2009) a question was raised as to which time scale should be
1065 used in formulation of the *Damkölher* number: the time of an individual droplet evaporation at
1066 constant saturation deficit, or the phase relaxation time. This study, as well Pt. 2 show that the
1067 phase relaxation time is the answer. The mixing time is introduced via the turbulent diffusion
1068 coefficient which is a natural measure characterizing the diffusion rate and, in particular,
1069 determines the propagation rate of the fronts in the fields of droplet concentration and other
1070 microphysical parameters. The turbulent diffusion coefficient is widely used to describe mixing
1071 in cloud models at resolved scales.

1072 The analysis was performed within a wide range of Da (from 1 to 500) and of R (from -
1073 1.5 to -0.1). The final LWC and the humidity in the mixing volume are determined by the mass
1074 conservation and do not depend on Da (see also Pt. 1 and Pt. 2). At the same time, the droplet
1075 concentration, as well as the shape of DSD and their parameters strongly depend on Da.

1076 It is shown that the mixing of air volumes with initially different thermodynamical and
1077 microphysical parameters consists of two stages characterized by two time periods: the time

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

This finding allows to delimit between mixing types. We presented two quantitative criteria on the $Da - R$ plane that allow to delimit three mixing regimes: homogeneous, intermediate and inhomogeneous. These criteria are based on comparison of the characteristic duration mixing and the evaporation rates. According to the criteria, at Da below about 5, mixing can be regarded as homogeneous, i.e. the main microphysical changes take place during the homogeneous stage. At $5 < Da < 30 \div 50$, the changes in the microphysical parameters are more significant at the inhomogeneous stage than at the homogeneous stage. In this case, the mixing can be regarded as intermediate. Finally, at Da exceeding several tens, the spatial microphysical gradients remain until the final equilibrium stage is reached. In this case, the mixing can be regarded as inhomogeneous. At Da exceeding a few hundred the deviations from predictions based of the classical concept of extremely inhomogeneous become relatively small, which justifies attribute regarding this mixing as extremely inhomogeneous.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

Acknowledgements

This research was supported by the Israel Science Foundation (grant 1393/14), the Office of Science (BER), the US Department of Energy Award DE-SC0006788 and the Binational US-Israel Science Foundation (grant 2010446). Dr. Korolev's participation was supported by Environment Canada.

Appendix. List of symbols

Table A here

1206

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

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

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

1312

1313 **Table 2.** Comparison of analysis based on the classic concepts of mixing and the results of
1314 the present study

1315

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 effective radius remains constant	The effective radius varies only slightly (5-20 %) 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.

1333 **Tab. A. List of symbols**

1334 ("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	J kg^{-1}K^{-1}
$\mathscr{D}$	coefficient of water vapour diffusion in the air	m^2 s^{-1}
Da	the *Damkölher* number	nd
e	water vapor pressure	N m^{-2}
e_S	saturation vapour pressure above a flat water surface	N m^{-2}
F	$F=\frac{\rho_w L_w^2}{k_a R_v T^2}+\frac{\rho_w R_v T}{e_S(T)\mathscr{D}}$, coefficient	m^{-2} 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	J m^{-1}s^{-1}K^{-1}
K	turbulent diffusion coefficient	m^{2}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	J $kg^{-1}K^{-1}$
R_v	specific gas constant of water vapor	J $kg^{-1}K^{-1}$
S	$e/e_w - 1$, supersaruration 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	m^2s^{-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

Figures

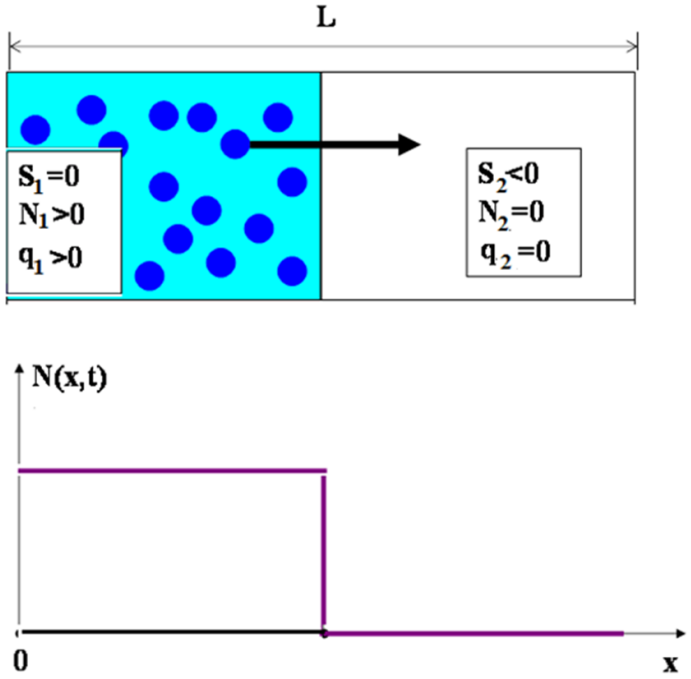

Fig.1. The schematic illustration of the 1D mixing problem considered in the study. The initial state at $t = 0$ is illustrated. The left volume of length $L / 2$ is a saturated cloudy volume; the right volume is a non-saturated air volume from the cloud environment.

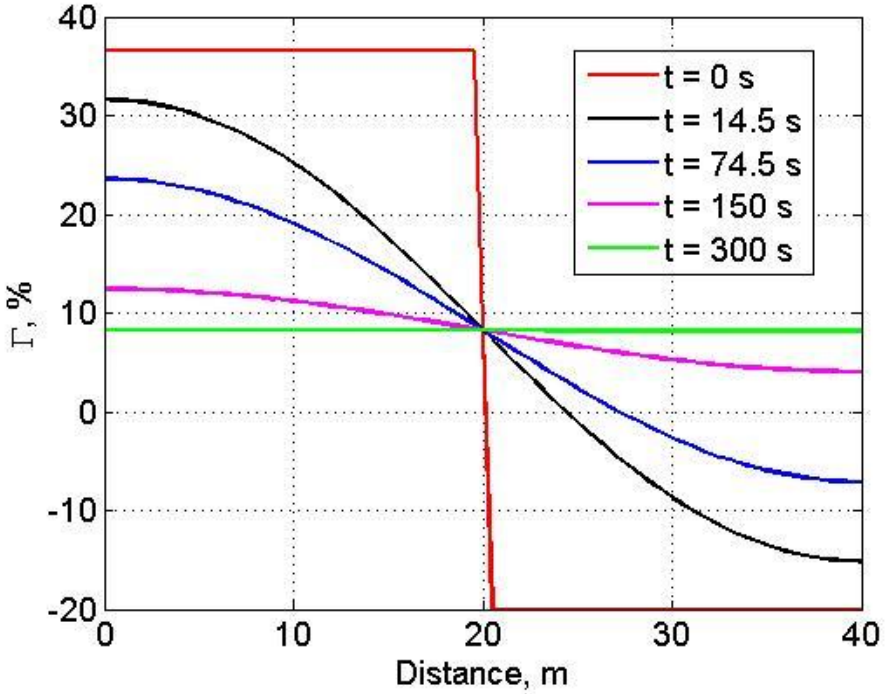

Fig. 2. An example of $\Gamma(x,t)$ evolution during mixing.

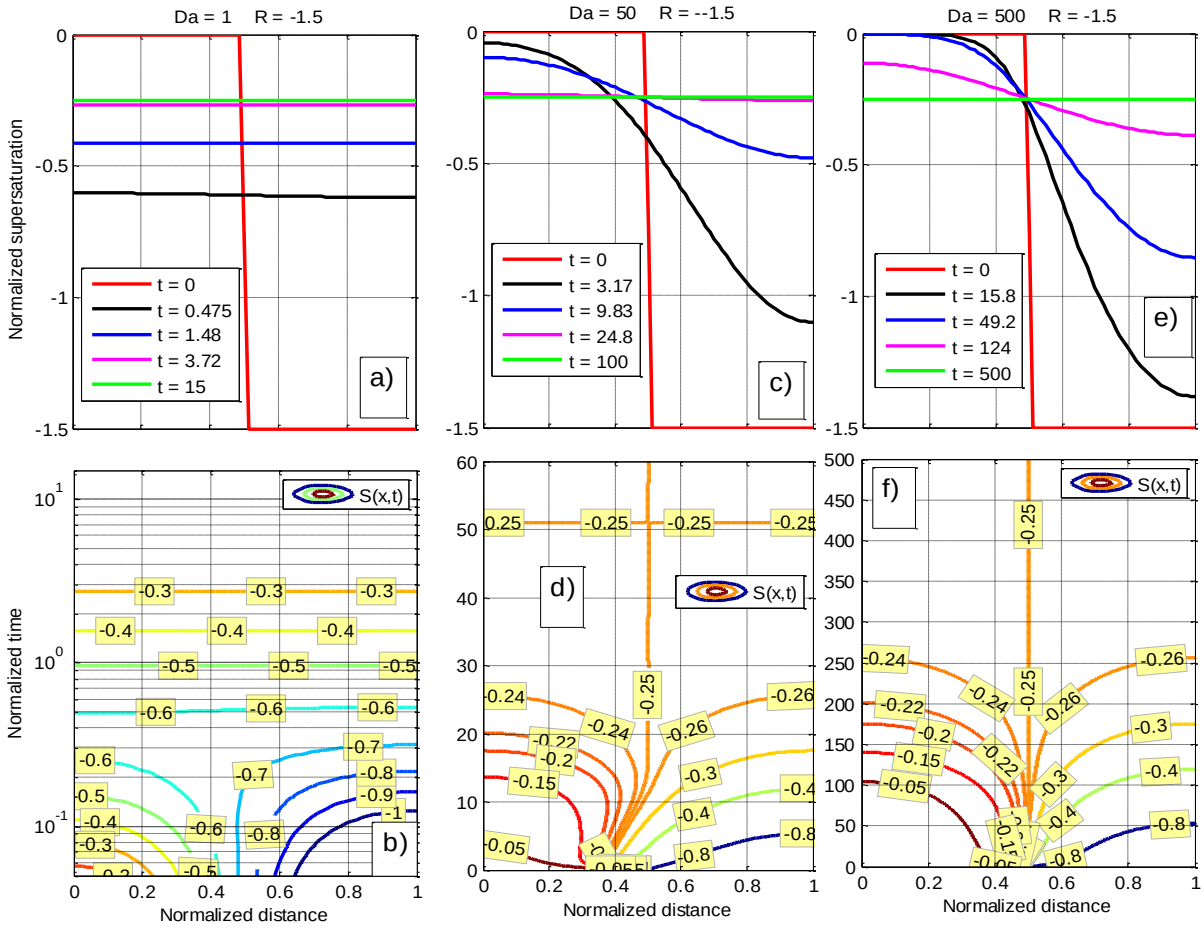

Fig. 3. Horizontal dependencies (upper row) and $\tilde{x}-\tilde{t}$ dependencies (lower row) of normalized supersaturation at $Da=1$, Da =50 and Da =500 and at $R=-1.5$. Panel b is plotted in semi-log scale.

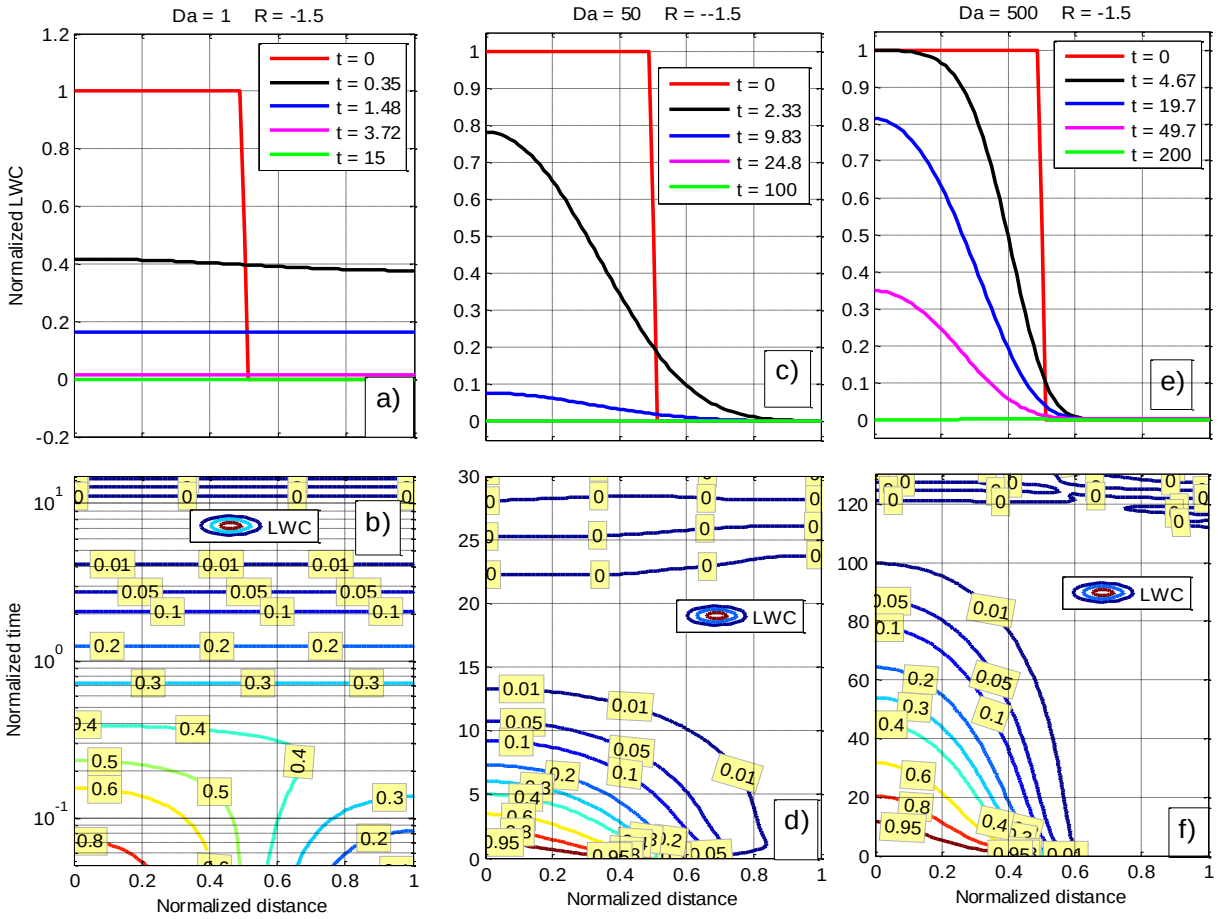

Fig. 4. The same as in Fig. 3, but for normalized LWC. Left bottom panel is plotted in semi-log scale.

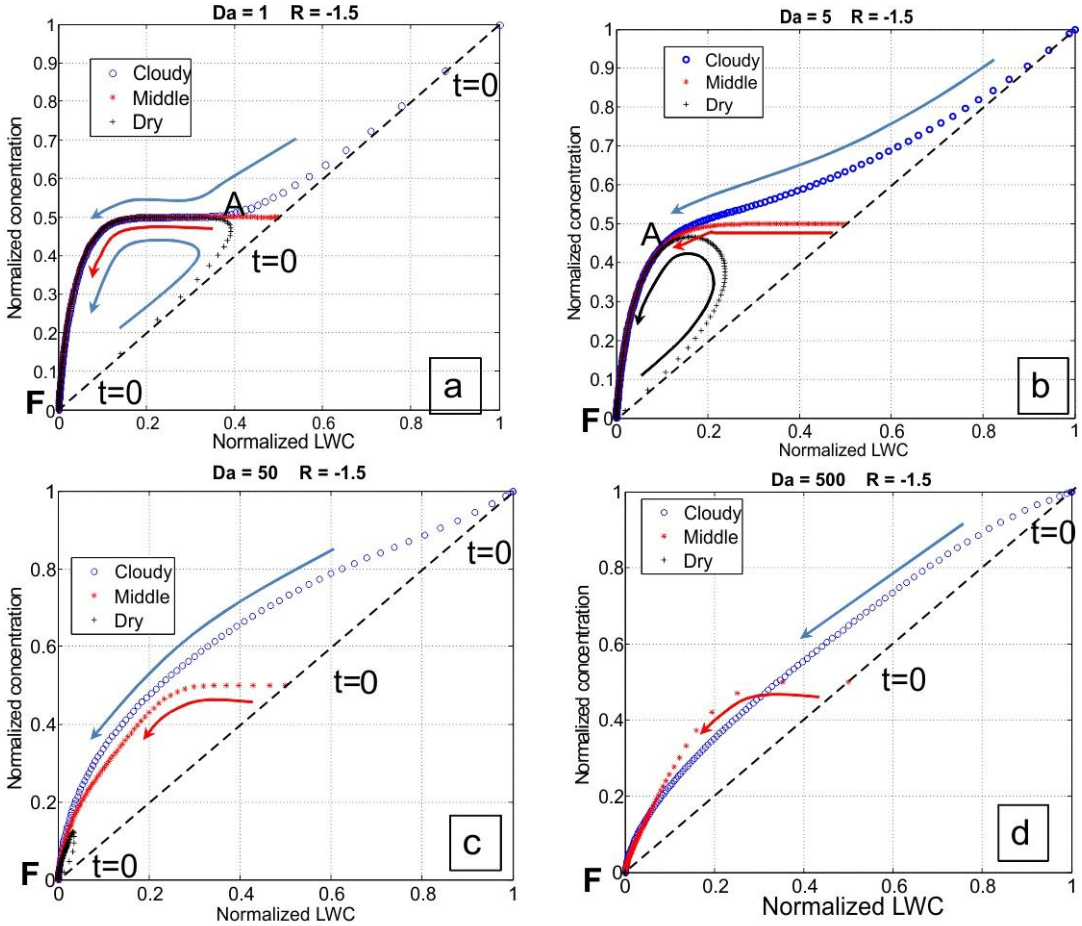

Fig. 5. Dependencies of normalized values of droplet concentration on normalized LWC at different Da and $R = -1.5$. Blue symbols mark the centre of the cloudy volume ($\tilde{x} = 1/4$), red symbols mark the interface between the cloudy volume and the dry volume ($\tilde{x} = 1/2$), and black crosses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

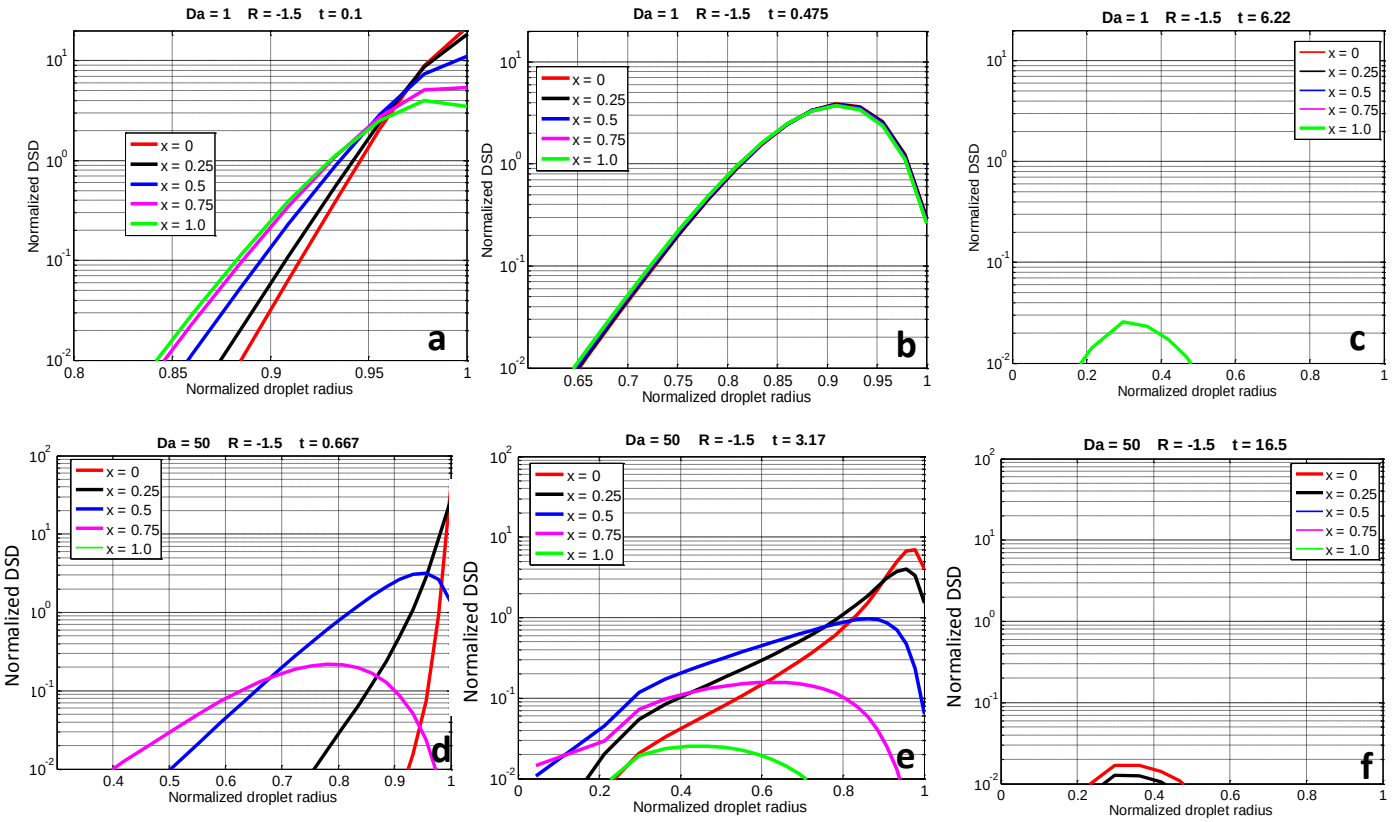

Fig. 6 Time evolution of DSD during droplet evaporation at $Da = 1$ (upper row) and $Da = 50$ (bottom row). In each panel, the normalized DSD are shown at different values of horizontal coordinate $\tilde{x}$. Different panels show DSD at different time instances.

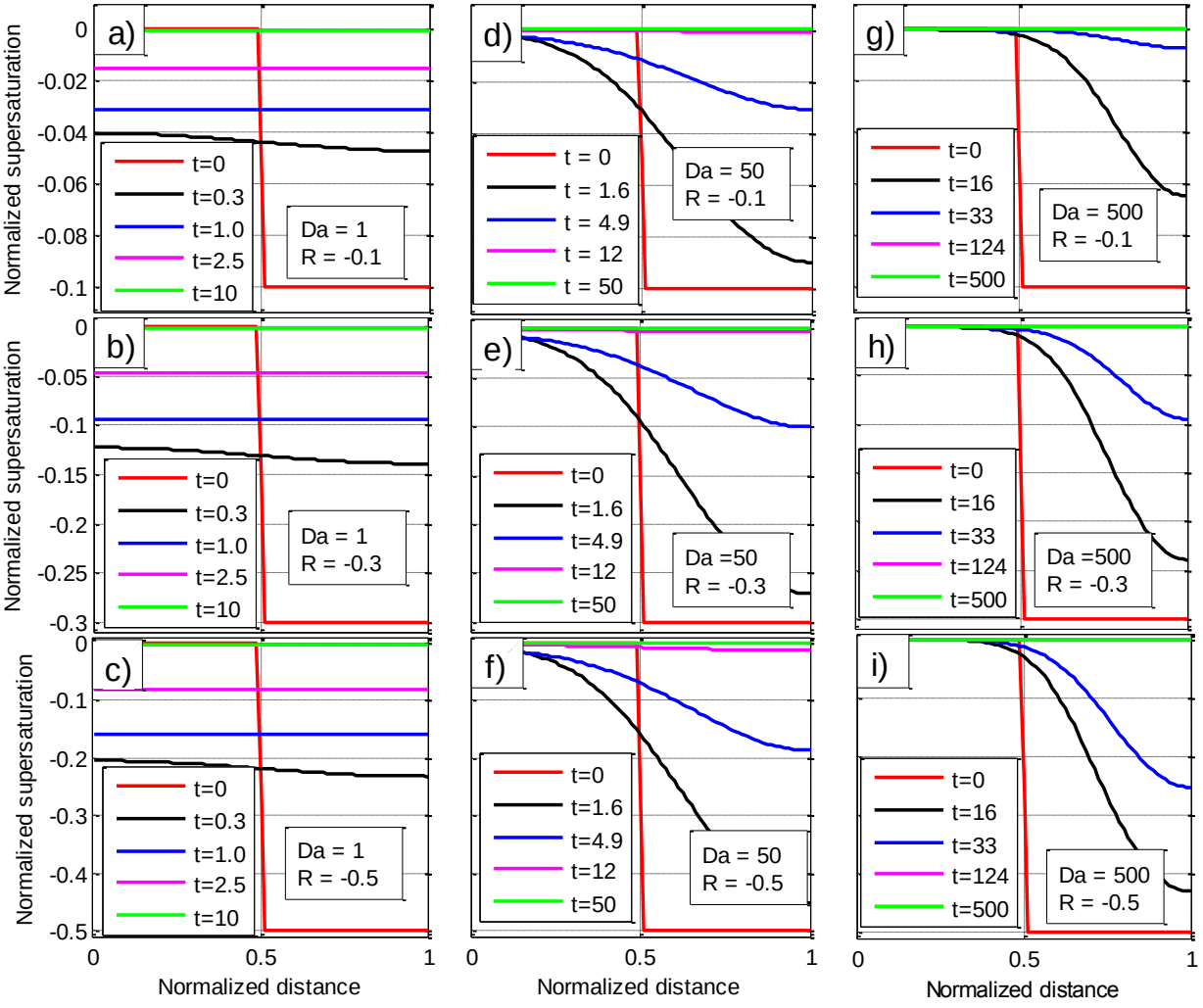

Fig. 7. Profiles of normalized supersaturation at different Da and different $R > -1$.

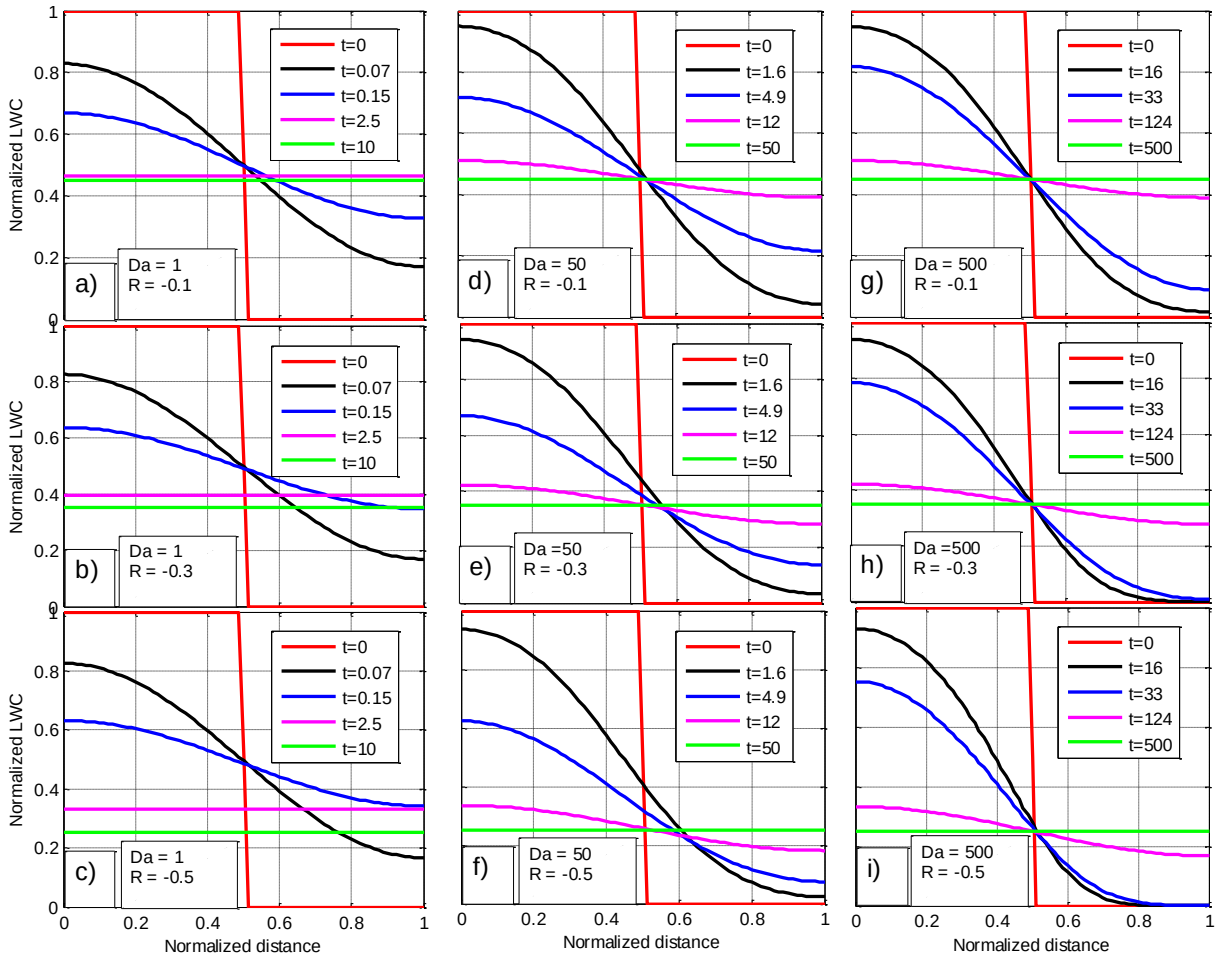

Fig. 8. Profiles of normalized LWC at different Da and at different $R > -1$.

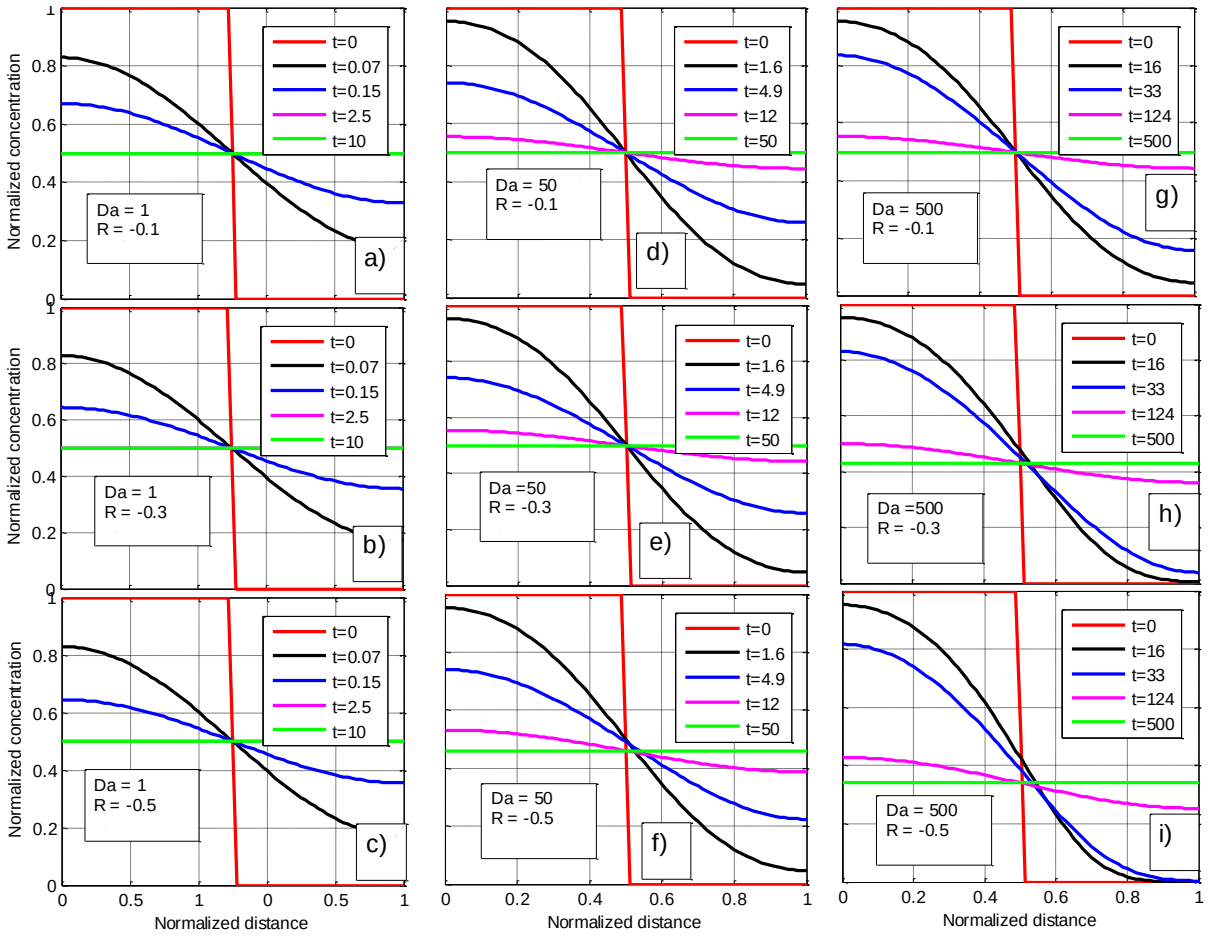

Fig. 9. Profiles of normalized droplet concentration at different Da and at different $R > -1$.

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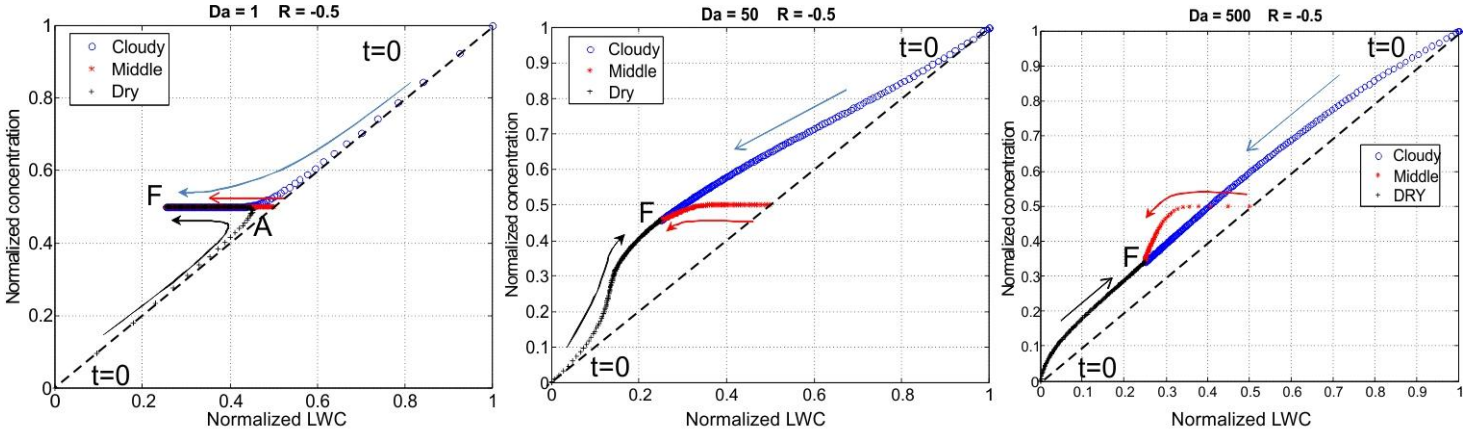

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1526 **Fig. 10**. Dependencies of normalized values of droplet concentration on normalized LWC
1527 at different *Da* and at $R = -0.5$. Blue circles mark the centre of the cloudy volume ($\tilde{x} = 1/4$),
1528 red symbols mark the initial interface ($\tilde{x} = 1/2$) and black crosses mark the centre of the
1529 initially dry volume ($\tilde{x} = 3/4$). Arrows show the direction of movement of the points with
1530 time. Point "F" marks the final stationary state of the system. The dashed line indicates the
1531 relationship between $\tilde{N}$ and $\tilde{q}$ in extremely inhomogeneous mixing (according to the classical
1532 concept).

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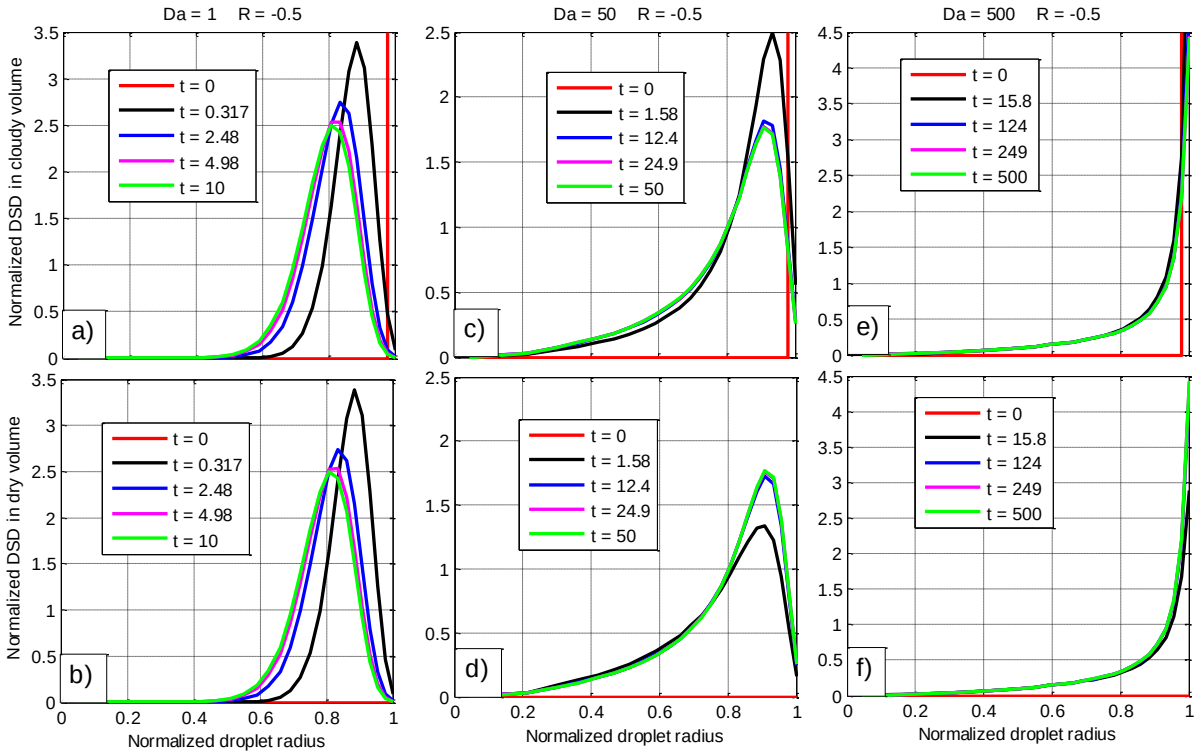


Fig. 11 Examples of DSD evolution in the initially cloudy volume ($\tilde{x}=1/4$) (upper row) and in the initially dry volume ($\tilde{x}=3/4$) (lower row) at $R=-0.5$ and at different values of Da.

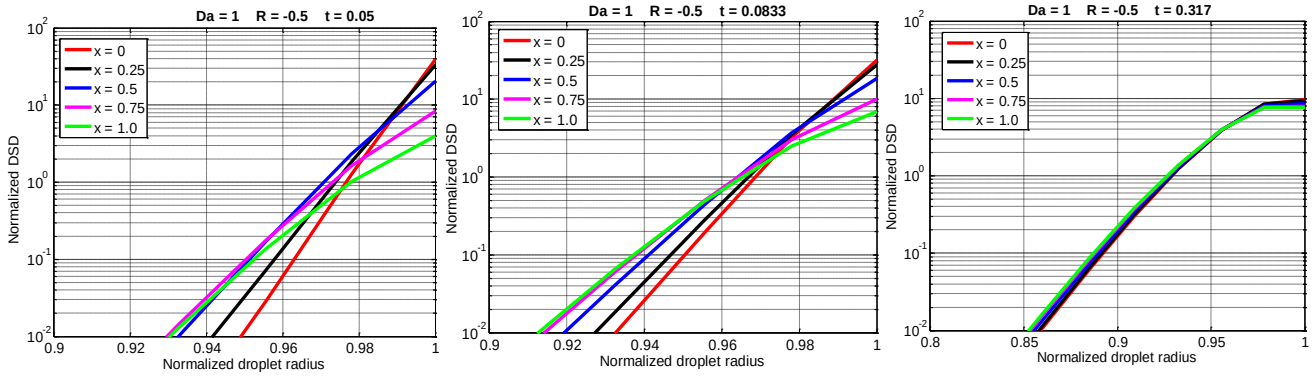


Fig. 12. DSD at different $\tilde{x}$ at the beginning of the mixing process for $Da = 1$ and $R = -0.5$.

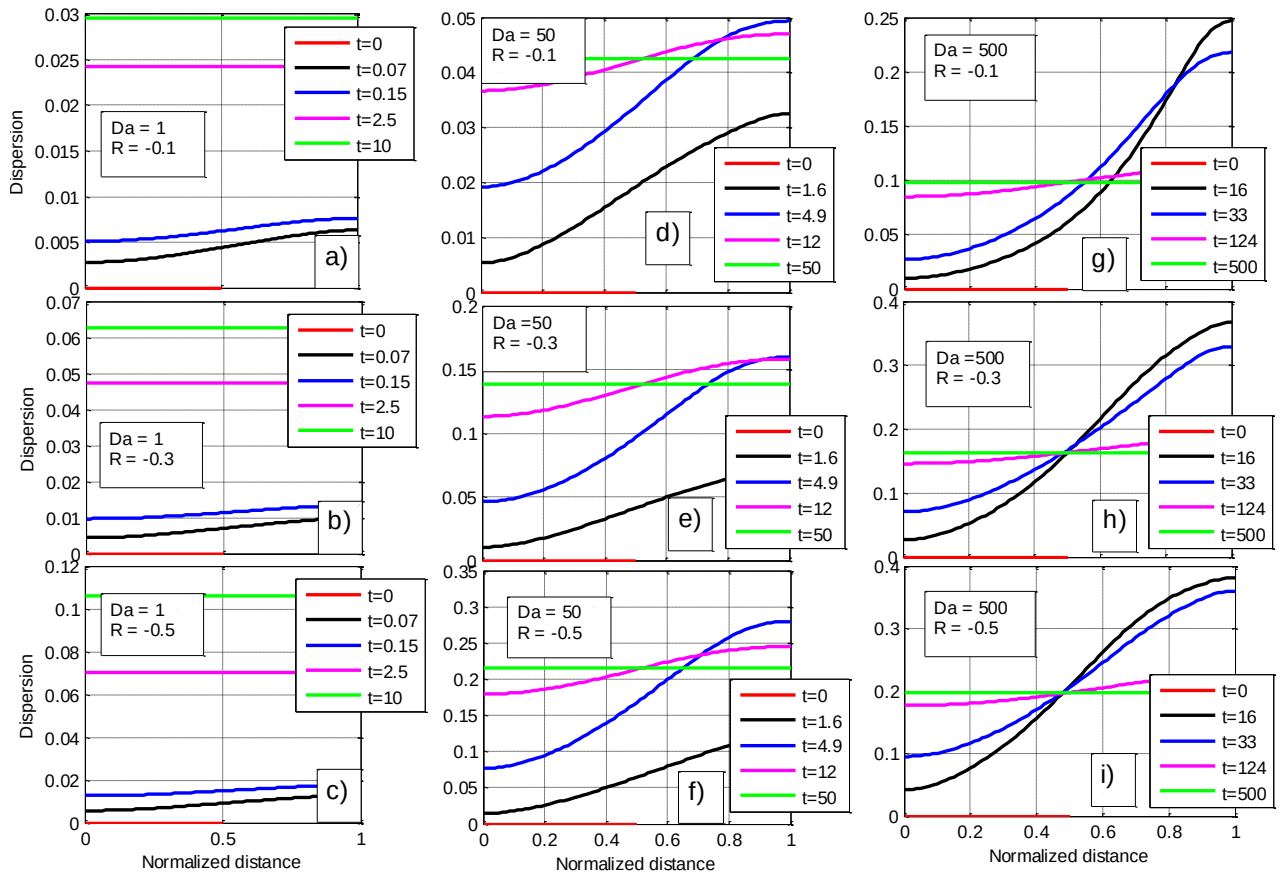


Fig. 13. Spatial dependencies of the relative DSD dispersion at different time instances and at different values of Da and different $R > -1$

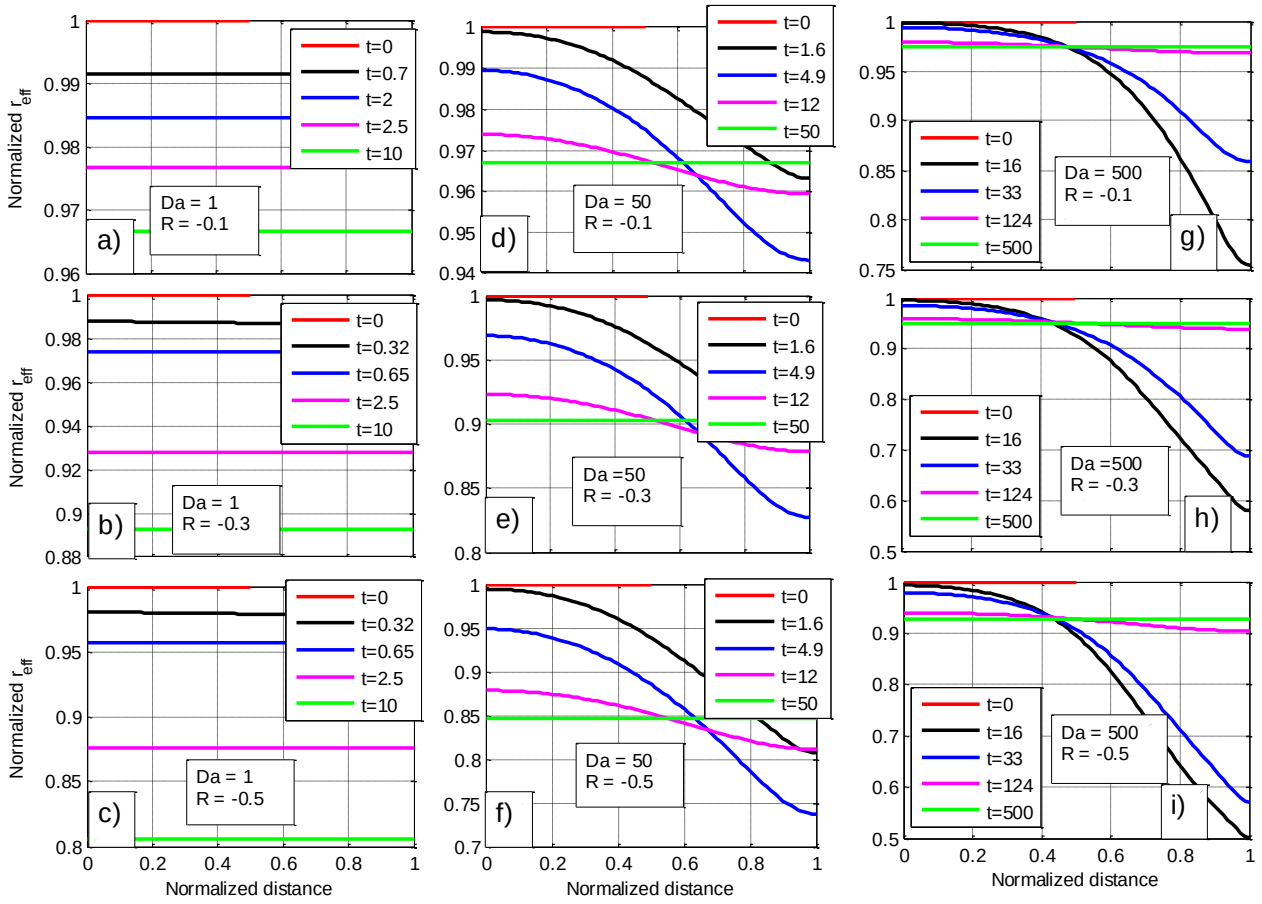


Fig. 14. Spatial dependencies of the effective radius at different time instances and at different values of Da and different $R > -1$

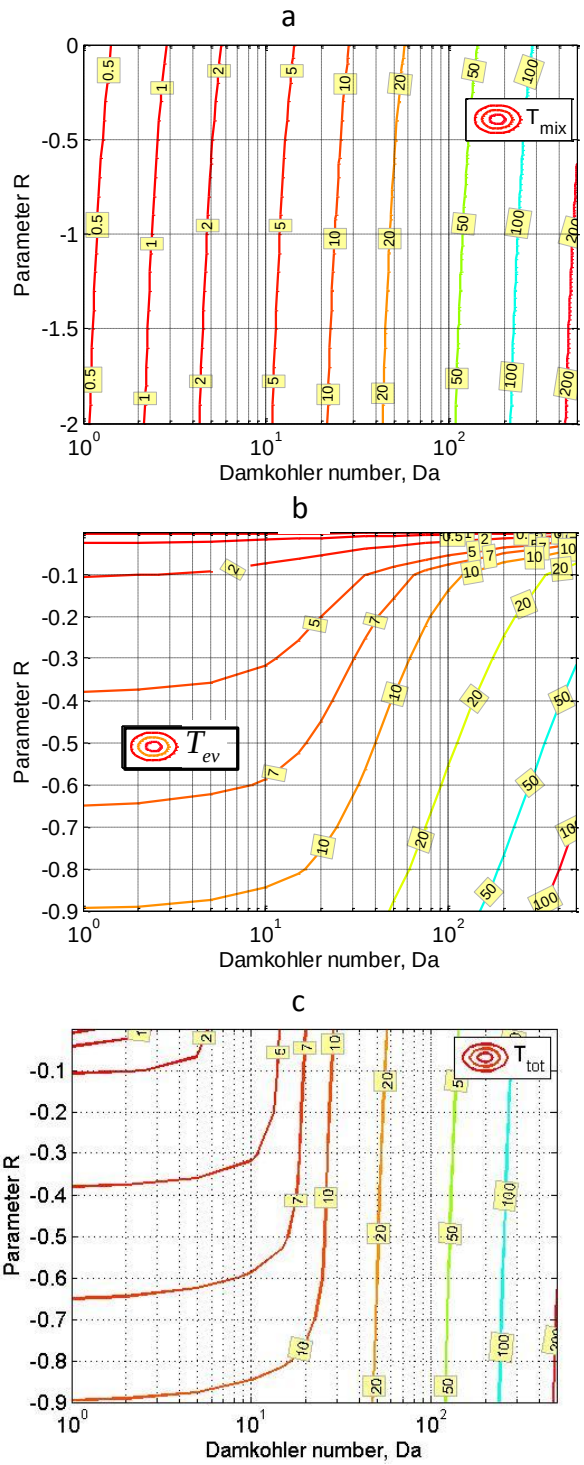
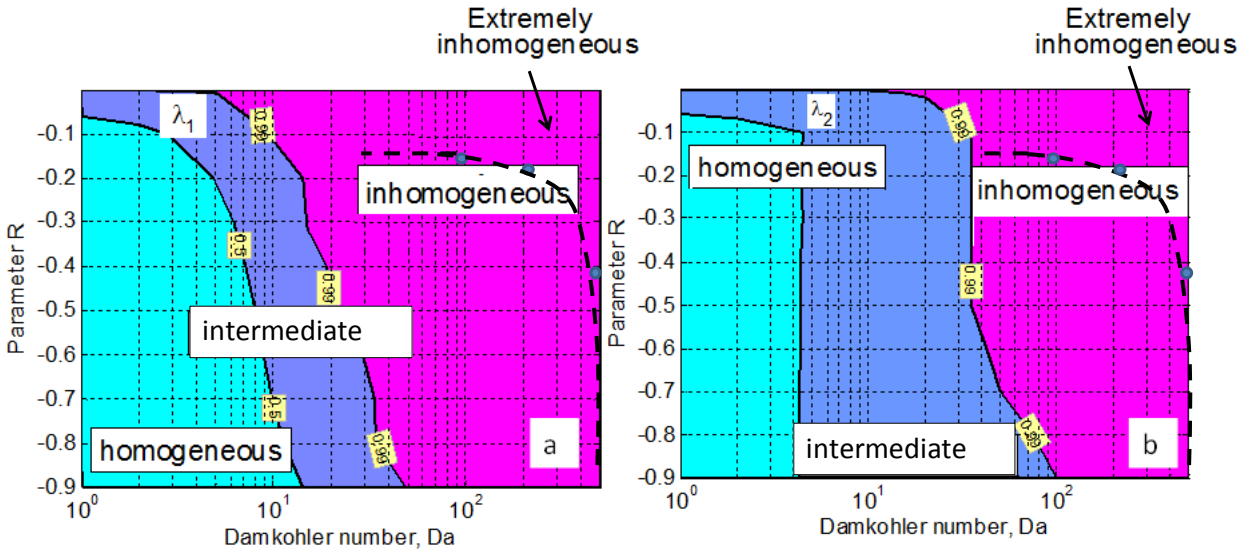
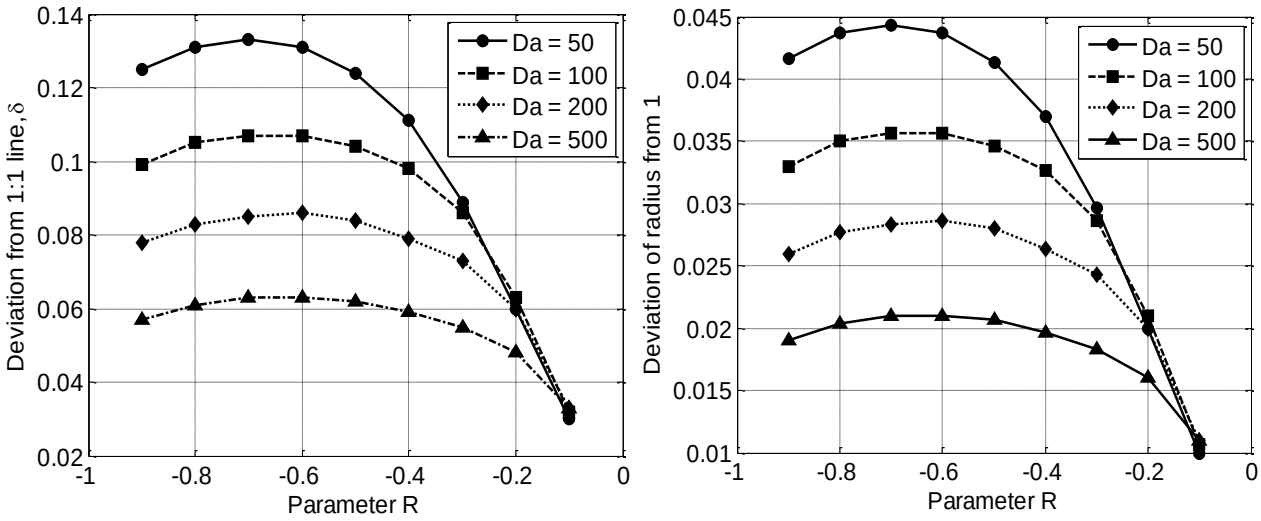


Fig. 15. Contours of normalized mixing duration times on $Da-R$ plane. (a) mixing time T_{mix}, (b) evaporation time T_{ev}, and (c) the total duration mixing time T_{tot}.



(a) **(b)**

Fig. 16. (a) The boundaries between mixing types on the $Da-R$ plane designed according to criteria $\lambda_1 = \frac{T_{mix}}{T_{tot}}$; **(b)** The boundaries between mixing types on the $Da-R$ plane designed according to criterion $\lambda_2 = \frac{2\langle \tilde{q}(T_{mix})\rangle - 1}{R}$ (Eq. 41). Dashed lines indicate the line corresponding to 2% deviation from the initial mean volume radius.



(a) (b)

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.